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1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU): as a highly efficient bicyclic amidine catalyst promoted solvent-free and one-pot synthesis of 1*H*-pyrazolo [1, 2-*b*] phthalazine-5,10-dione derivatives

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1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU) as a highly efficient bicyclic amidine catalyst promoted one-pot multi-component synthesis of biologically active 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives *via* one-pot four-component condensation reaction of phthalimide, hydrazine monohydrate, aryl aldehydes and malononitrile under solvent-free conditions through simple filter with no necessity of chromatographic purification steps. Use of safe, non-volatile, non-corrosive, highly efficient, readily available and easy to handle of catalyst, one-pot reaction, high yields and short reaction times, economical and convenient synthesis, solvent-free conditions and operational simplicity are among the other added advantages that make this approach an attractive alternative for the synthesis of these biologically active compounds.

Keywords: 1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU), Highly efficient bicyclic amidine catalyst, 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives, Solvent-free conditions, One-pot procedure.

Among the various nitrogen-containing heterocyclic compounds, 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives have received considerable attention due to their various biological and pharmacological activities[1, 2] such as anticancer[3], anti-inflammatory[4], anti micrbiological[5] and they have been reported to possess vasorelaxant[6], cardiotonic[7], anticonvulsant[8] and antifungal[9].

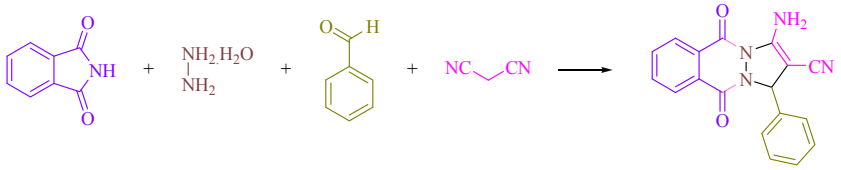
Between the known procedures for the synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives, the most straightforward method for synthesis of these systems involves a four-component tandem reaction of phthalimide/phthalic anhydrid, hydrazine monohydrate, aromatic aldehyde derivatives and malononitrile or three-component reaction of phthalhydrazide, aryl aldehyde derivatives and malononitrile utilizing a variety of homogeneous and heterogeneous catalysts, such as $Ce(SO4)2.4H_2O$[10], $SBA\text{-}Pr\text{-}SO_3H$[11], $InCl_3$[12], $NiCl_2.6H_2O$[13], [Bmim] OH[14], Ultrasound-assisted[15], P-TSA[16], STA[17], CuI nanoparticles[18], PTSA/[Bmim]Br[19] and TBBAD[20]. Although these protocols find certain merits of their own, still they suffer from a number of demerits such as relying on multi-step conditions, use of toxic organic solvents or catalysts containing transition metals, tedious work-up procedure, troublesome waste discarding, high reaction time, and low yields. Thus, a search for general, clean, efficient, feasible, and high yielding routes to this class of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives remains a valid exercise. Based on the above considerations and in continuation of our efforts to develop efficient methodologies[21-23] via multi-component reactions[24-28], finally, we have reported DBU as a cost-effective and easy to handle catalyst[29-31] for one-pot four-component condensation of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives under solvent-free conditions. We speculated that use of neutral organic bases that have high basicity, and can form a stable protonated species, may suppress the formation of enaminonitrile and other side products. 1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU) fulfills these requirements, and has been used in many organic transformations in recent years[32]. It is a sterically hindered amidine base and especially useful where side reactions due to the inherent nucleophilicity of basic nitrogen are a problem[33]. DBU is one of the strongest organic neutral base (pKa=12) and the +M effect of the adjacent nitrogen stabilizes the protonated species[29]. Furthermore, one of the source of environmental pollutions is the usage of organic

solvents under reflux conditions and the need for column chromatography to purity the products. In this present work, the products were obtained through simple filter with no need column chromatographic separation.

Results and Discussion

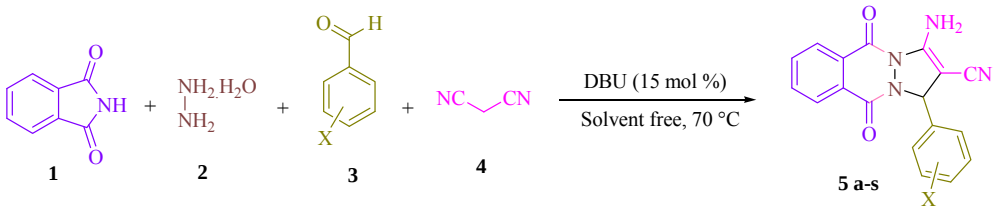
At beginning we performed four-component condensation of phthalimide (1.0 mmol), hydrazine monohydrate (1.0 mmol), benzaldehyde (1.0 mmol) and malononitrile (1.0 mmol) in the present of DBU (15 mol%) under solvent-free at 70°C, the product **5a** was found in 86%, which was confirmed by ^{1}H NMR spectroscopy. Encouraged by this result, we chosen this reaction as a model reaction to study the reaction conditions further for the synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives (**5a-s**). The catalyst plays an important role in the success of the reaction in terms of rate of the reaction and yields. In order to optimize the reaction conditions, quantity of the catalyst required was determined. No product could be detected in the absence of the catalyst even after 12 h (Table I, entry 1). Then, 5 mol% DBU was used to perform the reaction. But it requires slightly long reaction time and low yields (Table I, entry 2). Therefore, the loading of catalyst was gradually increased from 5 mol% to 20 mol% (Table I). It was found that 15 mol% of DBU is optimal to carry out the reactions in a short duration (Table I, entry 4). The use of excess of catalyst did not alter either reaction time or yield of the product (Table I, entry 10). Thus, the use of 15 mol% DBU is ideal to achieve the desired product in high yields. We also investigated different temperatures for the model reaction (Table I). It was observed that fast reaction occurred on raising the temperature from rt to 80°C and the yield of preferred product increased significantly (Table I). We were satisfied to find that the reaction proceeded smoothly and almost complete conversion of reactants was observed at 70°C to afford the desired product (**5a**) in 86% yields within 2 h (Table I, entry 4). Further increase in the temperature did not affect the product yield (Table I, entry 9). Having optimized reaction conditions, we synthesized a series 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives via phthalimide (**1**, 1.0 mmol), hydrazine monohydrate (**2**, 1.0 mmol), aldehyde derivatives (**3**, 1.0 mmol) and malononitrile (**4**, 1.0 mmol) (**5a-s**) using 15 mol% DBU as the catalyst under solvent-free conditions at 70°C (Scheme I) and the results are summarized in Table II.

Table I — Optimization of the reaction condition [a]



Entry	DBU (mol %)	Temperature (^{0}C)	Time (h)	Isolated Yields (%)
1	Catalyst free	70	12	No product
2	5	70	6	43
3	10	70	3	69
4	15	70	2	86
5	15	rt	12	No product
6	15	40	6	32
7	15	50	4	51
8	15	60	3	72
9	15	80	2	86
10	20	70	2	87

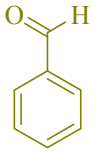
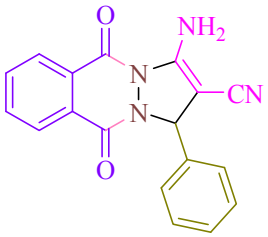
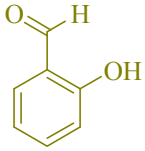
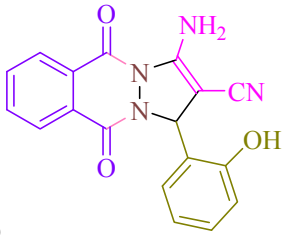
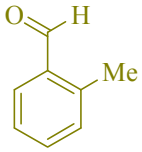
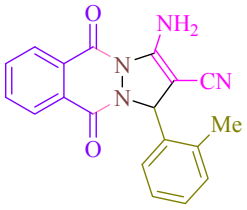
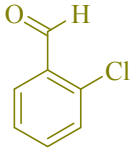
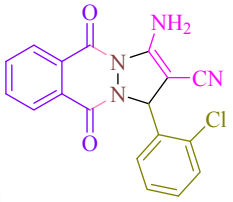
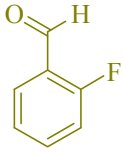
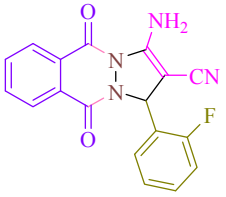
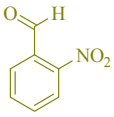
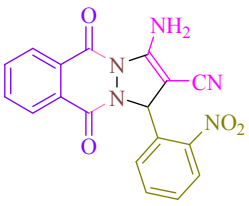
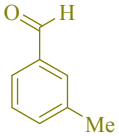
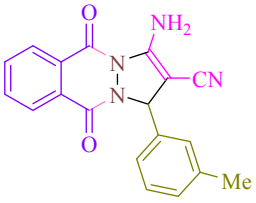
[a] Reaction conditions: phthalimide, hydrazine monohydrate, benzaldehyde and malononitrile (1:1:1:1) and DBU was heated at 70 °C for the appropriate time.



1 **2** **3** **4** **5 a-s**

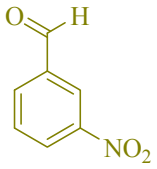
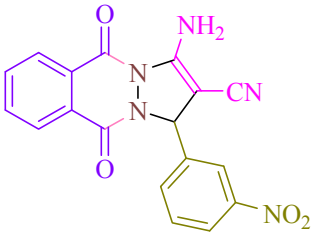
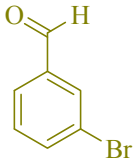
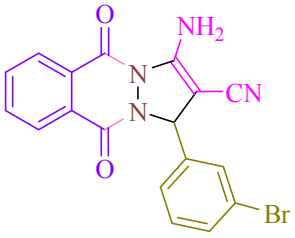
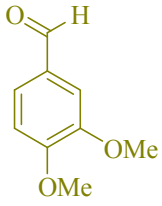
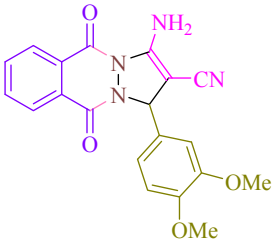
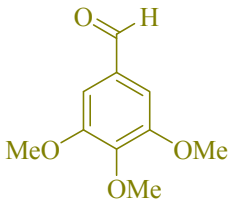
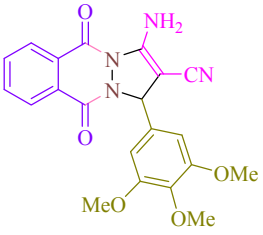
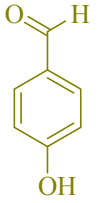
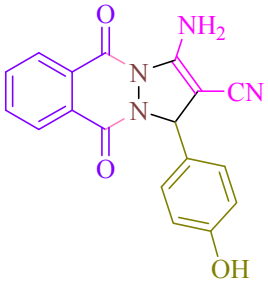
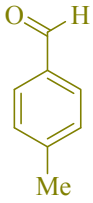
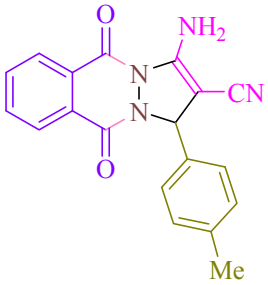
Scheme I — Synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives.

Table II — DBU-catalyzed synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives under solvent-free conditions.

Entry	Ar	Product	Time (h)	Isolated Yields (%)	m.p. °C	Lit. m.p. °C
1		 **5a**	2	86	272-274	270-272 [18]
2		 **5b**	3	78	210-212	212-214 [14]
3		 **5c**	2	92	247-249	248-250 [18]
4		 **5d**	3	80	257-259	257-259 [17]
5		 **5e**	2	90	269-271	268-270 [12]
6		 **5f**	2.5	86	266-268	265-266 [11]
7		 **5g**	2	89	251-253	250-252 [18]

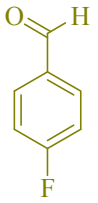
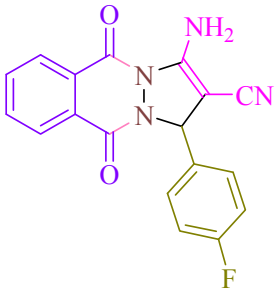
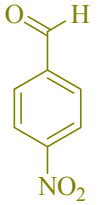
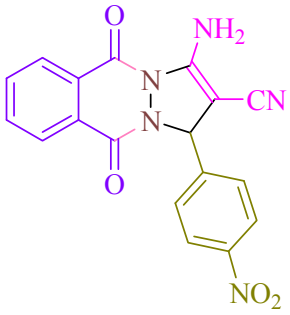
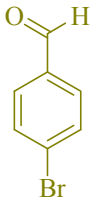
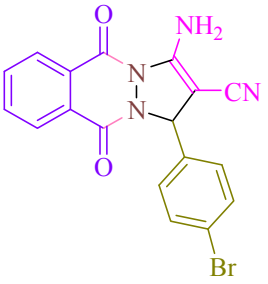
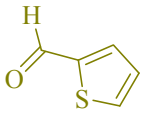
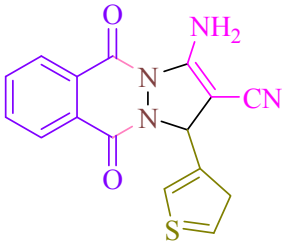
(*contd.*)

Table II — DBU-catalyzed synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives under solvent-free conditions. (*contd.*)

Entry	Ar	Product	Time (h)	Isolated Yields (%)	m.p. °C	Lit. m.p. °C
9		 **5i**	2.5	88	268-270	269-271 [19]
10		 **5j**	3.5	79	272-274	270-272 [11]
11		 **5k**	3	85	152-154	150-152 [20]
12		 **5l**	3	81	255-257	253-255 [12]
13		 **5m**	4	75	269-271	270-272 [12]
14		 **5n**	2	88	254-256	253-255 [18]

(*contd.*)

Table II — DBU-catalyzed synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives under solvent-free conditions.

Entry	Ar	Product	Time (h)	Isolated Yields (%)	m.p. °C	Lit. m.p. °C
16		 **5p**	2	90	264-266	263-265 [11]
17		 **5q**	3	87	226-228	228-229 [17]
18		 **5r**	4	81	266-268	265-267 [11]
19		 **5s**	3	82	246-248	244-246 [18]

Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 1*H*-pyrazolo[1,2-b]phthalazine-5,10-dione derivatives are shown in Table III. Table IV shows the comparison of ^{1}H NMR data. This study reveals that caffeine has shown its extraordinary potential to be an alternative inexpensive and highly efficient catalyst for synthesis of these biologically active nitrogen-containing heterocyclic compounds, in addition to the use of solvent-free conditions with excellent yield and short reaction times in the reaction are the notable advantages this present methodology.

Experimental Section

Melting points all compounds were determined using an Electro thermal 9100 apparatus. Also, nuclear magnetic resonance, ^{1}H NMR spectra were recorded on a Bruker DRX-400 Avance instruments with DMSO-d_6 as solvents. In this article, all reagents and solvents were purchased from Merck, Fluka and Acros chemical companies were used without further purification.

General procedure for preparation of 1-H-pyrazolo[1,2-b]phthalazine-5,10-dione derivatives (5a-s):

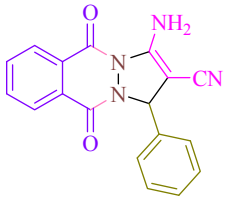
Table III — Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 1*H*-pyrazolo[1,2-b]phthalazine-5,10-dione derivatives [a]

Entry	Catalyst	Conditions	Time/Yield (%)	References
1	$InCl_3$	Water, Reflux	1.5h/85	[12]
2	$NiCl_2.6H_2O$	EtOH, Reflux	3h/87	[13]
3	*p*-TSA	[Bmim]Br, 100°C	3h/94	[16]
4	STA	Solvent-free, 70°C	20 min/94	[17]
5	CuI nanoparticles	MeCN, Reflux	27 min/91	[18]
6	TBBAD	Solvent-free, 80-100°C	15 min/89	[20]
7	DBU	Solvent-free, 70°C	2h/86	This work

[a] Based on the four-component reaction of benzaldehyde, phthalimide, hydrazine monohydrate and malononitrile.
Also ^{1}HNMR data of products have been compared with literature for synthesis of 1*H*-pyrazolo [1, 2-b] phthalazine-5, 10-dione derivatives are shown in Table IV.

A mixture of phthalimide (**1**, 1.0 mmol), hydrazine monohydrate (**2**, 1.0 mmol) and DBU (15 mol %) was heated for 2h at 70°C. Then aromatic aldehyde (**3**, 1.0 mmol) and malononitrile (**4**, 1.0 mmol) were added and the mixture was heated for the appropriate time. After completion of the reaction (by Thin layer chromatography TLC) the mixture was cooled to rt the solid products were filtered and then were be recrystallized from ethanol to give pure compounds (**5a-s**). Products have been characterized by melting points and ^{1}H NMR spectroscopy. Spectra data some of known products are represented below:

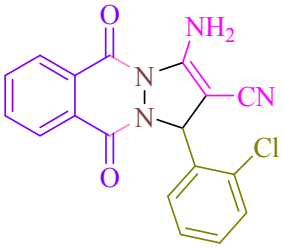
3-Amino-1-(phenyl)-5,10-dihydro-5,10-dioxo-1H-pyrazolo[1,2-b]phthalazine-carbonitrile (5a)



5a

Yield: 86%; m.p. 272-274°C; ^{1}H NMR (400 MHz, DMSO-d_6): 6.14 (1H, s, $H_{benzylic}$), 7.33-7.48 (5H, m, H_{Ar}), 7.97-8.29 (6H, m, NH_2 and H_{Ar}).

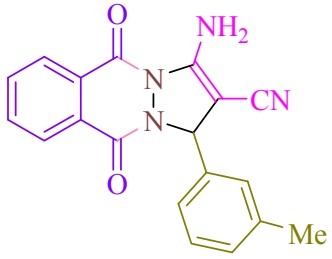
3-Amino-1-(2-chlorophenyl)-5,10-dihydro-5,10-dioxo-1H-pyrazolo[1,2-b]phthalazine-2-carbonitrile (5d)



5d

Yield: 80%; m.p. 257-259°C; ^{1}H NMR (400 MHz, DMSO-d_6): 6.47 (1H, s, $H_{benzylic}$), 7.39-7.65 (4H, m, H_{Ar}), 7.91-8.31 (6H, m, NH_2 and H_{Ar}).

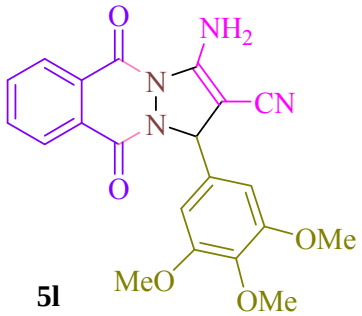
3-Amino-1-(3-methylphenyl)-5,10-dihydro-5,10-dioxo-1H-pyrazolo[1,2-b]phthalazine-2-carbonitrile (5 g)



5g

Yield: 89%; m.p. 251-253°C; ^{1}H NMR (400 MHz, DMSO-d_6): 2.30 (3H, s, CH_3), 6.08 (1H, s, $H_{benzylic}$), 7.14-7.26 (4H, m, H_{Ar}), 7.97-8.29 (6H, m, NH_2 and H_{Ar}).

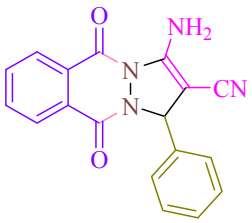
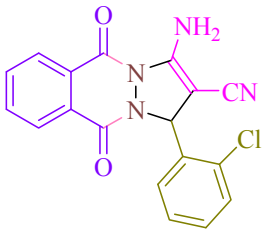
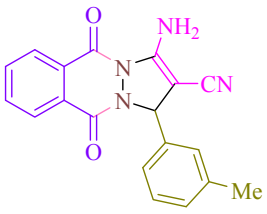
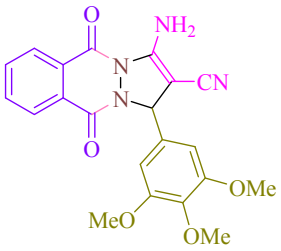
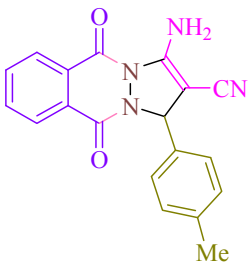
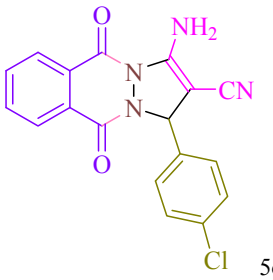
3-Amino-1-(3,4,5-trimethoxyphenyl)-5,10-dihydro-5,10-dioxo-1H-pyrazolo[1,2-b]phthalazine-2-carbonitrile (5l)



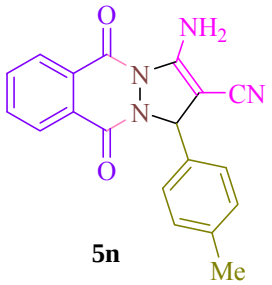
5l

Yield: 81%; m.p. 255-257°C; ^{1}H NMR (400 MHz, DMSO-d_6): 3.66 (3H, s, OCH_3), 3.76 (6H, s, 2×OCH_3), 6.07 (1H, s, $H_{benzylic}$), 6.78 (2H, s, H_{Ar}), 7.89- 8.29 (6H, m, NH_2 and H_{Ar}).

Table IV — Comparison of ^{1}HNMR data for synthesis of 1*H*-pyrazolo[1,2-*b*]phthalazine-5,10-dione derivatives.

Entry	Product	H Shift (found)	H Shift (lit)	References.
1	 5a	6.14 (1H, s, $H_{benzylic}$) 7.33-7.48 (5H, m, H_{Ar}) 7.97-8.29 (6H, m, NH_2 and H_{Ar})	6.12 (1H, s, $H_{benzylic}$) 7.29-7.47 (5H, m, H_{Ar}) 7.80-8.3 (6H, m, NH_2 and H_{Ar})	20
2	 5d	6.47 (1H, s, $H_{benzylic}$) 7.39-7.65(4H, m, H_{Ar}) 7.91-8.31 (6H, m, NH_2 and H_{Ar})	6.46 (1H, s, $H_{benzylic}$) 7.33-7.62 (4H, m, H_{Ar}) 7.87-8.30 (4H, m, H_{Ar}) 8.15 (2H, s, NH_2)	19
3	 5g	2.30 (3H, s, CH_3) 6.08 (1H, s, $H_{benzylic}$) 7.14-7.26 (4H, m, H_{Ar}) 7.97-8.29 (6H, m, NH_2 and ArH).	2.27 (3H, s, CH_3) 6.05 (1H, s, $H_{benzylic}$) 7.12-7.24 (4H, m, H_{Ar}) 7.96-8.26 (6H,m,Ar and NH_2)	18
4	 5l	3.66 (3H, s, OCH_3) 3.76 (6H, s, 2×OCH_3) 6.07 (1H, s, $H_{benzylic}$) 6.78 (2H, s, H_{Ar}) 7.89- 8.29 (6H, m, NH_2 and H_{Ar}).	3.64-3.73 (9H, s, OCH_3) 6.05 (1H, s, $H_{benzylic}$) 6.75 (2H, s, ArH) 7.94- 8.26 (6H, m, NH_2 and H_{Ar}).	20
5	 5n	6.15 (1H, s, $H_{benzylic}$) 7.43 (2H, d, J = 11.2 Hz, H_{Ar}) 7.54 (2H, d, J = 11.2 Hz, H_{Ar}) 7.88-8.28 (6H, m, NH_2 and H_{Ar})	6.14 (1H, s, $H_{benzylic}$) 7.39-7.52 (4H, m, H_{Ar}) 7.94-8.26 (6H, m, NH_2 and H_{Ar})	18
6	 5o	2.30 (3H, s, CH_3) 6.10 (1H, s, $H_{benzylic}$) 7.18 (2H, d, J = 8.0 Hz, H_{Ar}) 7.34 (2H, d, J = 8.0 Hz, H_{Ar}) 7.97-8.28 (6H, m, NH_2 and H_{Ar})	2.28 (3H, s, CH_3) 6.07 (1H, s, $H_{benzylic}$) 7.14-7.33 (4H, m, H_{Ar}) 7.94-8.25 (6H, m, NH_2 and H_{Ar})	18

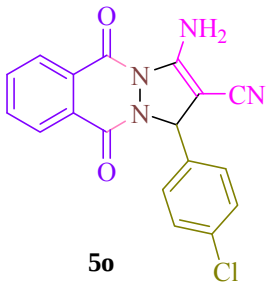
3-Amino-1-(4-methylphenyl)-5,10-dihydro-5,10-dioxo-1H-pyrazolo[1,2-b]phthalazine-2-carbonitrile (5n)



5n

Yield: 88%; m.p. 254-256°C; ^{1}H NMR (400 MHz, DMSO-d_6): 2.30 (3H, s, CH_3), 6.10 (1H, s, $H_{benzylic}$), 7.18 (2H, d, J = 8.0 Hz, H_{Ar}), 7.34 (2H, d, J = 8.0 Hz, H_{Ar}), 7.97-8.28 (6H, m, NH_2 and H_{Ar}).

3-Amino-1-(4-chlorophenyl)-5,10-dihydro-5,10-dioxo-1H-pyrazolo[1,2-b]phthalazine-2-carbonitrile (5o)



5o

Yield: 77%; m.p. 271-273°C; ^{1}H NMR (400 MHz, DMSO-d_6): 6.15 (1H, s, $H_{benzylic}$), 7.43 (2H, d, J = 11.2 Hz, H_{Ar}), 7.54 (2H, d, J = 11.2 Hz, H_{Ar}), 7.88-8.28 (6H, m, NH_2 and H_{Ar}).

Conclusion

In conclusion, Facile and efficient synthetic route for preparation of 1*H*-pyrazolo[1,2-*b*]phthalazine-5,10-dione derivatives catalyzed by DBU as a versatile, highly efficient bicyclic Amidine and easily available catalyst under solvent-free conditions was studied. This method presented is one-pot approach for the synthesis of these biologically active compounds with many merits in comparison with other reported results including easy-to-handle catalyst, short reaction times, excellent yields, facile reaction profiles and solvent-free conditions.

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