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ONE POT SYNTHESIS OF DIHYDROPYRIMIDONES BY USING NICKEL-FERRITE NANOCATALYST

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ABSTRACT:
Dihydropyrimidone derivatives have been synthesized by grinding a Meldrum's acid, urea/thiourea with aryl aldehyde in the presence of Ni-ferrite nanocatalyst. The comparative studies were done with respect to the yield, reaction simplicity and work-up of the reaction. The structures of all the synthesized compounds were confirmed by chemical test along with TLC, physical parameters such as melting, boiling point and spectral technique such as IR, ^{1}H NMR and ^{13}C NMR.

KEYWORDS: Grinding, Nanocatalyst, Multicomponent.

INTRODUCTION:

Dihydropyrimidone derivatives have been studied for their pharmacological properties, making significance role in drug development. Substituted dihydropyrimidone show interesting biological properties like calcium channel blockers[i] and antihypertensive agents[ii]. Dihydrpyrimidones have been discovered to improve the health of cancer patients[iii]. Industrial importance of DHP and DHPM derivatives have developed the catalysts and techniques for their synthesis, such as sulfuric acid, fresh lemon juice, Ni nanoparticles (NPs), the use of solar thermal energy, ionic liquid, polymers and microwave radiation[iv-viii]. Derivatives of dihydropyrimidone have been reported to possess potent anti-ulcer activity[ix-xi].

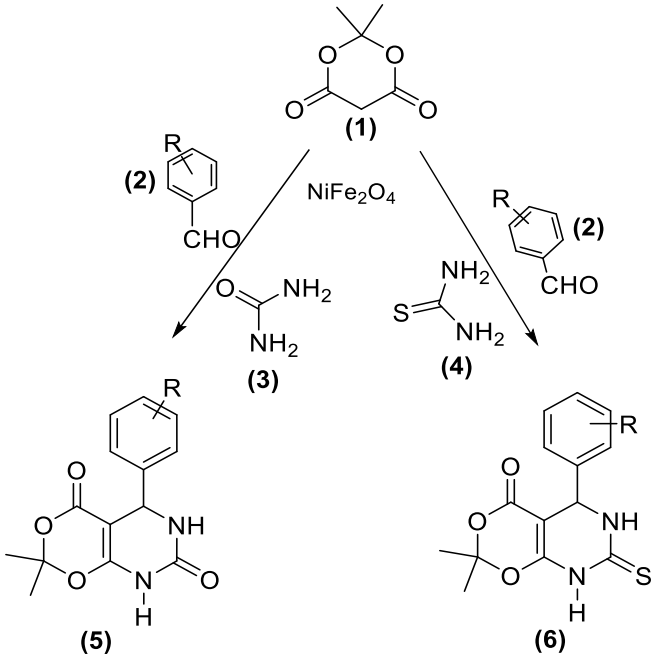
EXPERIMENTAL

Melting points of all synthesized compounds were determined in open capillary tubes and are uncorrected. The purity of the compounds was monitored by TLC. ^{1}H NMR spectra were recorded on Varian 600 MHz NMR spectrophotometer using $CDCl_3$/DMSO-d_6 as solvent and TMS as an internal standard (chemical shifts in δ ppm).

General Procedure:

Mixture of Meldrum's acid (0.01 mol), aromatic aldehydes (0.01 mol) and urea/thiourea (0.01 mol) were ground in presence of catalytic amount of Ni-ferrite nanocatalyst. The progress of the reaction was monitored by TLC. After completions of the reaction, the reaction mixture was poured onto ice-cold water, solid separated was filtered off, washed with water, dried and recrystallized from 25% ethanol to obtain pure compound.

General Scheme



Characterization data of compounds **5 (Urea derivative)** and **6 (Thiourea derivative)**

Compounds	R	Mol. Formula	m.p. °C	Yield %	
				Without Nano Catalyst	With Nano catalyst
5a	H	$C_{14}H_{14}N_20_4$	212-214	68	78
5b	4-Cl	$C_{14}H_{13}N_20_4Cl$	170-172	64	71
5c	4-OCH_3	$C_{15}H_{16}N_20_5$	130-132	77	85
6a	H	$C_{14}H_{14}N_20_3S$	200-202	69	80
6b	4-Cl	$C_{14}H_{13}N_20_3SCl$	156-158	80	87
6c	4-OCH_3	$C_{15}H_{16}N_20_4S$	120-122	74	82

Spectral Data:

Compound 5a: *7H,8H,10H-8,10-diaza-2,4-dioxa-5,9-dioxo-7-phenylbicyclo [4.4.0] dec-1(6)-ene*

IR (cm^{-1}): 1670(C=O), 1725(C=O), 3295(NH)**,**

^{1}H NMR(δ ppm): 1.71(6H,s,2xCH_3), 7.2 (5H,m,Ar- H), 8.15 (2H,s,2xNH), 8.25 (1H,s,CH),

^{13}C NMR(δ ppm): 27.325(2xCH_3), 105 and 110(tetrahedral carbon), 124.78 (CH), 116.36-131.7 (C=C,Ar-C),160.476 (C=O),165.985 (C=O).

Compound 5b: *7H,8H,10H-8,10-diaza-2,4-dioxa-5,9-dioxo-7-(4-chloro)phenylbicyclo [4.4.0] dec-1(6)-ene*

IR (cm^{-1}): 1670(C=O), 1725(C=O), 3295(NH)**,**

^{1}H NMR(δ ppm): 1.71(6H,s,2xCH_3), 7.2 (5H,m,Ar- H), 8.25 (2H,s,2xNH), 8.25 (1H,s,CH), ^{13}C NMR(δ ppm): 27.325(2xCH_3), 106 and 111(tetrahedral carbon), 124.78 (CH), 116.36-131.7 (C=C,Ar-C),160.476 (C=O),165.985 (C=O).

Compound 5c: *7H,8H,10H-8,10-diaza-2,4-dioxa-5,9-dioxo-7-(4-methoxy) phenylbicyclo [4.4.0] dec-1(6)-ene*
IR (cm^{-1}): 1660(C=O), 1730(C=O), 3290(NH)**,**
^{1}H NMR(δ ppm): 1.72(6H,s,2xCH_3), 3.87 (3H,s,OCH_3), 7.2 (4H,m,Ar- H), 8.2 (2H,s,2xNH), 8.3 (1H,s,CH),
^{13}C NMR(δ ppm): 27.325(2xCH_3), 55.75(OCH_3),105 and 110(tetrahedral carbon), 124.78 (CH), 116.36-131.7 (C=C,Ar-C),160.476 (C=O),165.985 (C=O).
Compound 6a: *7H,8H,10H-8,10-diaza-2,4-dioxa-5-oxo-9-thioxo-7-phenylbicyclo [4.4.0] dec-1(6)-ene*
IR (cm^{-1}): 1680 (C=O), 1240 (C=S), 3310(NH)**,**
^{1}H NMR(δ ppm): 1.72(6H,s,2xCH_3), , 7.0-7.15 (5H, m,Ar- H), 8.2 (2H,s,2×NH), 8.4(1H,s,CH), ^{13}C NMR(δ ppm):, 27.325(2xCH_3), 104.12 and 109(2x tetrahedral carbon), 120.53-130.788 (C=C,Ar-C), 156.982 (C=O), 163.512 (C=S).
Compound 6b: *7H,8H,10H-8,10-diaza-2,4-dioxa-5-oxo-9-thioxo-7(4-chloro)phenylbicyclo [4.4.0] dec-1(6)-ene*
IR (cm^{-1}): 1680 (C=O), 1240 (C=S), 3315(NH)**,**
^{1}H NMR(δ ppm): 1.72(6H,s,2xCH_3), , 7.0-7.15 (5H, m,Ar- H), 8.2 (2H,s,2×NH), 8.4(1H,s,CH), ^{13}C NMR(δ ppm):, 27.325(2xCH_3), 104.12 and 109(2x tetrahedral carbon), 120.53-130.788 (C=C,Ar-C), 156.982 (C=O), 163.512 (C=S).
Compound 6c: *7H,8H,10H-8,10-diaza-2,4-dioxa-5-oxo-9-thioxo-7(4-methoxy)phenylbicyclo [4.4.0] dec-1(6)-ene*
IR (cm^{-1}): 1680 (C=O), 1240 (C=S), 3310(NH)**,**
^{1}H NMR(δ ppm): 1.72(6H,s,2xCH_3), 3.70(3H,s, -OCH_3), 7.0-7.15 (5H, m,Ar- H), 8.2 (2H,s,2×NH), 8.4(1H,s,CH), ^{13}C NMR(δ ppm):, 27.325(2xCH_3), 56.75(-OCH_3), 104.12 and 109(2x tetrahedral carbon), 120.53-130.788 (C=C,Ar-C), 156.982 (C=O), 163.512 (C=S).

RESULTS AND DISCUSSION

The target molecules 7H,8H,10H-8,10-diaza-2,4-dioxa-5,9-dioxo-7-(substituted) phenylbicyclo [4.4.0] dec-1(6)-ene **(5) (a-c)** and 7H,8H,10H-8,10-diaza-2,4-dioxa-5-oxo-9-thioxo-7-(substituted) phenylbicyclo [4.4.0] dec-1(6)-ene **(6) (a-c)**,were synthesized in good yield by the multicomponent reaction of aromatic aldehydes, Meldrum's and urea/thiourea using catalytic amount of Ni-ferrite nanocatalyst.

CONCLUSIONS

Dihydropyrimidones were prepared in good yield by using multicomponent reaction. Use of Ni- ferrite nanocatalyst found to be very effective with high atom economy.

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