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SIMPLE AND EFFICIENT SYNTHESIS OF NEWBENZO[4,5]IMIDAZO[1,2-A]PYRIMIDINE DERIVATIVES USING ACETIC ACID AS CATALYST IN ETHANOL MEDIUM

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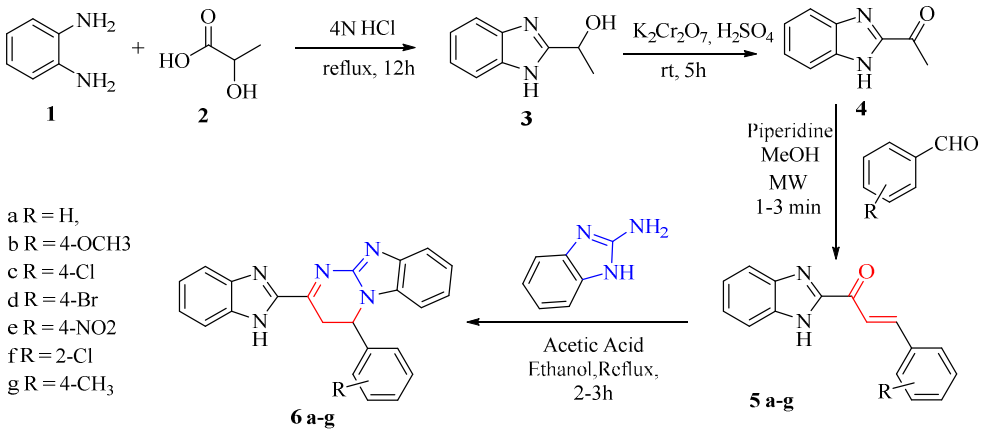
ABSTRACT
A series of new 2-(1H-benzo[d]imidazol-2-yl)-4-phenylbenzo[4,5]imidazo[1,2-a]pyrimidine derivatives**6a-g**were synthesized by simplecondensation reaction between 1-(1H-benzo[d]imidazol-2-yl)-3-phenylprop-2-en-1-one derivatives**5a-g** and 2-aminobenzimidazolein the presence of catalytic amount of acetic in in ethanol are heated under reflux for 2-3 hours . The yield of the synthesized compounds varied from 89-94%. The structures of the compounds obtained were characterized and confirmed by IR,^{1}H-NMR,^{13}C-NMR.

INTRODUCTION
During the last decades, chemists have been interesting on heterocyclic compounds and their various derivatives, especially the one that contains nitrogen, considering that it's excite in nucleic acids, vitamins, proteins and important molecular systems as well as their applications in pharmaceutical and chemical fields [i-iii]. Benzimidazole and pyrimidine are one of these heterocyclic aromatic organic compounds containing nitrogen [iv-v].
The benzimidazole ring system has been found to be an integral part of Vitamin-B12 and in the form of 5,6-dimethyl-1-(α-D-ribofuranosyl) benzimidazole [vi].The pyrimidine ring system has wide occurrence in nature as substituted and ring fused compounds and derivatives, including the nucleotides cytosine, thymine and uracil, thiamine (vitamin B1) and alloxan.[vii]
Over the past years, several Pyrimidobenzimidazoles derivatives have been synthesized and widely screened for their biological activities[viii-xi]. These classes of hetrocycles have found applications in diverse pharmacological areas such as anticancer,[xii]antiviral,[xiii-

[xiv]antmicrobial,[xv-xvi]antibacterial,[xvii]antifungal,[xviii-xix] anti-inflammatory,[xx]antiproliferativeactivity,[xxi] antihypertensive and hypotensive,[xxii] potential antineoplastic agents,[xxiii]antioxidant,[xxiv-xxv]antidiabetics, antibiotic, and antiarrythmic,[xxvi-xxvii]. Due to this wide range of biological and pharmaceutical activities and also industrial applications.Pyrimidobenzimidazoles moieties have received much attention in developing new therapeutic agents. The results of the synthesis of some new benzoimidazo[1,2-a]pyrimidine derivatives having are reported in this work.The structures of the compounds obtained were characterized and confirmed by IR,1HNMR,13C-NMR.

RESULTS AND DISCUSSION

In this work, A simple and practical method for the preparation of new 2-(1H-benzo[d]imidazol-2-yl)-4-phenylbenzo[4,5]imidazo[1,2-a]pyrimidine derivatives **6a-g**through the condensation reaction of 1-(1H-benzo[d]imidazol-2-yl)-3-phenylprop-2-en-1-onederivatives**5a-g**and 2-aminobenzimidazole**7** in the presence of catalytic amount of acetic in ethanol are heated under reflux for 2-3 hours**(Table 2)**.The reaction conditions were optimized by conducting the reaction model at different solvents such as methanol, water, ethanol, DMF, Butanol and also under solvent-free condition **(Table 1)**.The intermediates**3, 4** and **5** have been prepared under a method described previously[xxviii]. A Claisen-Schimdt condensation between 2-acetylbenzimidazole **4** and substituted aromatic aldehydes using the microwave conditions in the presence of a catalytic amount of piperidine leading to obtain the derivatives of 1-(1H-benzo[d]imidazol-2-yl)-3-phenylprop-2-en-1-one derivatives **5 a-g.**The precursor **3**was prepared by condensing the O-phenylenediamine **1** with lactic acid **2** in HCl 4N using the condensation of Phillip method[xxiv].An oxidation has been carried out on the precursor **3** by potassium dichromate in the sulfuric acid medium leading to the formation of 2-acetyl benzimidazole**4**.The synthetic pathway compounds are provided in **(Scheme 01)**.The purity of the synthesized compounds was monitored by TLC and the structures of all compounds were supported by spectral data. The IR, ^{1}H NMR and^{13}C-NMR are consistent with the proposed structures.



Scheme 01: synthetic route for the preparation of the new 2-(1*H*-benzo[*d*]imidazol-2-yl)-4-phenyl-3,4-dihydrobenzo[4,5]imidazo[1,2-*a*]pyrimidine **6 a-g**

Table 1: The synthesis of **6a-g** compounds by using different solvents.

Entry	**Catalyst (0.5eq)**	**Solvent**	**Time (min)**	**Temp (°C)**	**Yield[a] (%)**
1	AcOH	Free Solvent	24	80	75
2	AcOH	***Ethanol***	***3h***	***Reflux***	***94***
3	AcOH	Methanol	5	Reflux	85
4	AcOH	Chloroform	24	Reflux	-
5	AcOH	DMF	12	Reflux	67
6	AcOH	Butanol	24	Reflux	81

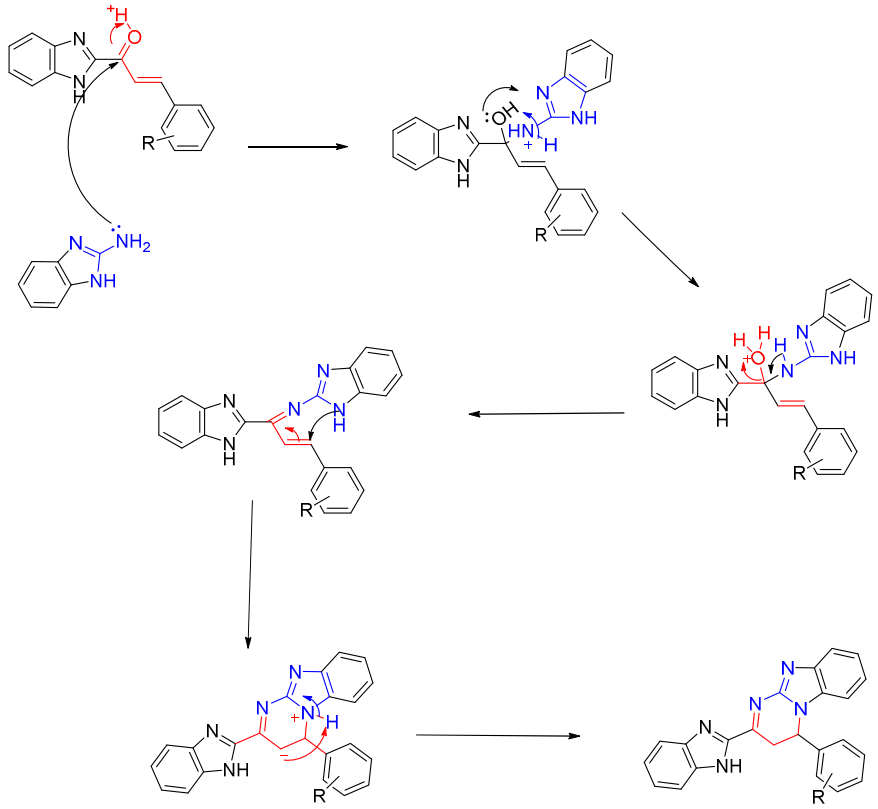
Table 3: Synthesis of products **9a-g**by the reactions of 5a-g with 2-aminobenzimidazole in the presence of AcOH (0.52mmol), at Reflux in Ethanol.

	R	Product	Time (h)	Yield[b] (%)	M.P (°C)
5a	H	**6a**	3	**92**	234–236
5	4-OCH_3	**6b**	3	**94**	222–224
3	4-ClC_6H_5	**6c**	2.5	**93**	236–237
4	4-Br	**6d**	2	**92**	226-227
5	4-NO_2	**6e**	3	**91**	232–233
6	2-Cl	**6f**	2	**93**	224–225
7	4-CH_3	**6g**	1.5	**89**	245–246

a. Reaction conditions: **5a** (1 mmol), 2-aminobenzimidazole (1 mmol) in the presence of AcOH (0.52mmol), at Reflux in Ethanol.
b. Isolated yield

In IR spectra, an absorption band 3248-3291 cm^{-1} showing the existence of the NH bond in all compounds**6a-g**which.The C=N stretching absorption for benzimidazole compounds appeared nearly around 1 cm-1 ,and absorption region of the C=C stretching at 1470 cm–1,The absorption bands of α,β-unsaturated carbonyl system was absent in the spectra of compounds **6a-g** due to the reaction with 2-aminobenzimidazole.
In the result of ^{1}HRMN ,the signal of NH displayed at ~11.5 ppm in the spectra of compounds**6a-g**, corresponding at benzimidazole and we observed two peaks, the first as a triplet at 5.08 for the CH of pyrimidine ring and the second represented as doublets at 2.9 for CH2 of pyrimidol[1,2-a]benzimidazole. The protons of α,β-unsaturated system was absent due to the reaction with 2-aminobenzimidazole. Other protons appeared in the expected region.

^{13}C NMR confirmed the suggested structure of compounds **6a-g**, by the absent of signals of carbonyl carbon atoms of the chalcone fragment, while the apperencetwo signals of carbonsof thepyrimidol[1,2-a]benzimidazole in the products of **6 a-g**, the first at ~34 ppm was assigned to CH_2 group and the scond at ~55 ppmwas assigned to CH.



Scheme 02: Mechanism propozed for the synthesis of the 2-(1*H*-benzo[*d*]imidazol-2-yl)-4-phenyl-3,4-dihydrobenzo[4,5]imidazo[1,2-*a*]pyrimidine **6 a-g**

CONCLUSION

In summary, using simple condensationreaction between 1-(1H-benzo[d]imidazol-2-yl)-3-phenylprop-2-en-1-one derivatives and 2-aminobenzimidazole in the presence of catalytic amount of acetic in ethanol are heated under reflux , Series of 2-(1H-benzo[d]imidazol-2-yl)-4-phenylbenzo[4,5]imidazo[1,2-a]pyrimidine derivatives were synthetized. The synthesized compounds were obtained in excellent yields and identified by melting point and characterized by IR, ^{1}H NMR and ^{13}C RMN.

EXPERIMENTAL SECTION

Most of the materials used in this work are Sigma Aldrich brand commercial products and have been used without further purification.The melting points were taken by using a Kofler bench melting point apparatus. The purity of the synthesized all coumponds was checked by TLC using silica gel-60 F 254 aluminium sheets using Ethyl acetate: Petrolium ether (3:7) as eluent and visualized in a ultra violet chamber. IR spectra were recorded on ALPHA's Platinum ATR single reflection diamond ATR spectrophotometer. The ^{1}HNMR and ^{13}CNMR spectras were recorded on a Bruker AC 300 MHZ FTNMR spectrophotometer in $CDCl_3$ and

DMSO.Chemical shift were recorded in parts per million (ppm) downfield from TMS as an internal reference and the following multiplicity abbreviations were used: s, singlet; d, doublet; t, triplet; q, quadruplet; m, multiplet. All chemicals were obtained from Merck and were used without further purification.

General procedure for synthesis of 1-(1H-benzo[d]imidazol-2-yl)-3-phenylprop-2-en-1-one derivatives (5a-g): A solution of 2-acetyl-benzimidazole (3.2g, 20 mmol) and the diversely substituted aromatic aldehyde (24mmol) in 40 ml of absolute methanol were taken in a flask. 0.2mmol of Piperidine was added and the reaction mixture was irradiated in a domestic microwave oven for 1 minute to 3 minutes at the power of 360 watts, the reaction is followed by thin layer chromatography using silica gel. the reaction mixture cooled to room temperature.Then poured onto crushed ice water. The product obtained is filtered and washed with distilled water. The compounds obtained recrystallized in suitable solvent[xxviii].

1-(1H-benzo[d]imidazol-2-yl)-3-phenylprop-2-en-1-one (5a). Yield (93%). Pale-yellow crystals. Mp 196–197°C. IR spectrum, ν, cm^{-1}: 3241 (N–H), 1656 (C=O), 1592(C=N).^{1}H NMR (300 MHz, DMSO-d_6) δ 13.49 (s, 1H, NH), 8.13 (d, J = 16.1 Hz, 1H), 7.98 (d, J = 16.1 Hz, 1H), 7.86 (d, J = 2.9 Hz, 2H), 7.73 (s, 2H), 7.49 (d, J = 3.0 Hz, 3H), 7.37 (dd, J = 6.0, 3.0 Hz, 2H).^{13}C NMR (75 MHz, DMSO-d_6) δ 180.96, 148.98, 144.21, 134.29, 131.06, 129.11, 128.88, 124.42, 121.58.

1-(1H-benzo[d]imidazol-2-yl)-3-(4-methoxyphenyl) prop-2-en-1-one (5b): Yield (83%). Yellow crystals. Mp 185–187°C IR spectrum, ν, cm^{-1}: 3253 (N–H), 1651 (C=O),1573 (C=N), ^{1}H NMR (300 MHz,) δ 13.48 (s, 1H, NH); 8.03 (d, J = 15.9, 1H); 7.96 (d, J = 15.9, 1H); 7.76 (t, J = 14.5 Hz, 2H), 7.59 (d, J = 7.3 Hz, 1H), 7.55 – 7.42 (m, 2H), 7.38 (d, J = 2.9 Hz, 1H), 7.36 (d, J = 3.0 Hz, 1H). 3.79 (s, 3H). ^{13}C NMR (75 MHz, DMSO-d6) δ 181.48, 162.30, 149.11, 145.93, 143.72, 133.85, 131.11, 127.41, 126.29, 123.74, 121.84, 118.63, 114.51, 112.15, 55.45.

1-(1H-benzo[d]imidazol-2-yl)-3-(4-chlorophenyl) prop-2-en-1-one (5c) Yield (89%). Yellow crystals. Mp 201–202°C. IR spectrum, ν, cm^{-1}: 3261 (N–H), 1660 (C=O), 1594(C=N). ^{1}H NMR (300 MHz, DMSO-d_6) δ 13.48 (s, 1H), 8.12 (d, J = 16.1 Hz, 1H), 7.94 (d, J = 16.2 Hz, 1H), 7.87 (d, J = 8.4 Hz, 2H), 7.72 (s, 2H), 7.51 (d, J = 8.4 Hz, 2H), 7.36 (d, J = 3.1 Hz, 1H), 7.34 (d, J = 3.1 Hz, 1H).^{13}C NMR (75 MHz, DMSO-d_6) δ 181.07, 149.14, 142.88, 138.36, 135.83, 133.47, 130.72, 129.34, 124.63, 122.50.

1-(1H-benzo[d]imidazol-2-yl)-3-(4-bromophenyl) prop-2-en-1-one (5d). Yield (84%). Yellow crystals. Mp 222– 223°C. IR spectrum, ν, cm^{-1}: 3249 (N–H), 1659 (C=O), 1596 (C=N).^{1}H NMR (300 MHz, DMSO-d_6) δ 13.44 (s, 1H, NH), 8.14 (d, J = 16.1 Hz, 1H), 7.93 (d, J = 16.1 Hz, 1H), 7.83 (d, J = 8.3 Hz, 2H), 7.72 (s, 2H), 7.67 (d, J = 8.3 Hz, 2H), 7.38 (d, J = 3.0 Hz, 1H), 7.36 (d, J = 3.1 Hz, 1H).^{13}C NMR (75 MHz, DMSO-d_6) δ 181.42, 149.47, 143.34, 134.52, 134.13, 132.64, 132.49, 131.31, 130.94, 126.82, 125.01, 122.90.

1-(1H-benzo[d]imidazol-2-yl)-3-(4-nitrophenyl) prop-2-en-1-one (5e): Yield (91%). Yellow crystals. Mp 204–205°C . IR spectrum, ν, cm^{-1}: 3267 (N–H), 1668 (C=O), 1591 (C=N).^{1}H NMR (300 MHz, DMSO-d_6) δ 13.78 (s, 1H, NH), 8.70 (d, J = 16.3 Hz, 1H), 8.13 (d, J = 16.2 Hz, 1H), 8.10 (s, 1H), 7.76 (t, J = 14.5 Hz, 2H), 7.59 (d, J = 7.3 Hz, 1H), 7.55 – 7.42 (m, 2H), 7.38 (d, J = 2.9 Hz, 1H), 7.36 (d, J = 3.0 Hz, 1H). ^{13}C NMR (75 MHz, DMSO-d_6) δ 181.46, 149.49, 138.31, 134.16, 132.05, 131.66, 129.88, 128.16, 127.66, 124.21, 124.13.

1-(1H-benzo[d]imidazol-2-yl)-3-(2-chlorophenyl) prop-2-en-1-one (5f): Yield (93%). Yellow crystals. Mp 204–205°C . IR spectrum, ν, cm^{-1}: 3267 (N–H), 1668 (C=O), 1591 (C=N).^{1}H NMR (300 MHz, DMSO-d_6) δ 13.52 (s, 1H, NH), 8.26 (d, J = 16.1 Hz, 1H), 8.15 (d, J = 18.1 Hz, 1H), 8.10 (s, 1H), 7.76 (t, J = 14.5 Hz, 2H), 7.59 (d, J = 7.3 Hz, 1H), 7.55 – 7.42 (m, 2H), 7.38 (d, J = 2.9 Hz, 1H), 7.36 (d, J = 3.0 Hz, 1H).^{13}C NMR (75 MHz, DMSO-

d_6) δ 180.46, 148.49, 138.41, 134.26, 132.05, 131.66, 129.88, 128.16, 127.66, 124.26, 124.07.

1-(1H-benzo[d]imidazol-2-yl)-3-(p-tolyl) prop-2-en-1-one(5g). Yield (86%). Yellow crystals. Mp 195–196°C. IR spectrum, ν, cm^{-1}: 3251 (N–H), 1653 (C=O),1577 (C=N), ^{1}H NMR (300 MHz, DMSO-d6) δ 13.54 (s,1H, NH); 8.30 (d, J = 16,1H,); 7.96 (d, J = 16,1H); 7.76 (t, J = 14.5 Hz, 2H), 7.59 (d, J = 7.3 Hz, 1H), 7.55 – 7.42 (m, 2H), 7.38 (d, J = 2.9 Hz, 1H), 7.36 (d, J = 3.0 Hz, 1H). 3.83 (s, 3H). ^{13}C NMR (75 MHz, DMSO-d6) δ 182.38, 161.30, 149.15, 146.73, 141.72, 133.85, 131.11, 127.41, 126.29, 123.74, 121.84, 118.63, 114.51, 112.15, 55.45.

General procedure for synthesis 2-(1H-benzo[d]imidazol-2-yl)-4-phenylbenzo[4,5]imidazo [1,2-a]pyrimidine (6 a-g):2-amino benzimidazole (1 mmol) and**5 a-g** compounds (1 mmol) were taken in ethanol in a 50 mL round bottomed flask and stirred for 5 min. AcOH (0.5mmol) was added and the reaction mixture was stirred at reflux for 2-3 h. The progress of the reaction was monitored by TLC. After completion of the reaction, the crude mixture was partitionedbetween H_2O and EtOAc. The aqueous layer was extracted with EtOAc (3×15 mL). The combined organic phases were dried ($MgSO_4$) and concentrated under reduced pressure. The compounds obtained recrystallized in ethanol absolute.

2-(1H-benzo[d]imidazol-2-yl)-4-phenylbenzo[4,5]imidazo[1,2-a]pyrimidine (6a): Yield (92%).yellowish green solid. Mp234–236°C.IR spectrum, ν, cm–1: 3267 (N–H), 1591 (C=N).^{1}H NMR (300 MHz, DMSO-d6)δ 11.06 (s, 1H), 8.13 – 8.04 (m, 1H), 7.74 (dd, J = 1.5, 7.5 Hz, 1H), 7.64 – 7.37 (m, 3H), 7.36 – 7.24 (m, 4H), 7.28 – 7.20 (m, 2H), 7.24 – 7.11 (m, 1H), 7.17 – 7.03 (m, 1H), 7.04 (dtd, J = 0.8, 1.6, 6.5 Hz, 1H), 5.08 (t, J = 0.6, 9.1 Hz, 1H), 2.77 (d, J = 9.3 Hz, 2H).^{13}C NMR (75 MHz, DMSO-d_6)δ 161.43, 150.69, 145.06, 142.41, 141.49, 139.80, 137.58, 135.30, 128.57, 127.79, 127.57, 125.08, 121.43, 121.31, 121.17, 118.59, 117.11, 113.49, 111.10, 53.59, 32.71.

2-(1H-benzo[d]imidazol-2-yl)-4-(4-methoxyphenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (6b) : Yield (94%). yellowish grey solid. Mp222–224°C.. IR spectrum, ν, cm–1: 3235 (N–H), 1598(C=N). ^{1}H NMR (300 MHz, DMSO-d6) δ 11.03 (s, 1H), 7.68 – 7.59 (m, 1H), 7.64 – 7.52 (m, 3H), 7.35 – 7.17 (m, 6H), 6.94 – 6.84 (m, 2H), 5.00 (t, J = 0.6, 9.2 Hz, 1H), 3.81 (s, 3H), 3.02 (d, J = 9.3 Hz, 2H^{13}C NMR (75 MHz, DMSO)δ 161.43, 158.29, 150.69, 145.06, 142.41, 141.49, 137.58, 135.30, 134.78, 128.75, 125.08, 121.43, 121.31, 121.17, 118.59, 117.11, 114.14, 113.49, 111.10, 55.40, 53.56, 32.13.

2-(1H-benzo[d]imidazol-2-yl)-4-(4-chlorophenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (6c) : Yield (93%). yellowish green solid. Mp236–237°C.IR spectrum, ν, cm–1: 3259 (N–H), 1589 (C=N). ^{1}H NMR (300 MHz, DMSO-d6) δ 11.18 (s, 1H), 7.86 – 7.77 (m, 1H), 7.77 – 7.69 (m, 1H), 7.58 (dd, J = 3.5, 5.8 Hz, 2H), 7.40 – 7.11 (m, 7H), 7.04 (ddq, J = 0.7, 1.5, 7.6 Hz, 1H), 5.16 – 5.03 (m, 1H), 2.92 (d, J = 9.3 Hz, 2H).^{13}C NMR (75 MHz, DMSO-d_6) δ 161.43, 150.69, 145.06, 142.41, 141.49, 138.97, 137.58, 135.30, 130.25, 129.25, 128.53, 125.08, 121.43, 121.31, 121.17, 118.59, 117.11, 113.49, 111.10, 55.28, 34.49.

2-(1H-benzo[d]imidazol-2-yl)-4-(4-bromophenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (6d) : Yield (92%). yellowish green solid. Mp226-227°C.IR spectrum, ν, cm–1: 3271 (N–H), 1584 (C=N). ^{1}H NMR(300 MHz, DMSO-d6) δ 11.08 (s, 1H), 7.97 – 7.89 (m, 1H), 7.81 – 7.72 (m, 1H), 7.68 – 7.52 (m, 3H), 7.47 (dd, J = 1.5, 7.2 Hz, 1H), 7.35 – 7.22 (m, 3H), 7.21 (ddd, J = 1.7, 7.3, 7.8 Hz, 1H), 7.15 – 7.06 (m, 2H), 5.00 (td, J = 0.6, 9.2 Hz, 1H), 3.10 (d, J = 9.3 Hz, 2H).^{13}C NMR (75 MHz, DMSO-d_6) δ 161.43, 150.69, 145.06, 142.41, 141.49, 138.28, 137.58, 135.30, 130.14, 126.69, 125.08, 121.92, 121.43, 121.31, 121.17, 118.59, 117.11, 113.49, 111.10, 54.96, 34.24.

2-(1H-benzo[d]imidazol-2-yl)-4-(4-nitrophenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (6e) : Yield (91%). yellowish green solid. Mp 232–233°C. ^{1}H NMR (300 MHz, DMSO-d6) δ 11.11 (s, 1H), 8.09 (dd, J = 1.5, 7.6 Hz, 1H), 8.00 (dd, J = 1.5, 7.6 Hz, 1H), 7.72 – 7.17 (m, 11H), 5.00 (tt, J = 0.6, 9.2 Hz, 1H), 2.91 (d, J = 9.3 Hz, 2H).^{13}C NMR (75 MHz,DMSO-d_6)δ 161.43, 150.69, 146.79, 145.06, 143.06, 142.41, 141.49, 137.58, 135.30, 126.32, 125.08, 124.79, 121.43, 121.31, 121.17, 118.59, 117.11, 113.49, 111.10, 55.75, 33.78

2-(1H-benzo[d]imidazol-2-yl)-4-(2-chlorophenyl)benzo[4,5]imidazo[1,2-a]pyrimidine (6f) : Yield (93%). yellowish brawn solid. Mp224–225°C.IR spectrum, ν, cm–1: 3261 (N–H), 1575 (C=N). ^{1}H NMR (300 MHz, DMSO-d6)δ 11.05 (s, 1H), 7.68 – 7.59 (m, 1H), 7.63 – 7.54 (m, 3H), 7.59 – 7.53 (m, 1H), 7.46 (td, J = 1.4, 7.7 Hz, 1H), 7.35 – 7.04 (m, 5H), 6.99 (ddd, J = 1.6, 6.7, 8.0 Hz, 1H), 5.01 (td, J = 0.6, 9.7 Hz, 1H), 2.76 (d, J = 9.6 Hz, 2H).^{13}C NMR (75 MHz, DMSO-d6) δ 161.43, 150.69, 145.06, 142.41, 141.49, 140.83, 137.58, 135.30, 134.74, 131.03, 130.16, 128.79, 126.09, 125.08, 121.43, 121.31, 121.17, 118.59, 117.11, 113.49, 111.10, 53.99, 33.56.

2-(1H-benzo[d]imidazol-2-yl)-4-(p-tolyl)benzo[4,5]imidazo[1,2-a]pyrimidine (6g) : Yield (89%). yellowish brawn solid.Mp245–246°C.IR spectrum, ν, cm–1: 3263(N–H), 1585(C=N). ^{1}H NMR (300 MHz, DMSO-d6) δ 11.08 (s, 1H), 7.68 – 7.52 (m, 4H), 7.45 – 7.24 (m, 3H), 7.29 – 7.02 (m, 6H), 5.09(t, J = 0.6, 9.1 Hz, 1H), 2.88 (d, J = 9.3 Hz, 2H), 2.30 (s, 3H).^{13}C NMR (75 MHz, DMSO-d_6)δ 161.43, 150.69, 145.06, 142.41, 141.49, 138.80, 137.58, 136.92, 135.30, 129.66, 126.09, 125.08, 121.43, 121.31, 121.17, 118.59, 117.11, 113.49, 111.10, 54.96, 34.80, 18.97.

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