Iranian Journal of Organic Chemistry Vol. 6, No. 1 (2014) 1203-1209

A convenient and simple synthetic route to dihydropyrimido[4,5-*d*]pyrimidinetriones catalyzed by ZnO nanoparticle thin-film

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Received: December 2013; Revised: January 2014; Accepted: January 2014

Abstract: The zinc oxide nanoparticles thin-film (ZnO NPs thin-film) on glass substrate have been successfully prepared by the mild hydrothermal method and used as an efficient heterogeneous catalyst for the synthesis of dihydropyrimido[4,5-*d*]pyrimidinetrione derivatives *via* a three-component condensation of 6-aminouracils, aromatic aldehydes and urea in aqueous media at 70 ˚C. The reusability of the catalyst, use of water as a green solvent and easy isolation of the products along with high yields of products are the advantages of the present protocol.

Keywords: Aqueous media, Dihydropyrimido[4,5-*d*]pyrimidinetrione, Reusability of catalyst, ZnO Nanoparticls Thin-Film.

Introduction

Nanostructured metal oxides are a class of materials, which have been used as efficient heterogeneous catalysts in many various organic transformations [1-6]. Among the numerous metal oxide nanoparticles, ZnO NPs is a low-priced nano metal oxide which has attracted most interest due to its unique properties [7-13]. The use of conventional powder catalyst has disadvantages in stirring during the reaction and in separation of powder after the reaction. The preparation of film catalyst makes it possible to overcome these disadvantages and extend the industrial applications [14]. ZnO NPs thin-films are prepared by different techniques such as metal organic chemical vapor deposition, sol-gel, hydrothermal, thermal evaporation, oxidation and anodization [15-21].

On the other hand, pyrimidopyrimidine core structure belongs to an imprtant class of nitrogen-containing heterocyclic compounds, which are exhibited a wide spectrum of useful biological properties such as antitumour [22], antiviral [23], antioxidant [24], antibacterial [25], and hepatoprotective properties [26] activities. During the course of studies on the synthesis of substituted pyrimido[*d*]pyrimidine, a number of procedures have been reported [27-30].

Based on the above-mentioned importance of heterocycles having pyridopyrimidine scaffold, it is very interesting to research promising catalysts for the synthesis of these biologically interesting heterocyclic compounds. With this mind, the purpose of the present article was focused on the application of easily prepared ZnO NPs thin-film on glass substrate as an efficient heterogeneous catalyst for the synthesis of dihydropyrimido[4,5-*d*]pyrimidinetrione derivatives by a three-component condensation of 6-aminouracils, aromatic aldehydes and urea in aqueous media at 70 ˚C (Scheme **1**).

Results and discussion

We prepared first ZnO NPs thin-film by a facile hydrothermal solution method using zinc acetate dihydrate. In this process, the ZnO film synthesis includes three principal steps: (i) solution preparation;

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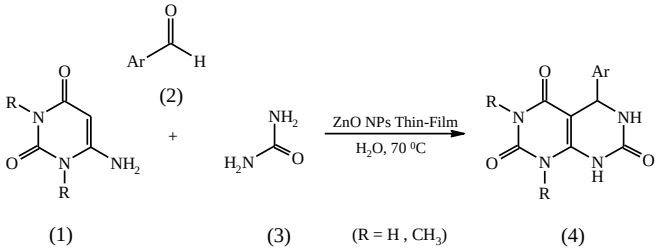
(ii) coating; and (iii) heat treatment. For the first step, the particle formation is discussed including nucleation and growth, particle size, morphology and colloids stability. These three steps involve several parameters such as: (i) nature and concentration of precursor; solvent and additive and solution aging time, for the chemical system; (ii) coating method, thickness and substrate for the coating step; and (iii) pre-and post-heat treatment for the last step.



Scheme 1: Synthesis of dihydropyrimido[4,5-*d*]pyrimidinetriones.

Figure **1a** shows the FT-IR spectrum of as-synthesized zinc acetate film, the absorption band at 1582 cm^{-1} is due to C=O arising from the bridging type metal-acetate bonding (M--OCOO-M), those at 1415 cm^{-1} is the C-O stretching frequencies in zinc acetate and at 1340 cm^{-1} is weakly C-O mode in acetic acid molecule (CH_3-COOH). Figure **1b** shows the FT-IR spectrum of zinc oxide film annealed at 400°C which shows an absence of absorption bands corresponding to organics and hydroxyls indicating complete removal of organics.

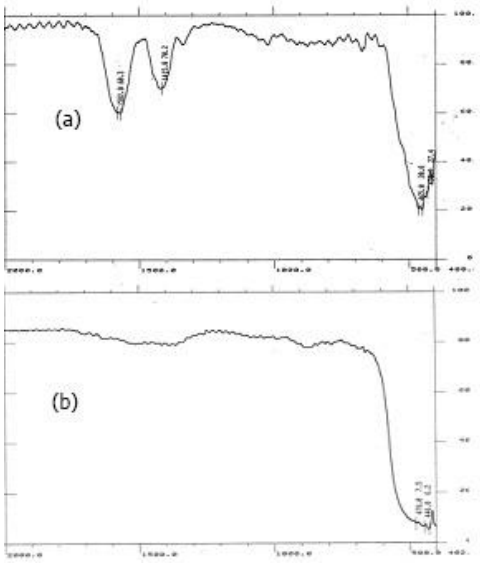


Figure 1: FT-IR spectra of (a) as synthesized zinc acetate film (b) ZnO NPs thin-film annealed at 400 ˚C.

X-ray diffraction (XRD) analyses of ZnO NPs thin-films annealed at 400 ˚C are depicted in Figure **2**. The spectra shows well defined diffraction peaks showing good crystallinity. One can see that the ZnO NPs thin-film exhibit the peaks at 2θ angle 31.9, 34.6 and 36.2˚ correspond to diffraction from planes (002), (101) and (110) respectively of hexagonal wurtzite ZnO (JCPDS:36-1451).

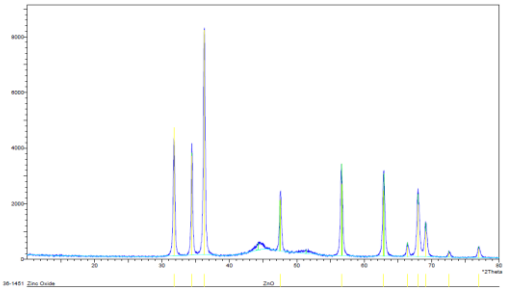


Figure 2: X-ray diffraction analyses of ZnO NPs thin-film annealed at 400 ˚C.

The optical absorption spectra of the thin-film in the UV–Vis wavelength range are presented in Figure **3**. It can be seen that the film has high transparency in the visible range. The absorption at higher wavelengths in the visible region is low at wavelength around 484 nm. Further, a sharp increase of absorption occurs about 200-400 nm indicating the ZnO absorption. With respect to the bulk absorption edge appearing at 373 nm at room temperature, a stronger absorption feature at about 361 nm is blue shifted about 12 nm at around 361 nm.

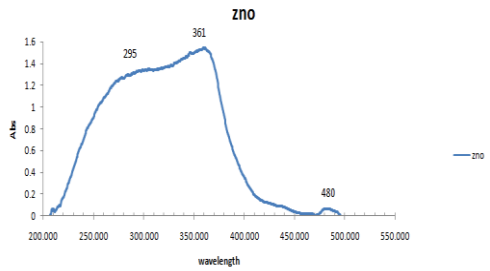


Figure 3: The optical absorption spectra of ZnO NPs thin-film.

Figure **4** shows the surface morphology of ZnO in thin-film. In general, film is homogeneous and continuous. Separate coating layers are not visible in sintered film. The random distribution of grains, suggests a random nucleation mechanism. Film thickness was found to be in the range of 366 nm.

6-Aminouracil, 4-bromobenzaldehyde and urea were first used as model substrates to study the catalytic activity of prepared ZnO NPs thin-film. To

demonstrate the efficiency of the catalyst a blank reaction was carried out under refluxing in aqueous media and in the absence of catalyst. After 8 h stirring, work-up and recrystallization from DMF/H_2O, only 32% of product was obtained (Table **1**, entry 1). Compared with various solvents, using H_2O as reaction media gave us the best result (Table **1**, entries 2-5). It is worth noting that, with decreasing the temperature to 60 ˚C the yield of product dropped mainly, meanwhile, an increase of the temperature from 70 ˚C to 90 ˚C, showed no measurable effects on the yield (Table **1**, entries 2 and 6-8).

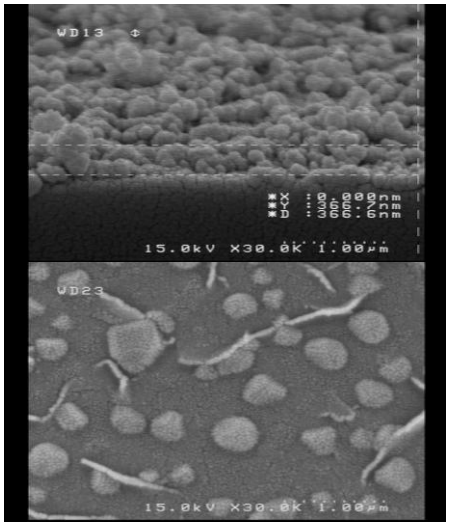


Figure 4: SEM image of ZnO NPs thin-film.

To examine the scope of presented method, we studied some substituted aromatic aldehydes with 6-aminouracil or 1,3-dimethyl-6-aminouracil to produce the corresponding products in high to excellent yields. The results are summarized in Table **2**. The structures of products (**4a-j**) were confirmed by IR, ^{1}H NMR spectral data and by elemental analyses.

A plausible mechanism for the formation of the product would be as follows: the ZnO facilate Knoevenagel condensation of aromatic aldehyde and 6-aminouracil for the formation of alkene, through Lewis acid sites (Zn^{2+}) which are coordinated to the oxygen of the aldehyde and active it for nucleophilic attack of 6-aminouracil. ZnO can also deprotonate urea which occurs in the presence of Lewis basic sites (O^{2-}), and acitive it for adding to alkene *via* Michael type addition. Intramolecular cyclization of Michael adduct gives product, after elimination of ammonia.

In view of environmental friendly methodologies, the recyclability of the catalyst is an important factor. For this reason, the reusability of the ZnO NPs thin-film was examined in model reaction under the optimized reaction conditions by separating film from the reaction mixture washing with DMF, and drying in vacuo. It was found that the ZnO film could be recycled for at least 7 cycles without significant change in activity (Figure **5**).

Conclusion

We have synthesised ZnO Nanoparticle thin-film by a facile hydrothermal process from zinc acetate, on glass substrate. The ZnO NPs thin-film was found to catalyze the three-component reaction of 6-aminouracils, aromatic aldehydes and urea efficiently, affording dihydropyrimido[4,5-*d*]pyrimidinetrione derivatives in high to excellent yields after an easy work-up. This catalyst is readily recovered after completion of the reaction and can be reused several times without deactivation. Finally, this general and simple procedure provides opportunities for practical application of ZnO NPs thin-film as an efficient heterogeneous catalyst.

Experimental

Materials and Methods:

All chemicals used in this work purchased from *Mreck* and *Fluka* in high purity. Melting points were determined with *Electrothermal 9100* melting point apparatus and were uncorrected. FT-IR spectra were obtained using a *Bruker, Equinox 55, Golden Gate Micro-ATR* spectrometer. ^{1}H NMR spectra were run on a *Bruker DRX-500 AVANCE* at 500 MHz using TMS as internal standard and DMSO-d_6 as solvent. Elemental analyses were carried out using a *Heraeus CHN-O-Rapid* analyzer. Microscopic morphology of catalyst was visualized by scanning electron microcopy *SEM*: *Philips XL30*. Power X-ray diffraction was recorded on a *Philips, X'-Pert* diffractometer using CuKα radiation (λ =1.54060Å) in the range 2θ = 5–80˚). UV-Vis spectrophotometery was carried out by *Aventes UV4200 Avaspec* spectrometer with drift accessory in the wavelength range from 200 to 500 nm.

General Procedure for the Preparation of ZnO NPs thin-film Catalyst:

For the preparation of the ZnO film, 1.6 g of zinc acetate dihydrate (99.98%) was dissolved into 50 ml of methanol, (>99.9%) and stirred on a magnetic stirrer for 10 min. Then, a small amount (0.2 ml) of glacial acetic acid (>99.9%) was added to the mixture and stirred at 40 ˚C for 10 min, the obtained transparent solution was transferred to an autoclave. The glass

substrate (15×5 mm) was precleaned with water and detergent, and then cleaned in methanol and acetone for 10 min each by using an ultrasonic cleaner and then cleaned with deionized water and dried. The hydrothermal technique was used for the deposition of the sample on the surface of glass substrate. The cleaned glass substrate was transferred to autoclave and aged at 60 ˚C for 48 h, and then the coated film was dried at 100 ˚C in air for 30 min. The process was repeated for second time to prepare film with desired thickness. Finally, the ZnO NPs thin-film was heat treated at 400 ˚C in a furnace.

Table 1: Synthesis of 5-(4-bromophenyl)-5,8-dihydropyrimido[4,5-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (4b) under different conditions.

Entry	Solvent	Catalyst (mol%)	Temp. (˚C)	Time (h)	Yield (%)[a]
1	H_2O	No catalyst	90	8	32
2	H_2O	ZnO NPs thin-film	70	0.5	94
3	EtOH	ZnO NPs thin-film	70	0.5	84
4	CH_2Cl_2	ZnO NPs thin-film	80	0.5	63
5	DMF	ZnO NPs thin-film	100	0.5	69
6	H_2O	ZnO NPs thin-film	60	0.5	62
7	H_2O	ZnO NPs thin-film	80	0.5	94
8	H_2O	ZnO NPs thin-film	90	0.5	96

[a]Isolated yield.

Table 2: ZnO NPs thin-film catalyzed three-component synthesis of dihydropyrimido[4,5-*d*]pyrimidinetriones 4a-4j.

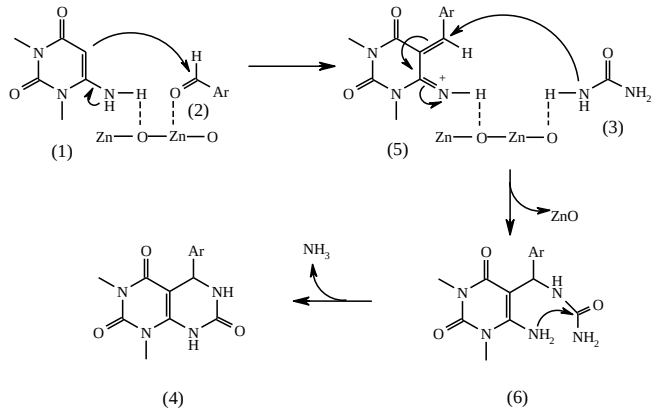
Product	Ar	R	Yield (%)[a,b]	M.P. (˚C)
4a	C_6H_5	H	97	245-247
4b	4-Br-C_6H_4	H	94	211-212
4c	4-Cl-C_6H_4	H	97	294-296
4d	4-OCH_3-C_6H_4	H	98	285-287
4e	4-CH_3-C_6H_4	H	98	249-251
4f	C_6H_5	CH_3	94	>300 (dec.)
4g	4-Br-C_6H_4	CH_3	93	>300 (dec.)
4h	4-Cl-C_6H_4	CH_3	93	>300 (dec.)
4i	4-OCH_3-C_6H_4	CH_3	89	>300 (dec.)
4j	4-CH_3-C_6H_4	CH_3	90	>300 (dec.)

[a]Yields refer to those of pure isolated products characterized by IR, ^{1}H NMR spectroscopy and elemental analysis.
[b]In all cases, reaction time was 0.5 h stirring at 70 ˚C.

General Procedure for the Synthesis of Compounds 4a-j:

A mixture of 6-aminouracil (2 mmol), aromatic aldehyde (2 mmol), urea (2.5 mmol) and water (20

mL) were taken in 50 mL round bottom flask. The ZnO NPs thin-film was then dipped into the sample and the mixture was stirred at 70 ˚C for about 0.5 h. The progress of the reaction was checked by TLC. After completion of the reaction, the ZnO thin-film was removed from the reaction mixture, for reuse by washing with DMF, and drying at 100 ˚C for several hours in a vacuum. The reaction mixture was then filtered. The residue was recrystallized from DMF and H_2O to afford the pure product.



Scheme 2: Possible mechanism for the formation of dihydropyrimido[4,5-*d*]pyrimidinetrione derivatives.

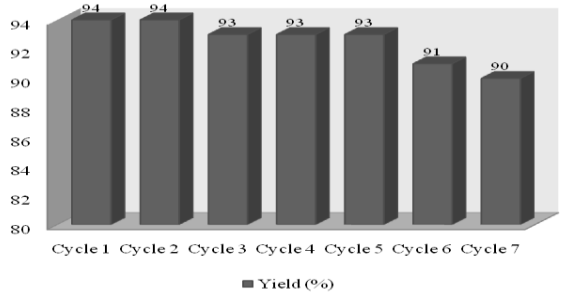


Figure 5: Recycling of ZnO NPs thin-film.

Selected spectral data:

5-Phenyl-5,8-dihydropyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4a):

Yield: 250 mg (97%). White powder. mp 245-247 ˚C (lit. 247-250 ˚C [28]). IR (KBr) *v* 3403, 3313, 3175, 1707, 1643, 1596 cm^{-1}. ^1H NMR (500 MHz, DMSO) *δ* 5.49 (s, 1H, CH), 7.16 (m, 5H, ArH), 7.55 (s, 1H, NH), 10.15 (s, 1H, NH), 11.43 (s, 2H, NH) ppm. Anal. for $C_{12}H_{10}N_4O_3$ (258.24): calcd. C 55.81, H 3.90, N 21.69; found: C 55.69, H 3.67, N 21.55%.

5-(4-Bromophenyl)-5,8-dihydropyrimido[4,5-d] pyrimidine-2,4,7(1H,3H,6H)-trione (4b):

Yield: 317 mg (94%). Pale yellow powder. mp 211-212 ˚C (lit. 210-212 ˚C [28]). IR (KBr) *v* 3376, 3262, 3039, 1699, 1620, 1515 cm^{-1}. ^1H NMR (500 MHz, DMSO) *δ* 5.60 (s, 1H, CH), 7.32 (m, 4H, ArH), 8.59 (s, 1H, NH), 10.02 (s, 1H, NH), 11.30 (s, 2H, NH) ppm. Anal. for $C_{12}H_9BrN_4O_3$ (337.13): calcd. C 42.75, H 2.69, N 16.62; found: C 42.59, H 2.75, N 16.44%.

5-(4-Chlorophenyl)-5,8-dihydropyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4c):

Yield: 284 mg (97%). White powder. mp 294-296 ˚C (lit. 294-295 ˚C [28]). IR (KBr) *v* 3392, 3250, 3074, 1716, 1637, 1554 cm^{-1}. ^1H NMR (500 MHz, DMSO) *δ* 5.21 (s, 1H, CH), 7.33 (m, 4H, ArH), 7.72 (s, 1H, NH), 8.55 (s, 1H, NH), 11.13 (s, 2H, NH) ppm. Anal. for $C_{12}H_9ClN_4O_3$ (292.68): calcd. C 49.25, H 3.10, N 19.14; found: C 49.12, H 3.26, N 19.05%.

5-(4-Methoxyphenyl)-5,8-dihydropyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4d):

Yield: 282 mg (98%). White powder. mp 258-287 ˚C (lit. 285-287 ˚C [28]). IR (KBr) *v* 3405, 3338, 3212, 1683, 1635, 1548 cm^{-1}. ^1H NMR (500 MHz, DMSO) *δ* 3.77 (s, 3H, OCH_3), 5.18 (s, 1H, CH), 6.95 (m, 4H, ArH), 8.30 (s, 1H, NH), 8.75 (s, 1H, NH), 11.12 (s, 1H, NH), 11.37 (s, 1H, NH) ppm. Anal. for $C_{13}H_{12}N_4O_4$ (288.26): calcd. C 54.17, H 4.20, N 19.44; found: C 54.08, H 4.06, N 19.53 %.

5-(4-Methylphenyl)-5,8-dihydropyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4e):

Yield: 267 mg (98%). White powder. mp 249-251 ˚C (lit. 248-250 ˚C [28]). IR (KBr) *v* 3387, 3295, 3086, 1698, 1606, 1584 cm^{-1}. ^1H NMR (500 MHz, DMSO) *δ* 2.87 (s, 3H, CH_3), 5.89 (s, 1H, CH), 7.07 (m, 4H, ArH), 7.93 (s, 1H, NH), 10.02 (s, 1H, NH), 10.93 (s, 2H, NH) ppm. Anal. for $C_{13}H_{12}N_4O_3$ (279.26): calcd. C 55.91, H 4.33, N 20.06; found: C 56.09, H 4.49, N 20.14%.

1,3-Dimethyl-5-phenyl-5,8-dihydropyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4f):

White powder. Yield: 269 mg (94%). mp >300 ˚C dec. (lit. 324 ˚C dec. [29]). IR (KBr) *v* 3328, 3115, 1698, 1643, 1585 cm^{-1}. ^1H NMR (500 MHz, DMSO) *δ* 3.09 (s, 3H, CH_3), 3.34 (s, 3H, CH_3), 5.22 (s, 1H, CH), 7.32 (m, 5H, ArH), 8.09 (s, 1H, NH), 9.81 (s, 1H, NH) ppm. Anal. for $C_{14}H_{14}N_4O_3$ (286.29): calcd. C 58.74, H 4.93, N 19.57; found: C 58.83, H 5.12, N 19.41%.

1,3-Dimethyl-5-(4-bromophenyl)-5,8-dihydro pyrimido [4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4g):

Yield: 340 mg (93%). Yellow powder. mp >300 ˚C dec. (lit. 307 ˚C dec. [29]). IR (KBr) *v* 3323, 3088, 1712, 1644, 1600 cm^{-1}. ^1H NMR (500 MHz, DMSO) δ 3.07 (s, 3H, CH_3), 3.34 (s, 3H, CH_3), 5.20 (s, 1H, CH), 7.41 (m, 4H, ArH), 8.10 (s, 1H, NH), 9.88 (s, 1H, NH) ppm. Anal. for $C_{14}H_{13}BrN_4O_3$ (365.19): calcd. C 46.05, H 3.59, N 15.34; found: C 45.94, H 3.45, N 15.27%.

1,3-Dimethyl-5-(4-chlorophenyl)-5,8-dihydro pyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4h):

Yield: 298 mg (93%). White powder. mp >300 ˚C dec. (lit. 312 ˚C dec. [29]). IR (KBr) *v* 3332, 3126, 1691, 1654, 1604 cm^{-1}. ^1H NMR (500 MHz, DMSO) δ 3.08 (s, 3H, CH_3), 3.35 (s, 3H, CH_3), 5.20 (s, 1H, CH), 7.35 (m, 4H, ArH), 8.10 (s, 1H, NH), 9.85 (s, 1H, NH) ppm. Anal. for $C_{14}H_{13}ClN_4O_3$ (320.73): calcd. C 52.43, H 4.09, N 17.47; found: C 52.32, H 3.99, N 17.39%.

1,3-Dimethyl-5-(4-methoxyphenyl)-5,8-dihydro pyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4i):

Yield: 282 mg (89%). Pale yellow powder. mp >300 ˚C dec. (lit. 307 ˚C dec. [29]). IR (KBr) *v* 3352, 3095, 1681, 1646, 1598 cm^{-1}. ^1H NMR (500 MHz, DMSO) δ 3.08 (s, 3H, CH_3), 3.37 (s, 3H, CH_3), 3.70 (s, 3H, OCH_3), 5.18 (s, 1H, CH), 7.10 (m, 4H, ArH), 8.03 (s, 1H, NH), 9.80 (s, 1H, NH) ppm. Anal. for $C_{15}H_{16}N_4O_4$ (316.31): calcd. C 56.96, H 5.10, N 17.71; found: C 56.86, H 5.17, N 17.84 %.

1,3-Dimethyl-5-(4-methylphenyl)-5,8-dihydro pyrimido[4,5-d]pyrimidine-2,4,7(1H,3H,6H)-trione (4j):

Yield: 270 mg (90%). Pale yellow powder. mp >300 ˚C (dec.) (lit. 315 ˚C dec. [29]). IR (KBr) *v* 3310, 3115, 1687, 1656, 1601 cm^{-1}. ^1H NMR (500 MHz, DMSO) δ 2.25 (s, 3H, CH_3), 3.10 (s, 3H, NCH_3), 3.35 (s, 3H, NCH_3), 5.19 (s, 1H, CH), 7.16 (m, 4H, ArH), 8.04 (s, 1H, NH), 9.81 (s, 1H, NH) ppm. Anal. for $C_{15}H_{16}N_4O_3$ (300.32): calcd. C 59.99, H 5.37, N 18.66; found: C 56.11, H 5.49, N 18.44%.

Acknowledgments

Partial support of this work by East Tehran Branch, Islamic Azad University is gratefully acknowledged.

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