Microwave-assisted Oxidation of 3,4-Dihydropyrimidin-2(*1H*)-ones

Hamid Reza Memarian, Hassan Sabzyan, and Asadollah Farhadi

Department of Chemistry, University of Isfahan, Isfahan 81746-73441, I. R. Iran

Reprint requests to Prof. Hamid R. Memarian. Fax: +98-311-6689732.
E-mail: memarian@sci.ui.ac.ir

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A new facile cost-effective method for the oxidation of acetyl and carboethoxy derivatives of 3,4-dihydropyrimidin-2(1*H*)-ones (DHPMs) by using potassium peroxydisulfate in water as solvent under microwave irradiation has been developed. Whereas the presence of the acetyl group instead of the carboethoxy group in position 5 decreases the rate of oxidation, the nature of the additional substituent (electron-donating or electron-withdrawing group) and its location on the phenyl ring (*ortho*-, *meta*- or *para*-position) attached to C-4 of the dihydropyrimidinone ring influence the rate of reaction. Increase in the rate of the reaction compared with that of the reaction under reflux condition is the advantage of the use of microwave irradiation for this oxidation method.

Key words: Dihydropyrimidinone, Green Solvent, Microwave Irradiation,
Potassium Peroxydisulfate, Pyrimidinone

Introduction

Application of microwave irradiation in optimization and acceleration of organic reactions has been rapidly increased in recent years. Microwave technique as an efficient heating source for organic reactions was first recognized in the mid 1980's [1] and is highly recommended over other thermal reactions for organic synthesis. Many reactions that typically require high temperatures and long reaction times have been accelerated using microwave irradiation with especially high yield and clean reaction conditions [2 – 4].

Nitrogen heterocyclic compounds, such as pyrimidines, are of synthetic interest because they constitute an important class of natural and synthetic products. Some pyrimidine derivatives are important drugs showing inhibition of the HIV virus [5].

The course of the dehydrogenation of 3,4-dihydropyrimidin-2(1*H*)-ones (DHPMs) as a route to pyrimidinones depends on the effects of the nature of the substituents located on the ring positions 4 and 5 on the rate of reaction. Many mild and powerful reagents such as MnO_2 [6], $FeCl_3$ [7], $RuCl_3/O_2$ in AcOH [8], pyridinium chlorochromate (PCC) [6], chloranil [6], $KMnO_4$/clay [6], 2,3-dichloro-5,6-dicyano-*p*-benzoquinone (DDQ) [6], $NaNO_2$/AcOH [6], ammonium ceric(VI) nitrate (CAN) in AcOH [9], the combination $Co(NO_3)_2 \cdot 6H_2O/K_2S_2O_8$ [10], Pd/C [6], Br_2 [11], and sulfur [12] were used as oxidants for DHPMs. However, for all of these reagents, the reaction was not efficient, either because of the hard reaction conditions required for the oxidation reaction to proceed, or in some cases, the occurrence of side reactions.

The peroxydisulfate ion is known as one of the strongest oxidizing agents in aqueous and protic organic solvents with a redox potential of about -2.01 V [13, 14]. The mechanism of the thermal decomposition of peroxydisulfate is believed to produce the sulfate radical ions which can abstract hydrogen from water to give hydroxyl radicals [15], which then facilitate the dehydrogenation (oxidation) of the compound. The cleavage of the O–O bond in the $S_2O_8{}^{2-}$ anion has a relatively high activation energy of around 130 kJ mol^{-1} and so is relatively slow at r. t. Therefore, acceptable oxidation rates can only be achieved above 50 °C [16, 17].

The application of peroxydisulfate as an oxidant in organic synthesis has been widely investigated [18 – 24]. Recently, we have reported on the oxidation of various ethyl 3,4-dihydropyrimidin-2(1*H*)-one-5-carboxylates by potassium peroxydisulfate (PPS) under reflux condition [25] and also by combination of heat and ultrasound irradiation (sono-thermal reaction) [26]. The advantage of microwave techniques in shortening of reaction times led us to carry out the same reactions under microwave irradiation to investigate the following points: (i) the effect of the acetyl

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group instead of the carboethoxy group in position 5; (ii) the effect of the nature of the additional substituent and its location on the phenyl ring located on C-4; (iii) the effect of microwave irradiation on the rate of reaction and comparison of the results with those obtained under thermal and sono-thermal conditions; (iv) the study the occurrence of possible side reactions under microwave conditions.

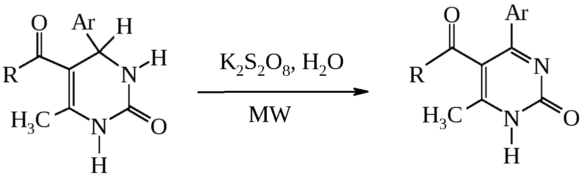
Results and Discussion

A solution of 0.23 mmol of various acetyl and carboethoxy derivatives of 3,4-dihydropyrimidin-2(1*H*)-ones and 0.23 mmol of potassium peroxydisulfate in 0.5 mL of water was irradiated by microwave at 900 Watt. The reaction was monitored by TLC until total disappearance of DHPMs (Scheme 1). The results are summarized in Table 1.

We have proposed earlier a reaction mechanism for the thermal oxidation of carboethoxy derivatives of 3,4-dihydropyrimidin-2(1*H*)-ones (**1a** – **1j**) by PPS (Scheme 2).

According to the proposed mechanism, thermal cleavage of PPS in the first step leads to the formation of potassium sulfate radicals, which abstract hydrogen from water in the second step to form hydroxyl radicals. Removal of hydrogen by the hydroxyl radical in the third step, followed by elimination of a second hydrogen atom from a dihydropyrimidinoyl radical intermediate in the last step, completes the reaction under formation of a pyrimidinone. The observed effect of the substituent in 4-position on the rate of reaction indicates that the removal of a hydrogen atom from the more covalent C–H bond compared with the less covalent N–H bond is more facile; therefore, step 3 should be the rate determining step. In fact, the stability of a dihydropyrimidinoyl radical intermediate, which is both a benzylic and an allylic radical, should lower the activation energy of its formation. This influences the rate of step 3 as the rate determining step.

The results presented in Table 1 indicate that in all cases the dihydropyrimidinone esters **1a** – **1j** were oxidized faster than the corresponding acetyl derivatives **1k** – **1v** under microwave irradiation. The results obtained from *ab initio* calculations at the B3LYP/6-31++G** level of theory for the optimized structures of these compounds can partially explain the observed comparative behavior of the two classes of compounds based on the following arguments: (i) the dihydropyrimidinone ring adopts a boat conformation, flattened at N1 toward an envelope conformation, with a pseudoaxial orientation of the C4-substituent. The extent of the deviation from planarity around C-4 depends on the orientation of the aryl group attached to this atom, especially on the position of an additional substituent at the phenyl ring; (ii) the dihedral angle of the ester carbonyl group or the acetyl moiety with respect to the C^5=C^6 bond depends on the type of the substituent on the C-4 atom.



1a-v **2a-v**

Scheme 1.

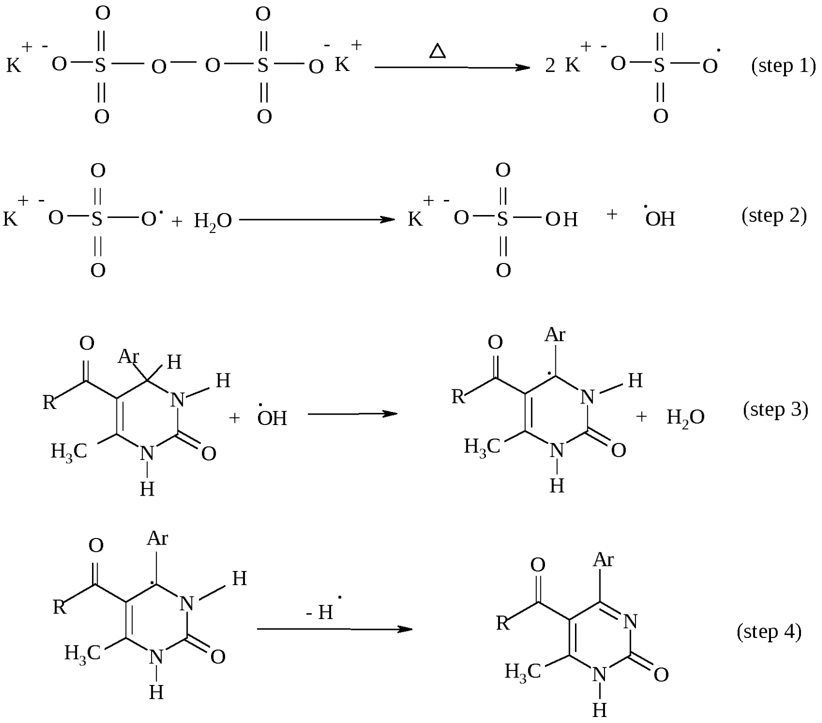
Table 1. Comparison of the reaction times of the oxidation of DHPMs under microwave irradiation.

DHPMs	Ar	R	Product	Time (sec)[a]	Yield (%)[b]
1a	C_6H_5-	C_2H_5O	**2a**	150	95
1b	4-$CH_3C_6H_4$-	=	**2b**	150	90
1c	4-$CH_3OC_6H_4$-	=	**2c**	60	95
1d	3-$CH_3OC_6H_4$-	=	**2d**	150	90
1e	2-$CH_3OC_6H_4$-	=	**2e**	90	93
1f	3-ClC_6H_4-	=	**2f**	90	90
1g	2-ClC_6H_4-	=	**2g**	90	95
1h	2-BrC_6H_4-	=	**2h**	60	90
1i	4-$NO_2C_6H_4$-	=	**2i**	60	87
1j	$PhCH_2CH_2$-	=	**2j**	120	90
1k	C_6H_5-	CH_3	**2k**	480	92
1l	4-$CH_3C_6H_4$-	=	**2l**	330	90
1m	4-$CH_3OC_6H_4$-	=	**2m**	300	88
1n	3-$CH_3OC_6H_4$-	=	**2n**	360	95
1o	2-$CH_3OC_6H_4$-	=	**2o**	240	90
1p	4-ClC_6H_4-	=	**2p**	300	93
1q	3-ClC_6H_4-	=	**2q**	300	95
1r	2-ClC_6H_4-	=	**2r**	240	90
1s	4-BrC_6H_4-	=	**2s**	390	90
1t	2-BrC_6H_4-	=	**2t**	360	92
1u	4-$NO_2C_6H_4$-	=	**2u**	390	87
1v	3-$NO_2C_6H_4$-	=	**2v**	420	90

[a] The times are given after total disappearance of DHPMs (100 % conversion according to TLC observation); [b] isolated yields.

The dihedral angle formed by the $C^{2'}$–$C^{1'}$–C^4–N^3 bonds depends on the stereoelectronic effect of the carboethoxy or acetyl groups located on C-5 and also on the location of the additional substituent on the phenyl ring located on C-4. Fig. 1 shows the optimized structures of **1a**, **1e**, **1k** and **1o** obtained at the B3LYP/6-31G(d,p) level of theory.

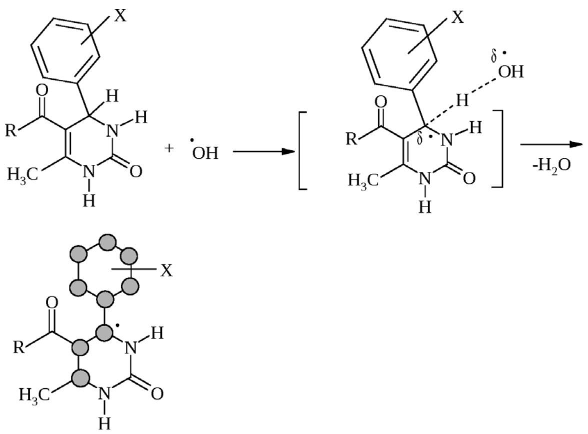
All experimental facts support our argument that the removal of 4-H by a hydroxyl radical is the rate determining step which is influenced by the above men-



Scheme 2.

tioned factors, since in the transition state the partial radical character formed on C-4 of the dihydropyrimidinone ring is stabilized by conjugation with the C^5=C^6 bond to form an allylic radical, and by forming a benzylic radical which is stabilized by interaction with the aromatic ring. The nature of the additional substituent and its position at the phenyl ring attached to C-4, such as the electron-donating species 4-methyl (**1b** or **1l**) or 4-methoxy (**1c** or **1m**) on the right position having no steric hindrance should stabilize the formation of the benzylic radical intermediate rather than an electron-withdrawing 4-nitro group (**1i** or **1u**), whereas the inductive effect of a methoxy group in the "wrong" position (**1d** or **1n**) should decrease the rate of reaction.

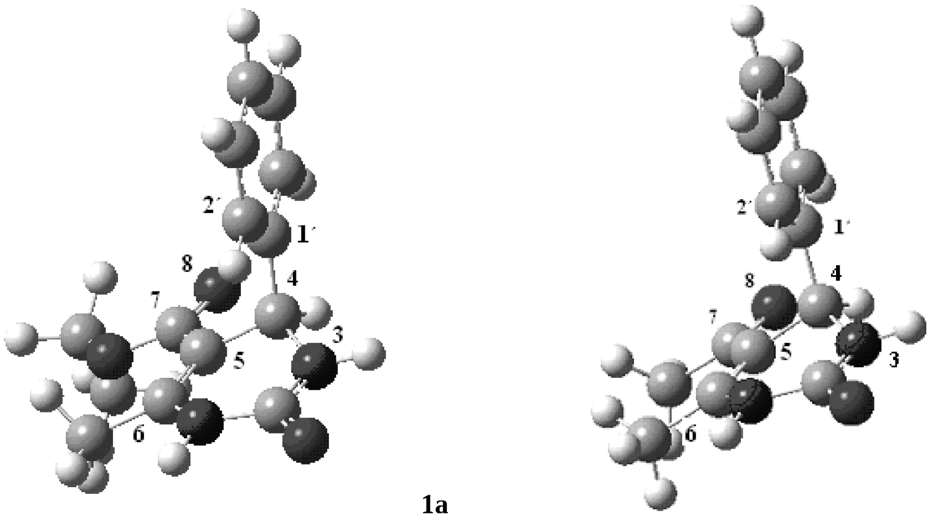
Scheme 3 shows the interaction of a hydroxyl radical with 4-H in the transition state leading to a dihydropyrimidinoyl radical intermediate. In this case, when the aromatic ring is perpendicular to the radical center formed on C-4, the stabilization of the radical center through conjugation with the aromatic ring is better, and simultaneously the steric hindrance for the linear approach of the hydroxyl radical to 4-H is diminished. This leads to an increase of the rate of reaction. In this case and as a comparative example, complete oxidation of **1e** and **1o** is observed after 90 and 240 s of irradiation, respectively. As a result, the balance of steric and electronic effects of the additional substituent on the phenyl ring located on C-4 and also those of the acetyl and the carboethoxy groups influence the rate of oxidation.



Scheme 3.

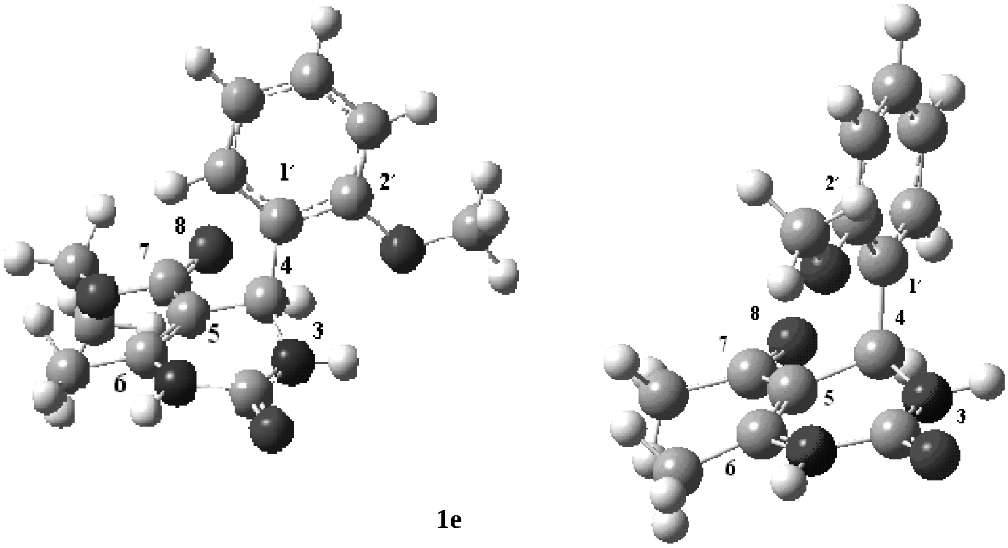
Effect of microwave energy

A comparison of the results presented in Table 1 with those obtained under thermal [25] or sonothermal [26] conditions indicates that, as expected,



1a **1k**

$O^8–C^7–C^5–C^6 = –174.0°$, $C^{2'}–C^{1'}–C^4–N^3 = –41.58°$, $O^8–C^7–C^5–C^6 = –175.0°$, $C^{2'}–C^{1'}–C^4–N^3 = 36.12°$.



1e **1o**

$O^8–C^7–C^5–C^6 = –178.8°$, $C^{2'}–C^{1'}–C^4–N^3 = +65.59°$, $O^8–C^7–C^5–C^6 = –169.8°$, $C^{2'}–C^{1'}–C^4–N^3 = –58.55°$.

Fig. 1. The optimized structures of **1a**, **1k**, **1e** and **1o** obtained at the B3LYP/6-31++G(d,p) level of theory.

a very fast reaction is observed under microwave irradiation, but the same products have been obtained in all reactions. The acceleration of oxidation under microwave irradiation is not only due to an increased temperature of the reaction medium as might be expected, although the temperature of water as solvent can be raised up to 105 °C under microwave exposure [27], above its normal boiling point of 100 °C, but other factors should also be considered for decreasing the reaction time under microwave irradiation.

It is true that the rapid heating by microwave exposure could be the reason of the increased rate of reaction under microwave conditions, but the specific effect derived from the microwave field, recognized as "microwave effect" should also be considered for the polar mechanism proposed in our study, especially when the polarity is increased during the reaction from the

Table 2. The effect of microwave power on the rate of oxidation of **1a** to **2a** by $K_2S_2O_8$ in H_2O.

Ratio $K_2S_2O_8$ / **1a**	Microwave power (Watt)	Time (sec)[a]	Yield **2a**[b] (%)
0.5 : 1	900	250	60
1 : 1	900	150	95
1 : 1	630	360 (70 °C)[c]	60
1 : 1	495	420 (60 °C)[c]	55
1 : 1	315	450 (55 °C)[c]	50
1 : 1	90	600 (50 °C)[c]	50

[a] The times after maximum progression of the reaction are given; [b] isolated yield; [c] the measured temperature after 30 s of MW irradiation.

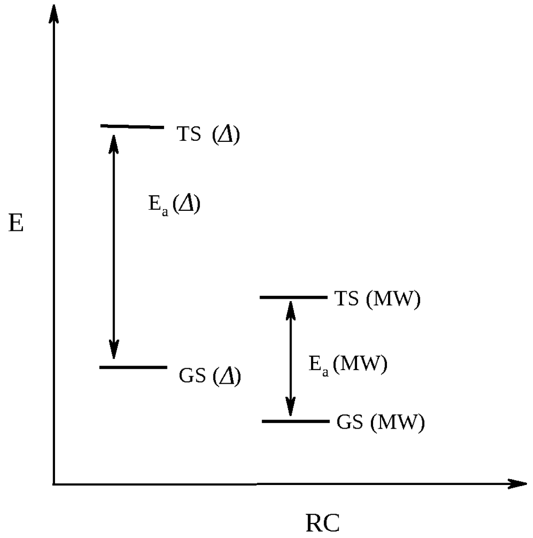


Fig. 2. Relative stabilization of the more polar transition state (TS) compared with the less polar ground state (GS).

ground state (GS) towards the transition state (TS) [28] (Fig. 2). Since the stabilization by dipole-dipole electrostatic interactions of the transition state is more effective than that of the ground state, an enhancement of the reactivity by a decrease in the activation energy should be expected using microwave irradiation. This may also be an explanation of the increased rate of oxidation using microwave irradiation compared with the reaction under reflux or sono-thermal conditions.

As a control experiment, we have carried out the oxidation of **1a** at different microwave power. The results presented in Table 2 indicate that (i) by increasing the microwave intensity, a decrease of the time of oxidation is observed; (ii) the optimized ratio of $K_2S_2O_8$/DHPM (1 : 1) indicates that the removal of two hydrogens is dependent on the presence of equimolar amounts of the oxidant and DHPM, because the reaction was not completed for the ratio 0.5 : 1 of oxidant to DHPM.

One reason is that by decreasing the microwave power the temperature is not raised too much; therefore, the cleavage of the O–O bond is slower, which is necessary for the reaction, but the important factor is the microwave effect, which depends on the applied microwave power. This phenomenon is clearly shown in Table 2.

Conclusion

We have developed an extremely simple, convenient, and efficient protocol for the oxidation of 3,4-dihydropyrimidin-2(1*H*)-one derivatives with potassium peroxydisulfate in water as solvent under microwave irradiation. In these oxidation reactions we used the lower molar ratio of DHPM : oxidant (1 : 1) compared with DHPM/CAN (1 : 5) in AcOH (55 – 68 % yield after 1 – 2.5 h) [9] and DHPM/$Co(NO_3)_2 \cdot 6H_2O/K_2S_2O_8$ (1 : 5 : 2.5) (69 – 81 % yield after 3 – 8 h) [10]. The balance of steric and electronic effects of the additional substituent on the phenyl ring located on C-4 and of the acetyl or the carboethoxy groups influence the rate of oxidation. The products obtained under microwave irradiation are the same as those obtained under thermal condition.

Experimental Section

Melting points were determined on a Stuart Scientific SMP2 apparatus and are uncorrected. IR spectra were recorded for solid samples using KBr discs on a Shimadzu IR spectrometer IR 435. The ^1H NMR spectra ([D$_6$]DMSO, standard TMS) were recorded with a Bruker 300 MHz machine. They are reported as follows: chemical shifts, (multiplicity, coupling constants *J* in Hz, number of protons, and assignment). Mass spectra were obtained on a Platform II spectrometer from Micromass; EI mode at 70 eV. UV spectra were taken with a Shimadzu UV-160 spectrometer.

*General procedure for the synthesis of 5-acetyl-6-methyl-3,4-dihydropyrimidin-2(1H)-ones (**1k – 1v**)*

A mixture of aldehyde (2 mmol), acetylacetone (0.26 g, 2 mmol), urea (0.18 g, 3 mmol) and NH_4Cl (0.05 g, 0.8 mmol) was heated with stirring at 100 °C for 3 h. After cooling, the reaction mixture was washed with cold water (2 × 50 mL), and the solid residue was recrystallized from ethanol [29].

General procedure for oxidation of 3,4-dihydropyrimidin-2(1H)-ones (DHPMs)

Potassium peroxydisulfate (61.5 mg, 0.23 mmol) was added to a solution of dihydropyrimidinones (DHPMs)

(0.23 mmol) in water (0.5 mL). The reaction mixture was irradiated under microwave irridiation for the times given in Table 1. The reaction mixture was irradiated in 30 s time intervals. Due to evaporation of water upon irradiation, water was added to keep a constant concentration of the reaction mixture during the irradiation. TLC monitoring of the reaction using *n*-hexane/ethyl acetate (2 : 1) as eluent was followed until the reaction was completed. Water was added, and the crude reaction mixture was extracted with 2 × 10 mL of diethyl ether. The ether was evaporated, and the residue was recrystallized from *n*-hexane/ethyl acetate. The spectral data of the products obtained by oxidation of the ester derivatives **2a** – **2j** were as reported earlier [26], and the data for acetyl derivatives **1k** – **1v** and the corresponding oxidation products **2k** – **2v** are presented here.

*5-Acetyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (**1k**)*

M. p. 228 – 230 °C (lit. [30]: 233 – 236 °C). – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 290.5 nm (4.10). – IR (KBr): ν = 1700 (CH_3CO), 1670 (CO), 1600 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 2.10 (s, 3H, CH_3), 2.28 (s, 3H, CH_3CO), 5.26 (d, J = 3.32 Hz, 1H, 4-H), 7.29 (m_c, 5H, H-aromatic), 7.81 (s, 1H, 1-NH), 9.16 (s, 1H, 3-NH).

*5-Acetyl-6-methyl-4-(4′-methylphenyl)-3,4-dihydropyrimidin-2(1H)-one (**1l**)*

M. p. 234 – 236 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 288.8 (3.56) and 241.4 nm (3.41). – IR (KBr): ν = 1640 (CH_3CO), 1590 (CO), 1505 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 2.07 (s, 3H, CH_3), 2.25 (s, 3H, CH_3CO), 2.27 (s, 3H, 4′-CH_3), 5.20 (s, 1H, 4-H), 7.12 (brd s, 4H, H-aromatic), 7.76 (s, 1H, 1-NH), 9.13 (s, 1H, 3-NH). – MS (EI, 70 eV): *m/z* (%) = 244 (14) $[M]^+$, 243 (25) $[M–H]^+$, 229 (49) $[M–CH_3]^+$, 201 (32) $[M–CH_3CO]^+$,153 (100) $[M–C_7H_7]^+$, 91 (30) $[Ph–CH_2]^+$.

*5-Acetyl-4-(4′-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1m**)*

M. p. 182 – 184 °C (lit. [30]: 168 – 170 °C). – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 286.4 nm (3.82). – IR (KBr): ν = 1650 (CH_3CO), 1580 (CO), 1430 (C=C) cm^{-1}.

*5-Acetyl-4-(3′-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1n**)*

M. p. 226 – 228 °C (lit. [31]: 228 – 230 °C, ethanol). – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 284.4 (3.89) and 239.6 nm (3.43). – IR (KBr): ν = 1670 (CH_3CO), 1590 (CO), 1425 (C=C) cm^{-1}.

*5-Acetyl-4-(2′-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1o**)*

M. p. 250 – 252 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 283.6 (4.42) and 240.2 nm (4.00). – IR (KBr): ν = 1670 (CH_3CO), 1580 (CO), 1430 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 2.00 (s, 3H, CH_3), 2.28 (s, 3H, CH_3CO), 3.81 (s, 3H, CH_3O), 5.56 (s, 1H, 4-H), 7.06 (m_c, 4H, H-aromatic), 7.33 (s, 1H, 1-NH), 9.11 (s, 1H, 3-NH). – MS (EI, 70 eV): *m/z* (%) = 260 (61) $[M]^+$, 259 (80) $[M–H]^+$, 245 (51) $[M–CH_3]^+$, 229 (92) $[M–CH_3O]^+$, 217 (85) $[M–CH_3CO]^+$, 153 (100) $[M–CH_3OC_6H_4]^+$.

*5-Acetyl-4-(4′-chlorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1p**)*

M. p. 249 – 251 °C (lit. [32]: 223 – 225 °C). – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 290.6 (4.04) and 240.0 nm (3.78). – IR (KBr): ν = 1690 (CH_3CO), 1615 (CO), 1420 (C=C) cm^{-1}.

*5-Acetyl-4-(3′-chlorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1q**)*

M. p. 285 – 287 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 291.4 (3.08) and 239.8 nm (2.70). – IR (KBr): ν = 1700 (CH_3CO), 1615 (CO), 1525 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 2.15 (s, 3H, CH_3), 2.30 (s, 3H, CH_3CO), 5.27 (d, J = 3.25 Hz, 1H, 4-H), 7.27 (m_c, 4H, H-aromatic), 7.87 (s, 1H, 1-NH), 9.28 (s, 1H, 3-NH). – MS (EI, 70 eV): *m/z* (%) = 266 (49) $[M^{37}Cl]^+$, 265 (64) $[M^{37}Cl–H]^+$, 264 (32) $[M^{35}Cl]^+$, 263 (79) $[M^{35}Cl–H]^+$, 249 (80) $[M^{35}Cl–CH_3]^+$ 229 (42) $[M^{35}Cl–^{35}Cl]^+$, 223 (28) $[M^{37}Cl–CH_3CO]^+$, 221 (74) $[M^{35}Cl–CH_3CO]^+$, 153 (100) $[M–ClC_6H_4]^+$, 141 (9) $[^{37}ClC_6H_4\text{-}CH{=}NH]^+$, 140 (17) $[^{37}ClC_6H_4\text{-}C{=}NH]^+$, 139 (19) $[^{35}ClC_6H_4\text{-}CH{=}NH]^+$, 138 (25) $[^{35}ClC_6H_4\text{-}C{=}NH]^+$.

*5-Acetyl-4-(2′-chlorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1r**)*

M. p. 262 – 264 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 291.0 (4.00) and 240.2 nm (3.62). – IR (KBr): ν = 1690 (CH_3CO), 1615 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 2.05 (s, 3H, CH_3), 2.33 (s, 3H, CH_3CO), 5.66 (s, 1H, 4-H), 7.36 (m_c, 4H, H-aromatic), 7.72 (s, 1H, 1-NH), 9.27 (s, 1H, 3-NH). – MS (EI, 70 eV): *m/z* (%) = 266 (4) $[M^{37}Cl]^+$, 265 (7) $[M^{37}Cl–H]^+$, 264 (10) $[M^{35}Cl]^+$, 263 (16) $[M^{35}Cl–H]^+$, 249 (10) $[M^{35}Cl–CH_3]^+$, 231 (6) $[M^{37}Cl–^{37}Cl]^+$, 229 (94) $[M^{35}Cl–^{35}Cl]^+$, 223 (6) $[M^{37}Cl–CH_3CO]^+$, 221 (72) $[M^{35}Cl–CH_3CO]^+$, 153 (100) $[M–ClC_6H_4]^+$, 141 (9) $[^{37}ClC_6H_4\text{-}CH{=}NH]^+$, 140 (17) $[^{37}ClC_6H_4\text{-}C{=}NH]^+$, 139 (19) $[^{35}ClC_6H_4\text{-}CH{=}NH]^+$, 138 (25) $[^{35}ClC_6H_4\text{-}C{=}NH]^+$.

*5-Acetyl-4-(4′-bromophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1s**)*

M. p. 232 – 233 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 290.8 (4.04) and 240.2 nm (3.85). – IR (KBr): ν = 1650 (CH_3CO), 1580 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 2.12 (s, 3H, CH_3), 2.28 (s, 3H, CH_3CO), 5.23 (s, 1H, 4-H), 7.18 (d, *J* = 6.90 Hz, 2′- and 6′-H), 7.51 (d, *J* = 6.77 Hz, 3′- and 5′-H), 7.88 (s, 1H, 1-NH), 9.23 (s, 1H, 3-NH).

*5-Acetyl-4-(2′-bromophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (**1t**)*

M. p. 254 – 257 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 292.2 (3.98) and 239.8 nm (3.70). – IR (KBr): ν = 1700 (CH_3CO), 1620 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 2.04 (s, 3H, CH_3), 2.33 (s, 3H, CH_3CO), 5.62 (d, *J* = 2.85 Hz, 1H, 4-H), 7.39 (m_c, 4H, H-aromatic), 7.69 (brd s, 1H, 1-NH), 9.28 (s, 1H, 3-NH). – MS (EI, 70 eV): *m/z* (%) = 267 (10) $[M^{81}Br-CH_3CO]^+$, 265 (11) $[M^{79}Br-CH_3CO]^+$, 231 (2) $[M^{81}Br-^{81}Br]^+$, 229 (97) $[M^{79}Br-^{79}Br]^+$, 214 (13) $[M^{79}Br-^{79}Br-CH_3]^+$, 185 (10) $[^{81}BrC_6H_4\text{-}CH=NH]^+$, 184 (4) $[^{79}BrC_6H_4\text{-}C=NH]^+$, 183 (3) $[^{79}BrC_6H_4\text{-}CH=NH]^+$, 182 (3) $[2\text{-}BrC_6H_4\text{-}C=NH]^+$, 153 (100) $[M-BrC_6H_4]^+$.

*5-Acetyl-6-methyl-4-(4′-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (**1u**)*

M.p. 229 – 230 °C (dec.) [lit. [29]: 230 °C (dec.)]. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 279.2 nm (4.07). – IR (KBr): ν = 1650 (CH_3CO), 1580 (CO), 1520 (C=C) cm^{-1}.

*5-Acetyl-6-methyl-4-(3′-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (**1v**)*

M. p. 286 – 288 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 282.6 nm (3.98). – IR (KBr): ν = 1650 (CH_3CO), 1585 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 2.19 (s, 3H, CH_3), 2.32 (s, 3H, CH_3CO), 5.45 (d, *J* = 3.24 Hz, 1H, 4-H), 7.99 (brd s, 1H, 1-NH), 8.11 (m_c, 4H, H-aromatic), 9.34 (s, 1H, 3-NH). – MS (EI, 70 eV): *m/z* (%) = 259 (5) $[M-OH]^+$, 258 (32) $[M-H_2O]^+$, 232 (7) $[M-CH_3CO]^+$, 228 (27) $[M-HNO_2]^+$, 153 (100) $[O_2NC_6H_4\text{-}CH=NH]^+$.

*5-Acetyl-6-methyl-4-phenylpyrimidin-2(1H)-one (**2k**)*

M. p. 162 – 163 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 307.5 (3.32) and 249.0 nm (2.52). – IR (KBr): ν = 1700 (CH_3CO), 1670 (CO), 1590 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 1.84 (s, 3H, 6-CH_3), 2.30 (s, 3H, CH_3CO), 7.52 (m_c, 5H, H-aromatic), 12.33 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 228 (42) $[M]^+$, 227 (49) $[M-H]^+$, 213 (100) $[M-CH_3]^+$, 185 (11) $[M-CH_3CO]^+$, 104 (61) $[C_6H_5\text{-}C=NH]^+$, 103 (7) $[C_6H_5\text{-}CN]^+$, 77 (44) $[C_6H_5]^+$.

*5-Acetyl-6-methyl-4-(4′-methylphenyl)-pyrimidin-2(1H)-one (**2l**)*

M. p. 219 – 221 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 320.0 (sh, 3.48) and 259.5 nm (3.70). – IR (KBr): ν = 1695 (CH_3CO), 1590 (CO), 1510 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 1.85 (s, 3H, CH_3), 2.29 (s, 3H, CH_3CO), 2.50 (s, 3H, 4′-CH_3), 7.31 (d, *J* = 7.83 Hz, 2H, 2-H′ and 6-H′), 7.37 (d, *J* = 8.52 Hz, 2H, 3-H′ and 5-H′), 12.26 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 242 (46) $[M]^+$, 241 (32) $[M-H]^+$, 227 (100) $[M-CH_3]^+$, 199 (11) $[M-CH_3CO]^+$, 117 (4) $[CH_3C_6H_4\text{-}C=NH]^+$, 116 (8) $[CH_3C_6H_4\text{-}CN]^+$, 91 (37) $[\text{-}C_6H_4\text{-}CH_3]^+$.

*5-Acetyl-4-(4′-methoxyphenyl)-6-methylpyrimidin-2(1H)-one (**2m**)*

M. p. 189 – 191 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 296.5 (3.08) and 255.0 nm (3.08). – IR (KBr): ν = 1670 (CH_3CO), 1680 (CO), 1425 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 1.87 (s, 3H, 6-CH_3), 2.28 (s, 3H, CH_3CO), 3.82 (s, 3H, CH_3O), 7.06 (d, *J* = 8.55 Hz, 2H, 2-H′ and 6-H′), 7.45 (d, *J* = 8.52, 2H, 3-H′ and 5-H′), 12.18 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 258 (84) $[M]^+$, 257 (40) $[M-H]^+$, 243 (100) $[M-CH_3]^+$, 227 (10) $[M-CH_3O]^+$, 215 (11) $[M-CH_3CO]^+$, 200 (18) $[M-CH_3CO-CH_3]^+$, 134 (71) $[CH_3OC_6H_4\text{-}CH=NH]^+$, 133 (4) $[CH_3OC_6H_4\text{-}C=NH]^+$, 132 (3) $[CH_3OC_6H_4\text{-}CN]^+$.

*5-Acetyl-4-(3′-methoxyphenyl)-6-methylpyrimidin-2(1H)-one (**2n**)*

M. p. 163 – 134 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 303.5 (3.75) and 250.0 nm (3.88). – IR (KBr): ν = 1675 (CH_3CO), 1595 (CO), 1425 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 1.85 (s, 3H, 6-CH_3), 2.28 (s, 3H, CH_3CO), 3.80 (s, 3H, 4′-CH_3O), 6.98 (d, *J* = 7.33 Hz, 2H, 4′-H), 7.03 (s, 1H, 2′-H), 7.12 (d, *J* = 8.20 Hz, 2H, 6′-H), 7.41 (t, *J* = 6.67 Hz, *J* = 7.55 Hz, 1H). – MS (EI, 70 eV): *m/z* (%) = 258 (3) $[M]^+$, 257 (2) $[M-H]^+$, 243 (2) $[M-CH_3]^+$, 200 (2) $[M-CH_3CO-CH_3]^+$, 134 (14) $[CH_3OC_6H_4\text{-}CH=NH]^+$, 133 (7), $[CH_3OC_6H_4\text{-}C=NH]^+$, 132 (8) $[CH_3OC_6H_4\text{-}CN]^+$, 77 (100).

*5-Acetyl-4-(2′-methoxyphenyl)-6-methylpyrimidin-2(1H)-one (**2o**)*

M. p. 166 – 167 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 303.0 (4.15) and 258.0 nm (4.15). – IR (KBr): ν = 1960 (CH_3CO), 1590 (CO), 1550 (C=C) cm^{-1}. – ^1H NMR (300 MHz, [D_6]DMSO): δ = 1.87 (s, 3H, CH_3), 2.32 (s, 3H, CH_3CO), 3.70 (s, 3H, CH_3O), 7.24 (m_c, 4H, H-aromatic), 12.12 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 258 (6) $[M]^+$, 257 (4) $[M-H]^+$, 243 (13) $[M-CH_3]^+$, 227 (100) $[M–CH_3O]^+$, 215 (22) $[M–CH_3CO]^+$, 134 (38) $[CH_3OC_6H_4\text{-}CH{=}NH]^+$, 133 (10) $[CH_3OC_6H_4\text{-}C{=}NH]^+$, 132 (6) $[CH_3OC_6H_4\text{-}CN]^+$.

5-Acetyl-4-(4′-chlorophenyl)-6-methylpyrimidin-2(1H)-one ***(2p)***

M. p. 235 – 237 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 313.0 (3.34) and 252.0 nm (3.56). – IR (KBr): ν = 1650 (CH_3CO), 1580 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 1.90 (s, 3H, CH_3), 2.30 (s, 3H, CH_3CO), 7.49 (d, *J* = 8.27 Hz, 2H, 3-H′ and 5-H′), 7.58 (d, *J* = 8.21 Hz, 2H, 2-H′ and 6-H′), 12.21 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 264 (27) $[M^{37}Cl]^+$, 263 (32) $[M^{37}Cl–H]^+$, 262 (78) $[M^{35}Cl]^+$, 261 (59) $[M^{35}Cl–H]^+$, 249 (69) $[M^{37}Cl–CH_3]^+$, 247 (99) $[M^{35}Cl–CH_3]^+$, 227 (17) $[M–Cl]^+$, 221 (5) $[M^{37}Cl–CH_3CO]^+$, 219 (13) $[M^{35}Cl–CH_3CO]^+$, 140 (42) $[^{37}ClC_6H_4\text{-}C{=}NH]^+$, 139 (19) $[^{37}ClC_6H_4\text{-}CN]^+$, 138 (87) $[^{35}ClC_6H_4\text{-}C{=}NH]^+$, 137 (12) $[^{35}ClC_6H_4\text{-}CN]^+$.

5-Acetyl-4-(3′-chlorophenyl)-6-methylpyrimidin-2(1H)-one ***(2q)***

M. p. 169 – 198 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 320 (sh, 3.99), 304 (4.03) and 259.0 nm (4.18). – IR (KBr): ν = 1650 (CH_3CO), 1580 (CO), 1430 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 1.90 (s, 3H, CH_3), 2.30 (s, 3H, CH_3CO), 7.54 (m_c, 4H, H-aromatic), 9.27 (s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 264 (21) $[M^{37}Cl]^+$, 263 (23) $[M^{37}Cl\ –H]^+$, 262 (58) $[M^{35}Cl]^+$, 261 (38) $[M^{35}Cl–H]^+$, 249 (45) $[M^{37}Cl–CH_3]^+$, 247 (100) $[M^{35}Cl–CH_3]^+$, 227 (14) $[M–Cl]^+$, 221 (50) $[M^{37}Cl–CH_3CO]^+$, 219 (90) $[M^{35}Cl–CH_3CO]^+$, 140 (36) $[^{37}ClC_6H_4\text{-}C{=}NH]^+$, 139 (18) $[^{37}ClC_6H_4\text{-}CN]^+$, 138 (78) $[^{35}ClC_6H_4\text{-}C{=}NH]^+$, 137 (9) $[^{35}ClC_6H_4\text{-}CN]^+$.

5-Acetyl-4-(2′-chlorophenyl)-6-methylpyrimidin-2(1H)-one ***(2r)***

M. p. 175 – 176 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 305.0 (3.34) and 259.0 nm (3.45). – IR (KBr): ν = 1700 (CH_3CO), 1620 (CO), 1430 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 1.90 (s, 3H, CH_3), 2.30 (s, 3H, CH_3CO), 7.47 (m_c, 4H, H-aromatic), 12.26 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 247 (5) $[M^{35}Cl–CH_3]^+$, 227 (100) $[M–Cl]^+$, 219 (3) $[M^{35}Cl–CH_3CO]^+$, 140 (24) $[^{37}ClC_6H_4\text{-}C{=}NH]^+$, 139 (13) $[^{37}ClC_6H_4\text{-}CN]^+$, 138 (47) $[^{35}ClC_6H_4\text{-}C{=}NH]^+$, 137 (8) $[^{35}ClC_6H_4\text{-}CN]^+$.

5-Acetyl-4-(4′-bromophenyl)-6-methylpyrimidin-2(1H)-one ***(2s)***

M. p. 239 – 240 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 332 (sh, 3.36), 304 (sh, 3.48) and 252.0 nm (3.76). – IR (KBr): ν = 1670 (CH_3CO), 1620 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 1.91 (s, 3H, CH_3), 2.30 (s, 3H, CH_3CO), 7.41 (d, *J* = 8.18 Hz, 2H, 3-H′ and 5-H′), 7.72 (d, *J* = 8.12, 2H, 2-H′ and 6-H′), 12.30 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 308 (38) $[M^{81}Br]^+$, 307 (32) $[M^{81}Br–H]^+$, 306 (37) $[M^{79}Br]^+$, 305 (26) $[M^{79}Br–H]^+$, 293 (81) $[M^{81}Br–CH_3]^+$, 291 (83) $[M^{79}Br–CH_3]^+$, 265 (60) $[M^{81}Br–CH_3CO]^+$, 263 (7) $[M^{79}Br–CH_3CO]^+$, 227 (24) $[M–Br]^+$, 185 (16) $[^{81}BrC_6H_4\text{-}CH{=}NH]^+$, 183 (61) $[^{79}BrC_6H_4\text{-}CH{=}NH]^+$, 182 (16) $[^{79}BrC_6H_4\text{-}C{=}NH]^+$, 181 (45) $[^{79}BrC_6H_4\text{-}CN]^+$.

5-Acetyl-4-(2′-bromophenyl)-6-methylpyrimidin-2(1H)-one ***(2t)***

M. p. 202 – 204 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 301.5 (3.89) and 264.0 nm (3.86). – IR (KBr): ν = 1700 (CH_3CO), 1615 (CO), 1420 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 1.85 (s, 3H, CH_3), 2.37 (s, 3H, CH_3CO), 7.43 (m_c, 3H, H-aromatic), 7.73 (d, *J* = 7.82 Hz, 1H, 6-H′), 12.36 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 227 (100) $[M–Br]^+$, 185 (10), $[^{81}BrC_6H_4\text{-}CH{=}NH]^+$, 184 (26) $[^{81}BrC_6H_4\text{-}C{=}NH]^+$, 183 (3) $[^{79}BrC_6H_4\text{-}CH{=}NH]^+$, 182 (6) $[^{79}BrC_6H_4\text{-}C{=}NH]^+$.

5-Acetyl-6-methyl-4-(4′-nitrophenyl)-pyrimidin-2(1H)-one ***(2u)***

M. p. 265 – 267 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 330 (sh, 3.71), 302 (sh, 3.86) and 262.0 nm (4.07). – IR (KBr): ν = 1665 (CH_3CO), 1600 (CO), 1505 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 2.35 (s, 3H, CH_3CO), 1.95 (s, 3H, CH_3), 7.73 (d, *J* = 8.52 Hz, 2H, 2-H′ and 6-H′), 8.33 (d, *J* = 8.49, 2H, 3-H′ and 5-H′), 9.27 (s, 1H, NH), 12.48 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 273 (20) $[M]^+$, 272 (13) $[M–H]^+$, 258 (100) $[M–CH_3]^+$, 256 (25) $[M–OH]^+$, 226 (12) $[M–HNO_2]^+$.

5-Acetyl-6-methyl-4-(3′-nitrophenyl)-pyrimidin-2(1H)-one ***(2v)***

M. p. 256 – 258 °C. – UV/Vis (CH_3CN): $\lambda_{max}(\lg\varepsilon_{max})$ = 305.0 (3.57) and 259.0 nm (3.85). – IR (KBr): ν = 1675 (CH_3CO), 1590 (CO), 1520 (C=C) cm^{-1}. – ^1H NMR (300 MHz, $[D_6]DMSO$): δ = 1.97 (s, 3H, CH_3), 2.35 (s, 3H, CH_3CO), 7.80 (t, *J* = 8.80 Hz, 1H, 5-H′), 7.88 (d, *J* = 7.49 Hz, 1H, 6-H′), 8.29 (s, 1H, 2-H′), 8.39 (d, *J* = 7.96 Hz, 1H, 4-H′), 12.48 (brd s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 273 (15) $[M]^+$, 272 (8) $[M–H]^+$, 258 (100) $[M–CH_3]^+$, 256 (30) $[M–OH]^+$, 226 (16) $[M–HNO_2]^+$.

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