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Synthesis and Characterization of Diazenyl-1,3-diphenylpropane-1,3-dione and 5-[Hydroxy(phenyl)methyl]-4-phenylpyrimidine Derivatives

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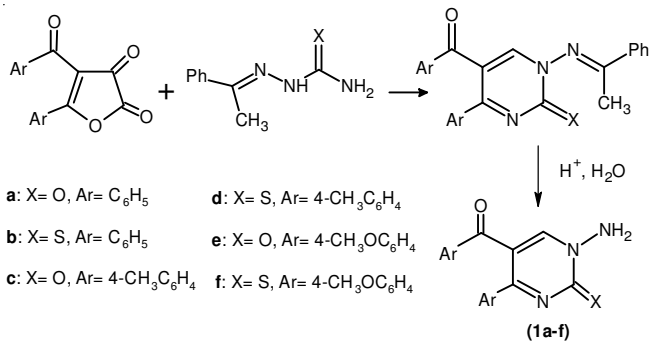
Various novel diazenyl-1,3-diphenylpropane-1,3-dione derivatives (**2a-f**) were prepared by the condensation of the diazonium salts of N-aminopyrimidine derivatives with β-diketones. Also, a series of novel 5-[hydroxy(phenyl)methyl]-4-phenylpyrimidine derivatives (**3a-f**) were synthesized from the reduction reactions of *N*-aminopyrimidine derivatives with $NaBH_4$. The structures of all the new synthesized compounds were characterized by the ^{13}C-NMR, ^{1}H-NMR and IR spectroscopic data and elemental analysis.

Keywords: N-Aminopyrimidines, Diazonium salts, β-Diketones, Reduction Reactions.

INTRODUCTION

Pyrimidine derivatives are well known pharmacophores exhibiting a wide spectrum of pharmacological activities. They show various interesting pharmacological properties including antiviral, antibacterial, antitumour and antiflammatory effects. Some of them are frequently encountered in many drugs used for the treatment of hypothyroidy, hypertension, cancer chemotherapy and HIV infection[1-5]. There are published reports about the synthesis of some pyrimidine derivatives from furan-2,3-diones[6-11] (**Scheme-I**). Also conformational analysis and quantum chemical calculations were carried out by means of MMP2, CNDO, MNDO and AM1 approximation methods for a series of compounds which are functionalized pyrimidine derivatives[8,9]. The N-aminopyrimidine derivatives **1a-f** which exhibit a free N-NH_2 moiety were applied in several subsequent reactions. Recently, the reactions of *N*-aminopyrimidine derivatives **1a-f** with several anhydrides, isocyanates, isothiocyanates and 1,3-dicarbonyl compounds have been reported in different solvents at various temperatures[12-17].



Scheme-I: Synthesis of *N*-aminopyrimidine derivatives **1a-f**

In view of these important properties, we decided both to prove the producibility of the reactions of *N*-aminopyrimidine derivatives **1a-f** with β-diketones and sodium borohydride and to extend our investigations related to the preparation of new heterocycles which include the pyrimidine ring in their structures. In the present study, the synthesis of some new diazenyl-1,3-diphenylpropane-1,3-dione derivatives **2a-f** were carried out (**Scheme-II**). In addition, in this work, a series of novel 5-[hydroxy(4-methylphenyl)methyl]-4-(methylphenyl)pyrimidine derivatives **3a-f** were synthesized by treating *N*-aminopyrimidine derivatives **1a-f** with $NaBH_4$ (**Scheme-III**). The newly synthesized compounds were established on the basis of elemental analysis, IR, ^{1}H and ^{13}C-NMR spectral studies.

EXPERIMENTAL

Melting points were determined on an Electrothermal 9200 apparatus and are uncorrected. Microanalyses were performed on a Leco-932 CHNS-O Elemental Analyzer, model 1108. A Shimadzu FT-IR-8400 model spectrophotometer was used for IR spectra (4000-400 cm^{-1} region), using ATR techniques. The ^{1}H- and ^{13}C-NMR spectra were measured with a Bruker Avance III 400 MHz spectrometer and the chemical shifts were recorded in ppm units. After completion of the reactions, the solvents were evaporated with a rotary evaporator (Buchi RE model 111). The reactions were followed by TLC using a DC Alufolien Kieselgel 60 F254 Merck and a Camag

TLC lamp (254/366 nm). Solvents and all other chemical reagents were purchased from commercial suppliers and were of reagent grade quality. Solvents were dried by refluxing with the appropriate drying agents and distilled before use.

General procedure for the synthesis of diazenyl-1,3-diphenylpropane-1,3-dione derivatives (2a-f): 1-Amino-5-benzoyl-4-phenyl-1*H*-pyrimidine-2-ones/-thiones (**1a-f**) (3.43 mmol) in 30 mL of EtOH was warmed until dissolved. The mixture was cooled to room temperature, 0.9 mL (10.29 mmol) of concentrated HCl was added and the reaction mixture was cooled to a temperature below 5 °C. And then the mixture was added to an ice cold solution of $NaNO_2$ (3.43 mmol) in water (10 mL) drop wise with constant stirring. Temperature was maintained below 5 °C. The diazonium salts of (**1a-f**) were reacted with dibenzoylmethane (molar ratio1:1) in the presence of NaOH. The reaction mixture continuously stirring for 5-9 h at 60-70 °C. Then the reaction mass was poured into ice-cold water and acidified. The solid obtained (**2a-f**) was filtered and recrystallized from alcohols and dried on P_2O_5.

2-[(E)-(5-Benzoyl-2-oxo-4-phenylpyrimidine-1(2*H*)-yl)diazenyl]-1,3-diphenylpropane-1,3-dione (2a): The obtained product **2a** was crystallized in *n*-butanol and dried on P_2O_5 and was obtained in 50 % yield as a colourless mass; m.p. 248-249 °C. IR: (KBr, ν_{max}, cm^{-1}): 3050 (aromatic C-H), 2930 (aliphatic C-H), 1730, 1680, 1650 (C=O), 1620 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 14.90 (s,1H, enol OH), 7.36-7.02 (m, 20H, Ar-H), 6.08 (s, 1H, C-H); ^{13}C-NMR (400 MHz, (DMSO-d_6): δ = 190.80-188.10 (benzoyl groups, C=O), 153.15 (pyrimidine ring, C=O), 150.05-110.42 (aromatic C), 74.82 ppm (C-H). Anal. Calcd. for $C_{32}H_{22}N_4O_4$: C, 72.99; H, 4.21; N, 10.64. Found: C, 72.89; H, 4.15; N, 10.50.

2-[(E)-(5-Benzoyl-2-thioxo-4-phenylpyrimidine-1(2*H*)-yl)diazenyl]-1,3-diphenylpropane-1,3-dione (2b): The obtained product **2b** was crystallized in *n*-butanol and dried on P_2O_5 and was obtained in 50 % yield as a yellow mass; m.p. 255-256 °C. IR (KBr, ν_{max}, cm^{-1}): 3050 (aromatic C-H), 2930 (aliphatic C-H), 1735 and 1645 (C=O), 1620 (C=C and C=N), 1245 (C=S), 800-620 cm^{-1} (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 14.80 (s, 1H, enol OH), 7.48-7.02 (m, 20H, Ar-H), 5.98 (s, 1H, C-H); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 191.50-187.10 (benzoyl groups, C=O), 170.15 (pyrimidine ring, C=S), 151.05-110.00 (aromatic C), 74.52 ppm (C-H). Anal. Calcd. for $C_{32}H_{22}N_4O_3S$: C, 70.83; H, 4.09; N, 10.33; S, 5.91. Found: C, 70.71; H, 4.01; N, 10.25; S, 5.80.

2-{(E)-[5-(4-Methylbenzoyl)-4-(4-methylphenyl)-2-oxopyrimidine-1(2*H*)-yl]diazenyl}-1,3-diphenyl- propane-1,3-dione (2c): The obtained product **2c** was crystallized in ethanol and dried on P_2O_5 and was obtained in 57 % yield as a colourless mass; m.p. 270-271 °C. IR (KBr, ν_{max}, cm^{-1}): 3040 (aromatic C-H), 2930 (aliphatic C-H), 1735, 1685 and 1655 (C=O), 1620 (C=C and C=N), 1245 (C=S), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 14.50 (s, 1H, enol OH), 7.56-7.05 (m, 18H, Ar-H), 6.08 (s, 1H, C-H); 2.14, 2.05 (s, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 190.80-188.10 (C=O, benzoyl groups), 153.15 (C=O, pyrimidine ring), 150.05-110.42 (aromatic C), 74.82 (C-H), 22.15, 21.64 ppm (2 × $\underline{C}H_3C_6H_4$-). Anal. Calcd. for $C_{34}H_{26}N_4O_4$: C, 73.63; H, 4.73; N, 10.10. Found: C, 73.45; H, 4.54; N, 10.22.

2-{(E)-[5-(4-Methylbenzoyl)-4-(4-methylphenyl)-2-thioxopyrimidine-1(2*H*)-yl]diazenyl}-1,3-dipheny-lpropane-1,3-dione (2d): The obtained product **2d** was crystallized in ethanol and dried on P_2O_5 and was obtained in 58 % yield as a yellow mass; m.p. 285-286 °C. IR (KBr, ν_{max}, cm^{-1}): 3040 (aromatic C-H), 2920 (aliphatic C-H), 1735, 1645 (C=O), 1620 (C=C and C=N), 1245 (C=S), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 14.62 (s, 1H, enol OH), 7.70-7.05 (m, 18H, Ar-H), 6.01 (s, 1H, C-H); 2.15, 2.05 (s, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 190.80-188.10 (C=O, benzoyl groups), 170.15 (C=S, pyrimidine ring), 158.15 (pyrimidine ring, C=S), 150.05-110.42 (aromatic C), 73.82 (C-H), 22.15, 21.64 ppm (2 × $\underline{C}H_3C_6H_4$-). Anal. Calcd. for $C_{34}H_{26}N_4O_3S$: C, 71.56; H, 4.59; N, 9.82; S, 7.63. Found: C, 71.38; H, 4.42; N, 9.64; S, 7.50.

2-{(E)-[5-(4-Methoxybenzoyl)-4-(4-methoxyphenyl)-2-oxopyrimidine-1(2*H*)-yl]diazenyl}-1,3-diphenylpropane-1,3-dione (2e): The obtained product **2e** was crystallized in ethanol and dried on P_2O_5 and was obtained in 63 % yield as a colourless mass; m.p. 273-274 °C. IR (KBr, ν_{max}, cm^{-1}): 3040 (aromatic C-H), 2935 (aliphatic C-H), 1730, 1650 C=O), 1620 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 14.56 (s, 1H, enol OH), 7.78-7.05 (m, 18H, Ar-H), 6.01 (s, 1H, C-H); 3.89, 3.86 (s, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 190.80-189.10 (C=O, benzoyl groups), 152.15 (C=O, pyrimidine ring), 151.05-115.42 (aromatic C), 73.82 (C-H), 55.90, 54.89 ppm (2 × $CH_3OC_6H_4$-). Anal. Calcd. for $C_{34}H_{26}N_4O_6$: C, 69.62; H, 4.47; N, 9.55. Found: C, 69.55; H, 4.36; N, 9.42.

2-{(E)-[5-(4-methoxybenzoyl)-4-(4-methoxyphenyl)-2-thioxopyrimidine-1(2*H*)-yl]diazenyl}-1,3-diphenyl-propane-1,3-dione (2f): The obtained product **2f** was crystallized in *n*-butanol and dried on P_2O_5 and was obtained in 67 % yield as a light yellow mass; m.p. 288-289 °C. IR (KBr, ν_{max}, cm^{-1}): 3040 (aromatic C-H), 2935 (aliphatic C-H), 1730-1650 C=O), 1620 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 14.52 (s, 1H, enol OH), 7.87-7.05 (m, 18H, Ar-H), 6.03 (s, 1H, C-H); 3.88, 3.84 (s, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 189.80-188.10 (C=O, benzoyl groups), 170.30 (C=S, pyrimidine ring), 155.01-112.22 (aromatic C), 72.82 (C-H), 55.40, 54.90 ppm (2 × $\underline{C}H_3OC_6H_4$-). Anal. Calcd. for $C_{34}H_{26}N_4O_5S$: C, 67.76; H, 4.35; N, 9.30; S, 5.32. Found: C, 67.58; H, 4.26; N, 9.15; S, 5.19.

General procedure for the synthesis of 5-[hydroxy-(phenyl)methyl]-4-phenylpyrimidine derivatives (3a-f): To a solution of 1-amino-5-benzoyl-4-phenyl-1*H*-pyrimidine-2-ones/-thiones (**1a-f**) (0.4 g, 1.36 mmol) in EtOH (50 mL) was added $NaBH_4$ (0.051 g, 1.36 mmol) in portions at 0.5 h intervals by stirring. The reaction mixture was refluxed for 1 h. It was then poured into ice-cold water and 5 mL of concentrated hydrochloric acid, after which it was extracted with $CHCl_3$. The $CHCl_3$ was evaporated *in vacuo* and crude product recrystallized from a suitable solvent (*i.e.* *n*-butanol or ethanol) and dried on P_2O_5.

1-Amino-5-[hydroxy(phenyl)methyl]-4-phenylpyrimidine-2(1*H*)-one (3a): The obtained product **3a** was crystallized in *n*-butanol and dried on P_2O_5 and was obtained in 54 % yield as a colourless mass; m.p. 255-256 °C. IR (KBr, ν_{max},

cm^{-1}): 3300 (O-H), 3200 (N-H), 3058 (aromatic C-H), 1650 (C=O), 1620 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.51 (s, 1H, pyrimidine C-H), 7.38-7.02 (m, 10H, Ar-H), 5.91 (s, 2H, N-H); 5.74, (s, 1H, C-H); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 152.80 (C=O, pyrimidine ring), 143.15-117.42 (aromatic C), 73.08 ppm (CH-OH). Anal. Calcd. for for $C_{17}H_{15}N_3O_2$: C, 69.61; H, 5.15; N, 14.33. Found: C, 69.49; H, 5.30; N, 14.15.

1-Amino-5-[hydroxy(phenyl)methyl]-4-phenylpyrimidine-2(1*H*)-thione (3b): The obtained product **3b** was crystallized in EtOH and dried on P_2O_5 and was obtained in 59 % yield as a light yellow mass; m.p. 235-236 °C. IR (KBr, ν_{max}, cm^{-1}): 3300 (O-H), 3220 (N-H), 3050 (aromatic C-H), 1620-1560 (C=C and C=N), 1220 (C=S), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.60 (s, 1H, pyrimidine C-H), 7.38-7.10 (m, 10H, Ar-H), 5.90 (s, 1H, O-H); 5.85, (s, 2H, N-H); 5.75 (s, 1H, C-H); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 148.80 (C=S, pyrimidine ring), 150.15-110.42 (aromatic C), 73.01 ppm (CH-OH). Anal. Calcd. for for $C_{17}H_{15}N_3OS$: C, 66.00; H, 4.89; N, 13.58; S, 10.36. Found: C, 66.17; H, 4.98; N, 13.40, S, 10.08.

1-Amino-5-[hydroxy(4-methylphenyl)methyl]-4-(methylphenyl)pyrimidine-2(1*H*)-one (3c): The obtained product **3c** was crystallized in EtOH and dried on P_2O_5 and was obtained in 64 % yield as a colourless mass; m.p. 247-248 °C. IR (KBr, ν_{max}, cm^{-1}): 3300 (O-H), 3220 (N-H), 3050 (aromatic C-H), 1620-1560 (C=C and C=N), 1220 (C=S), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.55 (s, 1H, pyrimidine C-H), 7.38-7.10 (m, 8H, Ar-H), 5.90 (s, 1H, O-H); 5.85, (s, 2H, N-H); 5.75 (s, 1H, C-H); 2.27 (s, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 151.80 (C=O, pyrimidine ring), 152.15-109.42 (aromatic C), 73.04 ppm (CH-OH), 21.44 ppm (2 × $\underline{C}H_3C_6H_4$-). Anal. Calcd. for for $C_{19}H_{19}N_3O_2$: C, 71.01; H, 5.96; N, 13.08. Found: C, 71.20; H, 5.80; N, 13.21.

1-Amino-5-[hydroxy(4-methylphenyl)methyl]-4-(methylphenyl)pyrimidine-2(1*H*)-thione (3d): The obtained product **3d** was crystallized in EtOH and dried on P_2O_5 and was obtained in 69 % yield as a yellow mass; m.p. 222-223 °C. IR (KBr, ν_{max}, cm^{-1}): 3300 (O-H), 3221 (N-H), 3040 (aromatic C-H), 1620-1560 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.40 (s, 1H, pyrimidine C-H), 7.48-7.05 (m, 8H, Ar-H), 5.80 (s, 1H, O-H); 5.85, (s, 2H, N-H); 5.74 (s, 1H, C-H); 2.27 (s, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 170.80 (C=S, pyrimidine ring), 152.15-109.42 (aromatic C), 73.12 ppm (CH-OH), 21.44 ppm (2 × $\underline{C}H_3C_6H_4$-). Anal. Calcd. for for $C_{19}H_{19}N_3OS$: C, 67.63; H, 5.68; N, 12.45; S, 9.50. Found: C, 67.50; H, 5.59; N, 12.32; S, 9.62.

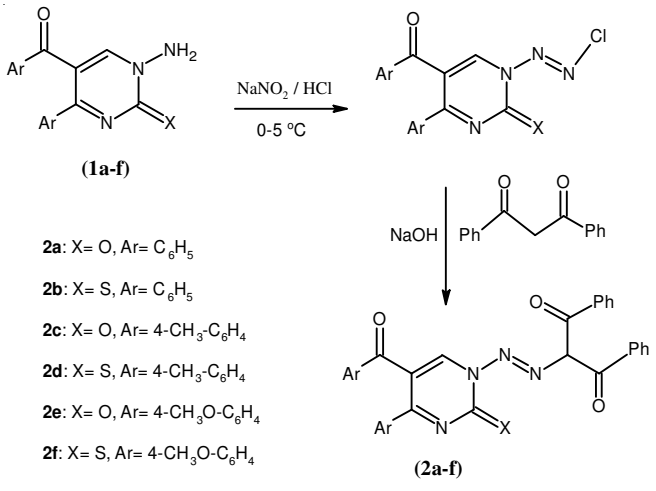
1-Amino-5-[hydroxy(4-methoxyphenyl)methyl]-4-(methoxyphenyl)pyrimidine-2(1*H*)-one (3e): The obtained product **3e** was crystallized in *n*-butanol and dried on P_2O_5 and was obtained in 58 % yield as a colourless mass; m.p. 261-262 °C. IR (KBr, ν_{max}, cm^{-1}): 3300 (O-H), 3221 (N-H), 3040 (aromatic C-H), 2940 (aliphatic C-H), 1620-1560 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.42 (s, 1H, pyrimidine C-H), 7.78-6.85 (m, 8H, Ar-H), 5.81 (s, 1H, O-H); 5.79, (s, 2H, N-H); 5.69 (s, 1H, C-H); 3.89, 3.87 (d, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 152.80 (C=O, pyrimidine ring), 146.15-110.22 (aromatic C), 75.07 (CH-OH), 54.64 (2 × $CH_3OC_6H_4$-). Anal. Calcd. for for $C_{19}H_{19}N_3O_4$: C, 64.58; H, 5.42; N, 11.89. Found: C, 64.45; H, 5.60; N, 11.70.

1-Amino-5-[hydroxy(4-methoxyphenyl)methyl]-4-(methoxyphenyl)pyrimidine-2(1*H*)-thione (3f): The obtained product **3f** was crystallized in *n*-butanol and dried on P_2O_5 and was obtained in 54 % yield as a colourless mass; m.p. 268-269 °C. IR (KBr, ν_{max}, cm^{-1}): 3300 (O-H), 3220 (N-H), 3030 (aromatic C-H), 2930 (aliphatic C-H), 1620-1560 (C=C and C=N), 800-640 (pyrimidine ring); ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.42 (s, 1H, pyrimidine C-H), 7.76-6.95 (m, 8H, Ar-H), 5.84 (s, 1H, O-H), 5.75, (s, 2H, N-H); 5.69 (s, 1H, C-H), 3.89, 3.87 (d, 6H, 2 × CH_3); ^{13}C NMR (400 MHz, (DMSO-d_6): δ = 170.40 (C=S, pyrimidine ring), 147.15-111.40 (aromatic C), 75.07 (CH-OH), 54.64 (2 × $\underline{C}H_3OC_6H_4$-). Anal. Calcd. for $C_{19}H_{19}N_3O_3S$: C, 61.77; H, 5.18; N, 11.37; S, 8.68. Found: C, 61.59; H, 5.02; N, 11.18; S, 8.80.

RESULTS AND DISCUSSION

Acetophenone semicarbazone and acetophenone thiosemicarbazone reacted with furan-2,3-diones and 1-methylenaminopyrimidine derivatives were synthesized. The *N*-aminopyrimidine derivatives (**1a-f**) were used as important materials in the synthesis of the target heterocycles (**Scheme-I**)[6-11]. Briefly, the reactions of **1a-f** were carried with 1,3-diketones and $NaBH_4$.

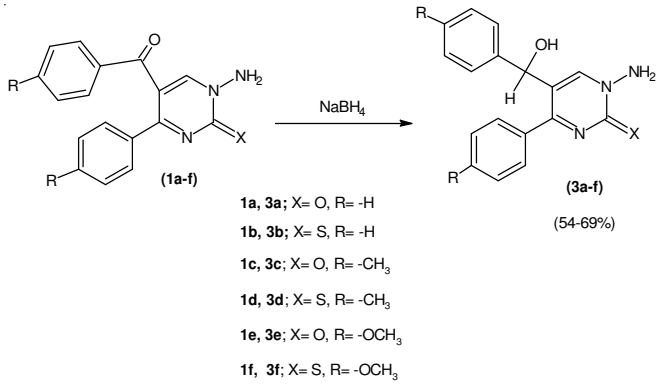
In this study, the diazonium salts of **1a-f** were condensed with dibenzoylmethane in the presence of NaOH with continuous stirring at 60-70 °C (**Scheme-II**). The reactions were monitored by thin-layer chromatography until complete consumption of the starting materials. After evaporation of the organic solvents and recrystallization from proper solvents (Experimental Section), **2a-f** were obtained in moderate yields (50-67 %). The products **2a-f** obtained were characterized by FT IR, elementary analysis, ^{1}H NMR and ^{13}C NMR. In the IR spectrum of compound **2a**, the C=O absorption peaks were seen at 1730, 1680 and 1650 cm^{-1}, respectively. In the ^{1}H NMR spectrum of compound **2a**, a peak observed at δ = 14.90 ppm was due to the enolic form. The ^{1}H NMR signals were found



Scheme-II: Synthesis of compounds **2a-f**

at δ = 7.36-7.02 (m, 20H, ArH). The ^{13}C NMR signals were detected at 190.80-188.10 (benzoyl groups, C=O), 153.15 (pyrimidine ring, C=O), 150.05-110.42 (aromatic C) and 74.82 ppm (CH). The elemental analysis data confirmed the structure of **2a**. The results of measurements of **2b-f** are given in the experimental section.

The reactions of *N*-aminopyrimidine derivatives (**1a-f**) with sodium borohydride led to the formation of the corresponding 5-[hydroxy(phenyl)methyl]-4-phenylpyrimidine-2(1*H*)-one derivatives (**3a-f**) in moderate yields (54-69 %) (**Scheme-III**). 1-Amino-5-[hydroxy(phenyl)methyl]-4-phenylpyrimidine-2(1*H*)-one (**3a**) was obtained in 54 % yield by treating **1a** with sodium borohydride and refluxing it in boiling ethanol for 1 h. In the FT-IR spectrum of compound **3a**, the OH and NH_2 absorption bands were found at 3300 and 3200 cm^{-1}. The absorption peak of the carbonyl group was observed at about 1650 cm^{-1}. The ^{1}H NMR signals were found at δ = 5.91 (s, 1H, OH), 5.80 (s, 2H, NH_2) and 5.74 (s, 1H, CH).The ^{1}H NMR signals of the aromatic rings were found at δ = 10.51 (s, 1H, pyrimidine ring.) and 7.38-7.02 ppm (m, 10 H, Ar-H). The ^{13}C NMR signals were observed at 152.80 (C=O, pyrimidine ring), 143.15-117.42 (aromatic C) and 73.08 ppm (CHOH). Finally, the elemental analysis data along with spectroscopic data confirmed the structure of compound **3a**. The other results of compounds **3b-f** are given in the experimental section.



Scheme-III: A Synthetic pathway for the preparation of compounds **3a-f**

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