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MICROWAVE ASSISTED SYNTHESIS AND CHARACTERIZATION OF SOME 2-SUBSTITUTED-4,5-DIPHENYLIMIDAZOLE DERIVATIVES

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ABSTRACT

A simple and highly efficient method for a three-component condensation of benzil, aldehydes and ammonium acetate under microwave irradiation in the presence of glacial acetic acid in solvent-free condition was carried out to afford the corresponding 2-substituted-4,5-diphenylimidazole derivatives in high yields. The remarkable advantages offered by this method are inexpensive, simple procedure, much faster (1-4 min) reactions and high yield of products.

Key words: 2-Substituted 4,5-diphenylimidazoles, Solvent-free, Microwave irradiation.

INTRODUCTION

The imidazole ring system is of particular interest as it is a component of histidine that produces histamine in metabolic process. The potency and wide applicability of the imidazole pharmacophore can be attributed to its hydrogen bond donor-acceptor capability as well as its high affinity for metals, which are present in many protein active sites (e.g. Zn, Fe, Mg). Among these imidazoles, 2,4,5-triphenylimidazoles can be used as light-sensitive materials in photography and known as inhibitors, fungicides and herbicides, plant growth regulators, anti-inflammatory, antithrombotic, therapeutic agents, and so on.

EXPERIMENTAL

Methods

Equimolar amount of neat reactants benzil, aldehydes and excess of ammonium acetate on exposure to microwave irradiation for optimized time gave excellent yield

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(Table 1) of 2-substituted-4,5-diphenyl-1H-imidazoles **(G1-12)** after triturating with small quantity of silica gel. Silica gel as acidic support under microwave irradiation can accelerate this new cyclocondensation reaction by increasing the reactivity.

Melting points of the compounds were determined using melting point apparatus and were reported uncorrected. Thin layer chromatography was performed on precoated silica gel G plates using methanol/chloroform (9 : 1) as developing solvent system and spots were visualized by UV lamp/iodine vapors in a tightly closed chamber.

IR spectra were recorded in KBr pellets using a Schimadzu-8700 FT-IR spectrophotometer. The ^{1}H NMR spectra were recorded in DMSO-d_6 by Bruker AVIII NMR spectrometers using tetramethylsilane as an internal standard. The mass spectra were recorded by JEOL GC Mate II Mass spectrophotometer operating as direct probe using EI technique.

Table 1: Reaction times and yields for compounds

Compd.	R_1	Melting point (^{0}C)	Reaction time (min)	Yield (%)
G1	4-Methoxyphenyl	82-84	3	90
G2	2-Nitrophenyl	160-162	3.5	88
G3	4-N,N-Dimethylphenyl	256-258	4	85
G4	4-Hydroxy-3-methoxyphenyl	252-253	3	92
G5	4- Hydroxy-3-ethoxyphenyl	255-256	3.5	90
G6	2- Hydroxyphenyl	85-86	3	94
G7	Furyl	170-173	3	82
G8	-CH=CH-C_6H_5	58-60	3	84
G9	Pthalyl	262-263	3	89
G10	3,4-Dimethoxyphenyl	158-160	4	86
G11	4-Methylphenyl	240-242	4	90
G12	4- Hydroxy-3,5-dimethoxyphenyl	260-261	3	96

RESULTS AND DISCUSSION

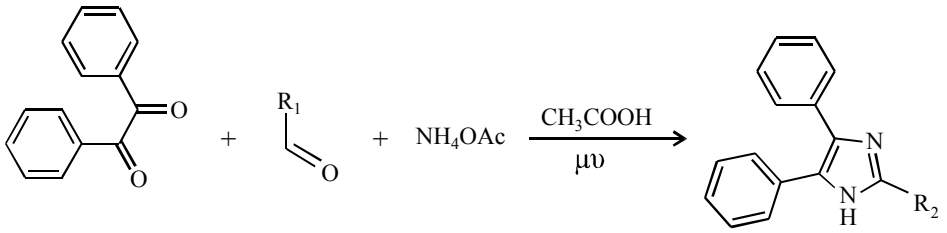
The structures of compounds **G1-12** were deduced from their ^{1}H NMR, mass and IR spectral data. The melting points and the other results are reported in Table 2.

Table 2: Analytical and spectral data for compounds G1-G12

Compd.	Molecula formula	% CHNO, found (calc.)				IR (in KBr) cm^1	Mass values m/e (M^+)	^1H NMR (DMSO, 500 MHz)
		C	H	N	O			
G1	$C_{22}H_{18}N_2O$	80.96	5.56	8.58	4.90	3316 (NH), 3062 (CH, Ar) 1660 (C=C), 1592 (C=N) 1211 (C-O-C)	326.14	12.43 (s,NH), 3.82 (s,-CH_3), 7.62-7.94 (m, ArH)
G2	$C_{21}H_{15}N_3O_2$	73.89	4.43	12.31	9.37	3438 (NH), 3066 (CH, Ar) 1675 (C=C), 1596 (C=N) 1525 (NO_2).	341.01	12.96 (s, NH), 8.0 (d, Ar), 7.44-7.94 (m, ArH)
G3	$C_{23}H_{21}N_3$	81.38	6.24	12.38	-	3421 (NH), 3027 (CH, Ar) 1616 (C=N), 2929s (-CH_3)	339.10	12.32 (s, NH), 2.96 (s,-CH_3) 6.78-6.81 (d, Ar), 7.897, 2 (d, Ar), 7.27-7.55 (m, ArH)
G4	$C_{22}H_{18}N_2O_2$	77.17	5.30	8.18	9.35	3511 (NH), 3058 (CH, Ar) 1604 (C=N), 1230 (C-O-C)	341.46	12.41 (s,NH), 3.86 (s,-CH_3), 5.0 (s, OH), 7.63 (d, Ar), 6.85 (d Ar), 7.28-7.63 (m, ArH)
G5	$C_{23}H_{20}N_2O_2$	77.51	5.66	7.86	8.98	3511 (NH), 3060 (CH, Ar) 1604 (C=N), 2975 (-CH_3) 1224 (C-O-C)	356.47	12.39 (s, NH), 1.37-1.40 (m,-CH_3), 4.09-4.13 (m, -CH_2), 6.85 -6.87 (d, Ar), 7.27-7.62(m,ArH)
G6	$C_{21}H_{16}N_2O$	80.75	5.16	8.97	5.12	3424 (NH), 3060 (CH, Ar) 1660 (C=C), 1594 (C=N)	312.72	12.95 (s, NH), 5.23 (s, OH), 6.93-6.99 (m, Ar), 7.62-7.93 (m, ArH)

Cont...

Compd.	Molecula formula	% CHNO, found (calc.)				IR (in KBr) cm[1]	Mass values m/e (M^+)	^{1}H NMR (DMSO, 500 MHz)
		C	H	N	O			
G7	$C_{19}H_{14}N_2O$	79.70	4.93	9.78	5.59	3448 (NH), 3064 (CH, Ar) 1660 (C=C), 1594 (C=N) 1211 (C-O-C)	286.71	12.82 (s, NH), 6.97-6.98 (d), 6.64-6.65 (m) (furan), 7.41-7.94 (m, ArH)
G8	$C_{23}H_{18}N_2$	85.68	5.63	8.69	-	3448 (NH), 3062 (CH, Ar) 1660 (C=C), 1592 (C=N)	321.71	12.59 (s, NH), 6.56 (d, -CH=CH-), 7.59-7.94 (m, ArH)
G9	$C_{22}H_{16}N_2O$	81.46	4.97	8.64	4.93	3448 (NH), 3064 (CH, Ar) 1660 (C=C), 1592 (C=N)	324.03	12.41 (s, NH), 9.18 (s,-CHO), 7.62-7.80 (m, ArH), 7.92-7.94 (d, Ar)
G10	$C_{23}H_{20}N_2O_2$	77.51	5.66	7.86	8.98	3421 (NH), 3062 (CH, Ar) 1660 (C=C), 1592 (C=N) 1253 (C-O-C), 2960 (CH_3)	356.81	12.53 (s, NH), 3.81-3.85 (d, CH_3), 7.05- 7.07 (d, Ar), 7.92-7.94 (d, Ar), 7.21-7.80 (m, ArH)
G11	$C_{22}H_{18}N_2$	85.13	5.85	9. 03	-	3450 (NH), 3062 (CH, Ar) 1660 (C=C), 1592 (C=N)	311.78	12.61 (s, NH), 2.35(s, CH_3), 7.92-7.94 (d, Ar), 7.97-7.98 (d, Ar), 7.62-7.82 (m, ArH)
G12	$C_{23}H_{20}N_2O_3$	74.18	5.41	7.52	12.89	3438 (NH), 3062 (CH, Ar) 1660 (C=C), 1594 (C=N) 1211 (C-O-C)	372.83	12.47 (s, NH), 3.85 (s, CH_3), 5.0 (s, OH), 7.92-7.93 (d, Ar), 7.37-7.82 (m, ArH)



CONCLUSION

This microwave assisted method simplifies the laborious procedures and offers considerable advantages, such as; elimination of solvents, the use of substances without any modification or activation, high yields, short reaction time and environmentally friendly character over the existing methodologies.

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