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Synthesis of Some Benzalacetophenones and Their Imino Derivatives

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> A group of five new benzalacetophenones (L_1 - L_5) were synthesized by the condensation of variably substituted aldehydes with acetophenone and acetyl furan, in the presence of 10 % NaOH and ethanol. Further more benzalacetophenones (L_1 - L_5) were converted to three series of their imino derivatives (L_1 - L_5 C₅). The chemical structure of these compounds was characterized by FTIR, EIMS, ¹H NMR, ¹³C NMR spectroscopy and CHN analysis.

> Key Words: Synthesis, Benzalacetophenone, Acetyl furan, Imino derivatives.

INTRODUCTION

Benzalacetophenones constitute a class of naturally occurring pigments, which are often referred to as chalcones. Kostanecki¹ first coined this term and did pioneering work in the synthesis of natural colouring compounds. An interesting feature of these compounds is that they serve as starting material for the synthesis of another class of naturally occurring pigments called flavones. Many patents have appeared in the literature, describing the usefulness of these compounds and their derivatives. These find applications as artificial sweeteners, oxidation inhibitors, stabilizers against light, heat and aging², visible and UV light³, colour photography⁴, scintillates⁵, polymerization catalysts⁶, fluorescent whiting agents⁷ and organic brightening additives8. Benzalacetophenone analogues have been reported to be bioactive compounds, e.g. furyl-analogues of benzalacetophenone^{9,10} have been reported to be a significant bacteriostatic agent against E. coli, S. aureus, B. micoidis, B. subtilis and S. lutea. These compounds also act as strong toxic accaricidal agents^{11,12}. Oxime derivatives of benzalacetophenone exhibit week herbicidal activity¹³. Benzalacetophenone and its semicarbazone derivatives possess high antifungal activity¹⁴. In continuation of our reported work¹⁵ on the synthesis of benzylacetophenones, we now report the synthesis of one new heterocyclic mimic of benzylaceto-phenone and another fifteen new imino derivatives of benzylacetophenone.

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EXPERIMENTAL

All melting points were determined in open capillaries using Gallenkemp melting point apparatus and are un-corrected. All chemicals were supplied by Fluka and Sigma Aldrich (Germany). R_f values were calculated by using pre-coated silica gel aluminum backed plates Kieselgel 60 F₂₅₄ Merck (Germany), in ethyl acetate:pet-ether (1:2). FTIR spectra were recorded on Bio-Rad Merlin using KBr discs. ¹H NMR spectra were recorded on Bruker (400 MHz) AM-250 using TMS as internal standard. EIMS were recorded on VG: 70 SE Mass Spectrometer. Purity of each compound was monitored by TLC.

Preparation of 1,3-difuranyl prop-2-en-1-one (L₁): 1,3-Difuranyl prop-2-en-1-one was synthesized by condensing 2-acetyl furan (0.01 mol) with benzaldehyde (0.01 mol) according to Claisen Schmidt condensation reaction^{16,17}.

Preparation of 1-(2'-hydroxy)-3-phenyl-prop-2-ene(L₅): 1-(2'-Hydroxy)-3-phenyl-prop-2-ene (L₅) was synthesized by condensing 2'-hydroxy acetophenone (0.01 mol) with benzaldehyde (0.01 mol) in 100 mL of distilled ethanol in a round bottom flask and 50 % aqueous solution of KOH was added drop wise to the reaction mixture with continuous stirring for about 0.5 h. The reaction mixture was then allowed to stand over night. Crushed ice was then added to it and was neutralized with dilute HCl, the product was filtered off and recrystallized from ethanol.

Preparation of 1, 3-difuranyl-pro-2-ene-1-oxime (L_1A_1): In a 250 mL round bottom flask, 5.83 g (0.084 mol) hydroxylamine hydrochloride, 35 mL of distilled water, 24 mL of 10 % NaOH solution, 5 g (0.024 mol) of furfural and enough ethanol was added drop wise while shaking, to obtain a clear solution. The reaction mixture was refluxed on a water bath for about 10 to 20 min. The resultant solution was cooled in an ice bath and crystallization was initiated by scratching the walls of the container with a glass rod. The contents of the flask were allowed to stand over night in a refrigerator. The crystals thus formed were collected by suction filtration, washed with ice cold water and recrystallized from ethanol. Same procedure was used for the synthesis of ($L_2A_2-L_5A_5$).

Preparation of phenyl hydrazone derivatives of 1,3-difuranyl prop-2-en-1-one (L₁B₁): To a solution of 5 g (0.024 mol) of 1,3-difuranyl prop-2-en-1-one (L₁) in 25 mL of glacial acetic acid, 2.35 mL (0.022 mol) of phenyl hydrazine was added gradually with continuous shaking and the mixture was allowed to stand for 1 h. Yellow crystals began to separate which were filtered on a Buchner funnel and washed with supper cooled ethanol. Recrystallization of the product (L₁B₁) was achieved with 90 % ethanol. Same procedure was used for the synthesis of (L₂B₂-L₅B₅). Vol. 19, No. 7 (2007) Synthesis of Benzalacetophenones and Their Imino Derivatives 5059

Preparation of 2,4 dinitrophenyl hydrazone derivatives of 1,3difuranyl prop-2-en-1-one (L_1C_1): To a solution of 4.42 g (0.022 mol) of 2,4 dinitrophenyl hydrazine in 24 mL of distilled water, 24 mL of concentrated sulfuric acid was added drop wise. The resultant solution was allowed to attain room temperature. 100 mL of 95 % ethanol was then added to this solution. Separately a solution of 2-furyl acetophenone 5 g (0.022 mol) in 100 mL of 95 % ethanol was prepared. This solution was then added slowly to the 2,4-dinitrophenyl hydrazine solution and was refluxed on a steam bath for 15-20 min. On cooling crystals of (L_1C_1) were formed and were collected by suction filteration, washed with cold ethanol and recrystallized with 98 % ethanol. Same procedure was used for the synthesis of ($L_2C_2-L_5C_5$).

RESULTS AND DISCUSSION

Benzalacetophenones L_{1-5} were synthesized by condensing acetophenone (a) with appropriate benzaldehyde derivatives (b) in dilute ethanolic sodium hydroxide solution at room temperature, their oxime derivatives $(L_1A_1-L_5A_5)$, Phenyl hydrazone derivatives $(L_1B_1-L_5B_5)$ and 2,4-dinitro phenylhydrazone derivatives $(L_1C_1-L_5C_5)$ were synthesized by the reaction of appropriate benzalacetophenone derivatives (L_{1-5}) , according to Scheme-I. Physical data of all synthesized benzalacetophenones, their oxime, phenyl hydrazone and 2,4-dinitrophenyl hydrazone derivatives are listed in Table-1.



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Compound	R_1	R_2	R ₃
L_1C_1	$\sqrt[]{}_{o}$	$\sqrt{2}$	NH NO ₂
L_2	\bigcirc	CI	_
L_2A_2	\bigcirc	CI CI	–OH
L_2B_2	\bigcirc		NH
L_2C_2		CI CI	NO2 NO2
L_3	\bigcirc	OMe	_
L_3A_3	\bigcirc	OMe	–OH
L_3B_3	\bigcirc	OMe	NH
L ₃ C ₃	\bigcirc	OMe	NH NO ₂
L_4	$_{o}$	\bigcirc	_
L_4A_4	$_{0}$	\bigcirc	–OH
L_4B_4	$\sqrt[]{}$	\bigcirc	
L_4C_4		$\hat{\mathbf{Q}}$	NH NO ₂
L_5	ОН	\bigcirc	_
L_5A_5	ОН	\bigcirc	–OH
L_5B_5	ОН		NH
L_5C_5	ОН		NH NO ₂
	S	cheme-I	NO ₂

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Scheme-I

PHYSICAL DATA OF SYNTHESIZED COMPOUNDS L ₁ -L ₅ C ₅							
Comme	m.f.	m.w.	m.p.	Yield (%)	Found (Calcd.) %		
Compd.			(°Ĉ)		С	Н	Ν
т	СНО	188	191-	73.00	70.8	4.32	_
\mathbf{L}_1	$C_{11} \Pi_8 O_3$	100	193	75.00	(70.21)	(4.25)	
L	CirHuOCl	242	97-	75.00	74.3	4.83	_
-2	015111001	2.2	99	72.00	(74.12)	(4.54)	
La	$C_{16}H_{14}O_{2}$	239	65-	82.00	79.4	5.23	_
5	10 14 2		6/		(80.67)	(5.88)	
L_4	$C_{13}H_{10}O_2$	233	94- 05	78.00	(78.12 (78.78)	4.89	_
			95 70		(70.70)	(3.03)	
L_5	$C_{11}H_8O_3$	224	78- 80	82.00	(80.35)	14.09	_
			195-		64 99	(14.20) 4.67	6.81
L_1A_1	$C_{11}H_9N$	155	196	68.00	(65.16)	(4.43)	(6.89)
			178-		69.29	4.51	5.69
L_2A_2	$C_{15}H_{12}NOCl$	257	180	65.00	(70.03)	(4.66)	(5.44)
T A		252	123-	C 1 00	75.12	5.67	5.20
L_3A_3	$C_{16}H_{15}NO_2$	253	125	64.00	(75.88)	(5.92)	(5.53)
ТА		212	252-	75.00	74.11	5.52	6.50
L_4A_4	$C_{13} \Pi_{11} \Pi O_2$	213	253	75.00	(73.90)	(5.21)	(6.63)
ΙΑ.	C.H.NO	223	211-	62 00	80.11	5.92	6.50
125/15	0151113110	225	213	02.00	(80.71)	(5.82)	(6.27)
L ₁ B ₁	$C_{17}H_{14}N_{2}O_{2}$	278	253-	73.00	72.67	6.29	9.82
-1-1	-17 14 2 2		255		(73.38)	(5.03)	(10.07)
L_2B_2	$C_{21}H_{17}N_2Cl$	332	121-	69.00	84.45	6.29	9.82
	21 1, 2		122		(84.50)	(0.04)	(9.39)
L_3B_3	$C_{22}H_{20}N_2O$	328	220-	71.00	80.31 (80.48)	(6.09)	8.40 (8.53)
			154-		80.11	(0.07)	9.51
L_4B_4	$C_{19}H_{16}N_2O$	228	154-	67.77	(79.16)	(5.9	(9.72)
			202-	60.01	80.11	5.12	9.11
L_5B_5	$C_{21}H_{19}N_2O$	314	205	69.01	(80.25)	(5.73)	(8.91)
ТС		269	198-	62.01	55.63	3.33	7.54
$\mathbf{L}_{1}\mathbf{C}_{1}$	$C_{17}H_{12}N_4O_6$	308	201	02.01	(55.43)	(3.26)	(7.60)
ТС	CHNOC	122	178-	50.07	59.71	3.55	12.98
$\mathbf{L}_2\mathbf{C}_2$	C_{21} C_{151} C_{4} C_{4} C_{1}	422	180	50.07	(59.71)	(3.55)	(13.27)
L ₂ C ₂	$C_{22}H_{10}N_4O_{\pi}$	418	201-	53.00	63.05	4.32	13.40
-,~,	-22-18-14-5	.10	203	22.00	(63.15)	(4.30)	(13.39)
L_4C_4	$C_{19}H_{15}N_4O_5$	378	210-	50.10	60.63	3.81	14.85
	17 15 7 5		112		(60.31)	(5.70)	(14.81)
L_5C_5	$C_{21}H_{16}N_4O_5$	404	211- 213	62.00	01.03	3.81 (3.96)	13.15
			415		(04.57)	(3.70)	(10.00)

Vol. 19, No. 7 (2007) Synthesis of Benzalacetophenones and Their Imino Derivatives 5061 TABLE-1

Benzalacetophenone *mimic* 1,3-difuranyl prop-2-en-1-one (L_1) was obtained as off white crystals. The molecular formula and molecular weight of the compound (L_1) are $C_{11}H_8O_3$ and 188, respectively. The FTIR spectrum showed stretching frequencies at 1506 cm⁻¹, 1690 cm⁻¹ which are characteristics of aromatic (C=C) and carbonyl (C=O), respectively.

¹H NMR spectrum of (L_1) showed two doublets at 6.67 and 7.54 ppm with large coupling constant of 15 and 17.6 Hz, which are respectively characteristic of the α and β protons as shown below:



The six aromatic protons of the two furan rings appear as doublets in the range of 6.61-7.80 ppm. ¹³C NMR spectrum showed two doublets at 127.4 and 131.5 ppm, which are characteristic of the two alkenyl carbons, carrying the α and β protons. Singlets at 153.6 and 151.6 ppm are characteristic of the two furan ring carbons, which are attached to carbonyl and alkenyl groups of the molecule, respectively. Doublets at 121.2, 112.6, 149.3, 111.4, 112.7 and 145.9 ppm are due to six carbon atoms of the two furane rings to which aromatic protons are attached. A singlet at 177.6 ppm showed the presence of a carbonyl carbon.

The mass spectrum of 1,3-difuranyl prop-2-en-1-one (L_1) showed molecular ion peak [M]⁺ at m/z 188 which is also the base peak and is in good agreement with the molecular weight of the compound. Physical and spectral (FTIR, ¹H NMR, ¹³C NMR, EIMS) data confirmed the structure of benzalacetophenone *mimic* (L_1) as 1,3-difuranyl prop-2-en-1-one. Similarly benzalacetophenone ($L_{2.5}$) were characterized on the basis of their physical and spectral data.

Oxime derivative of 1,3-diphenyl-pro-2-ene (L_1A_1) of benzalacetophenone *mimic* (L_1) was obtained as white granules. In the FTIR spectrum of this derivative, the band due to carbonyl of L_1 at 1690 cm⁻¹ disappeared and a new band at 1585 cm⁻¹ appeared, which is characteristic of (C=N). The molecular ion peak enhanced from 188 to 203 and in ¹H NMR and ¹³C NMR a significant singlet at 11.00 ppm and a peak at 164.6 ppm are due to the hydroxyl proton and the carbon doubly bonded to nitrogen, respectively. All other oxime derivatives ($L_2A_2-L_5A_5$) were similarly identified.

1,3-Diphenyl-pro-2-ene-1-phenylhydrazone ($\mathbf{L}_1\mathbf{B}_1$) was obtained as pale yellow crystals. There was not much difference between the FTIR spectra of ($\mathbf{L}_1\mathbf{A}_1$) and ($\mathbf{L}_1\mathbf{B}_1$) however, the molecular ion peak further increased to m/z = 278. ¹H NMR spectrum of 1,3-diphenyl-pro-2-ene-1-phenylhydrazone ($\mathbf{L}_1\mathbf{B}_1$) was almost similar to that of ($\mathbf{L}_1\mathbf{A}_1$) except for the presence of Vol. 19, No. 7 (2007) Synthesis of Benzalacetophenones and Their Imino Derivatives 5063

a singlet at 7.00 due to the imino proton instead of a hydroxyl proton of (L_1A_1) and a multiplet between 6.4-7.01 ppm due to the protons of phenyl ring attached to the imino group. ¹³C NMR spectrum of (L_1B_1) was also not very different form (L_1A_1) except that a singlet at 143.1, doublets at 116.3, 129.8, 118.8 were observed, which are due to the carbons of the phenyl ring attached to the imino moity. All other phenylhydrazone derivatives $(L_2B_2-L_5B_5)$ were characterized accordingly. Similarly all other 2",4"-dinitrophenylhydrazone derivatives $(L_2C_2-L_5C_5)$ were characterized on the basis of their physical and spectral data.

Complete spectral data (¹H NMR, ¹³C NMR, EIMS, FTIR) of all synthesized compounds (L_2 - L_5C_5) are tabulated in Tables 2-4.

Compd.	¹ H NMR (ppm)
L ₁	δH: 6.67 (d, 1H, 15Hz), 7.54 (d, 1H, 17.6Hz), 7.00 (dd, 1H), 6.65 (dd, 1H), 7.80 (dd, 1H), 7.23 (dd, 1H), 6.61 (dd, 1H), 7.72 (dd, 1H).
L ₂	7.56 (d, 1H, 15Hz), 7.90 (d, 1H, 15Hz), 7.24 (dd, 2H, 8.2, 8.5Hz), 7.22 (dd, 2H, 8.3, 2.5Hz), 7.81 (m, 2H), 7.45 (m, 1H), 7.54 (m, 2H)
L ₃	3.73 (S, 3H), 7.90 (d, 1H, 13.7Hz), 7.56 (d, 1H, 15Hz), 7.19 (dd, 2H, 7.2, 1.2Hz), 6.72 (dd, 2H, 8.3, 1.02Hz), 7.71 (m, 2H, 7.9, 2.3Hz), 7.45 (m, 1H, 1.8Hz), 7.54 (m, 2H, 7.9, 2.3Hz).
L_4	6.67 (d, 1H, 12.11), 7.54 (d, 1H, 13.7Hz), 6.61 (dd, 1H, 3.2, 1.8Hz), 7.23 (d, 1H, 3.5Hz), 7.72 (d, 1H, 1.02Hz), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H)
L_5	7.56 (d, 1H, 12.11Hz), 5.00 (s, 1H), 6.92 (m, 1H), 7.37 (m, 1H), 7.01 (m, 1H), 7.64 (m, 1H), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H).
L ₁ A ₁	5.6 (d, 1H, 15.01Hz), 6.67 (d, 1H, 12.2Hz), 11.00 (s, 1H), 6.3 (dd, 2H), 7.4 (dd, 1H), 6.3 (dd, 2H), 7.4 (dd, 1H)
L_2A_2	5.6 (d, 1H, 13.2Hz), 6.67 (d, 1H, 12.5Hz), 11.00 (1H), 7.3 (dd, 2H), 7.21 (dd, 2H), 7.6 (m, 2H), 7.3 (m, 2H), 7.14 (m, 1H).
L ₃ A ₃	5.6 (d, 1H, 13.2Hz), 6.67 (d, 1H, 11.5Hz), 10.20 (s, 1H), 7.6 (m, 2H), 7.3 (m, 2H), 7.14 (m, 1H), 7.19 (dd, 2H), 6.72 (dd, 2H), 3.73 (s, 1H).

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Compd.	¹ H NMR (ppm)
L_4A_4	5.6 (d, 1H, 14.5Hz), 6.67 (d, 1H, 11.6Hz), 11.52 (s, 1H), 7.3 (m, 2H) 7.21 (m, 2H), 7.14 (m, 1H), 6.3 (d, 1H, 3.2Hz), 6.4 (dd, 1H, 2.5, 1.2Hz), 7.54 (d, 1H, 1.5Hz)
L_5A_5	5.62 (d, 1H, 12.5Hz), 6.69 (d, 1H, 13.9Hz), 5.00 (s, 1H), 6.8 (m, 1H), 7.11 (m, 1H), 6.8 (m, 1H), 7.4 (m, 1H), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H), 6.46 (m, 2H), 7.01 (m, 1H), 6.62 (m, 1H).
L_1B_1	5.6 (d, 1H, 15.2Hz), 6.6 (d, 1H, 13.02Hz), 6.3 (dd, 4H) 7.4 (dd, 2H), 7.01 (s, 1H), 6.46 (m, 2H), 7.01 (m, 2H), 6.62 (m, 1H).
L_2B_2	7.90 (d, 1H, 15Hz), 7.56 (d, 1H, 15Hz), 7.6 (m, 2H) 7.31 (m, 2H), 7.14 (m, 1H), 7.02 (s, 1H), 7.22 (dd, 2H, 2.5, 6.9Hz), 6.24 (dd, 1H, 7.2, 6.2Hz), 6.6 (d, 1H, 15Hz), 7.00 (s, 1H) 6.46 (m, 2H), 7.01 (m, 2H), 6.46 (m, 2H).
L ₃ B ₃	5.6 (d, 1H, 11.9Hz), 6.6 (d, 1H, 12.7Hz), 11.52 (s, H, 7.6 (m, 2H), 7.3 (m, 2H), 7.14 (m, 1H), 7.19 (dd, 2H, 7.1, 2.2Hz), 6.72 (dd, 2H, 7.5, 1.2Hz), 3.73 (s, 3H), 6.6 (d, 1H, 15Hz), 7.00 (s, 1H) 6.46 (m, 2H), 7.01 (m, 2H),
L_4B_4	5.6 (d, 1H, 13.2Hz), 6.67 (d, 1H, 11.95Hz), 11.52 (s, 1H), 6.3 (d, 1H, 2.1Hz), 6.39 (d, 1H, 1.4Hz), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H) (d, 1H, 1.2Hz), 6.46 (m, 2H), 7.01 (m, 2H), 6.62 (m, 1H),
L_5B_5	5.6 (d, 1H, 14.02Hz), 6.67 (2.5Hz), 5.00 (s, 1H), 6.8 (m, 1H), 7.11 (m, 1H), 6.8 (m, 1H), 7.4 (m, 1H), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H), 6.46 (m, 2H), 7.01 (m, 1H), 6.62 (m, 1H)
L_1C_1	5.6 (d, 1H, 15Hz), 6.6 (d, 1H, 15Hz), 6.3 (dd, 4H) 7.4 (dd, 2H), 7.01 (s, 1H), 6.98 (m, 1H), 8.33 (m, 1H), 8.87 (m, 1H).
L ₂ C ₂	7.90 (d, 1H, 15Hz), 7.56 (d, 1H, 13.8Hz), 7.02 (s, 1H) 7.6 (m, 2H), 7.3 (m, 2H), 61.4 (m, 2H), 7.24 (dd, 2H, 2.2, 5.5Hz), 7.22 (dd, 2H, 2.5, 6.9Hz), 6.90 (dd, 1H, 7.2, 6.2Hz), 8.33 (dd, 1H, 6.9, 5.5Hz), 8.87 (dd, 1H, 4.5, 6.9Hz).
L ₃ C ₃	5.6 (d, 1H, 14.2Hz), 6.6 (d, 1H, 11.9Hz), 11.52 (s, 1H) 7.6 (m, 2H), 7.3 (m, 2H), 7.14 (m, 1H), 7.19 (dd, 2H, 7.1, 2.2Hz), 6.72 (dd, 2H, 7.5, 1.2Hz), 3.73 (s, 3H), 6.98 (dd, 1H, 5.1, 6.8Hz), 8.33 (dd, 1H, 1.2, 6.7Hz), 8.87 (dd, 1H, 6.2, 7.9Hz).
L ₄ C ₄	5.6 (d, 1H, 12.2Hz), 6.67 (d, 1H, 13.9Hz), 11.52 (s, 1H), 6.3 (d, 1H, 2.1Hz), 6.39 (d, 1H, 1.4Hz), 7.4 (d, 1H, 1.2Hz), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H), 6.98 (dd, 1H, 5.1, 6.8Hz), 8.33 (dd, 1H, 1.2, 6.7Hz), 8.87 (dd, 1H, 6.2, 7.9Hz).
L_5C_5	5.6 (d, 1H, 12.2Hz), 6.67 (d, 1H, 11.9Hz), 5.00 (s, 1H), 6.8 (m, 1H), 7.11 (m, 1H), 6.8 (m, 1H), 7.4 (m, 1H), 7.30 (m, 2H), 7.21 (m, 2H), 7.14 (m, 1H), 6.98 (m, 1H), 8.33 (m, 1H), 8.87 (m, 1H).

TABLE-3				
	C NIVIR DATA OF STINTHESIZED COMPOUNDS $L_1-L_5C_5$			
Compd	. ¹³ C NMR (ppm)			
L_1	δC: 127.4 (d, 1C), 131.5 (d, 1C), 177.8 (s, 1C), 153.6 (s, 1C), 121.2 (d, 2C), 112.6 (d, 1C), 149.3 (d, 1C), 151.6 (s, 1C), 111.4 (d, 1C), 112.7 (d, 1C), 145.9 (d, 1C).			
L ₂	121.4 (d, 1C), 145.2 (d, 1C), 189.7 (s, 1C), 137.9 (s, 1C), 129.9 (d, 2C), 129.3 (d, 2C), 134.6 (d, 1C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128.0 (s, 1C).			
L ₃	121.4 (d, 1C), 145.2 (d, 1C), 187.7 (s, 1C), 137.9 (s, 1C), 129.9 (d, 2C), 129.3 (d, 2C), 134.6 (d, 1C), 127.5 (s, 1C), 127.4 (d, 2C), 114.2 (d, 2C), 159.9 (s, 1C), 55.9 (q, 1C)			
L_4	121.4 (d, 1C), 145.2 (d, 1C), 177.8 (s, 1C), 153.6 (s, 1C), 121.2 (d, 1C), 112.6 (d, C), 149.3 (d, 1C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128.0 (d, 1C).			
L_5	121.4 (d, 1C), 145.2 (d, 1C), 189.7 (s, 1C), 122.7 (s, 1C), 161.8 (s, 1C), 116.9 (d, C), 136.0 (d, 1C), 121.9 (d, 1C), 131.3 (d, 2C), 145.2 (s, 1C), 128.7 (d, 2C), 128.0 (d, 1C).			
L_1A_1	126.6 (d, 1C), 128.8 (d, 1C), 164.6 (s, 1C), 143.2 (s, 1C), 109.5 (d, 2C), 143.9 (d, 1C), 151.6 (s, 1C), 111.4 (s, 1C), 112.7 (d, 1C), 145.9 (d, 1C).			
L_2A_2	120.6 (d, 1C), 142.5 (d, 1C), 164.6 (s, 1C), 133.2 (s, 1C), 129.2 (d, 2C), 128.9 (d, 2C), 131.1 (d, 1C), 133.3 (s, 1C), 127.8 (d, 2C), 128.8 (d, 2C), 133.6 (s, 1C).			
L ₃ A ₃	120.6 (d, 1C), 142.9 (d, 1C), 164.6 (s, 1C), 133.2 (s, 1C), 129.2 (d, 2C), 128.9 (d, 2C), 131.1 (d, 1C), 127.5 (s, 1C), 127.4 (d, 2C), 114.2 (d, 2C), 159.9 (s, 1C), 55.9 (q, 1C).			
L_4A_4	120.6 (d, 1C), 142.5 (d, 1C), 164.6 (s, 1C), 143.6 (s, 1C), 109.5 (d, 1C), 109.9 (d, C), 143.9 (d, 1C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128.0 (d, 1C).			
L_5A_5	120.6 (d, 1C), 142.5 (d, 1C), 164.6 (s, 1C), 118.4 (s, 1C), 161.1 (s, 1C), 116.0 (d, C), 132.5 (d, 1C), 121.5 (s, 1C), 130.6 (d, 2C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128 (d, 1C).			
L_1B_1	124.7 (d, 1C), 125.3 (d, 1C), 155.6 (s, 1C), 143.2 (s, 1C), 109.9 (d, 2C), 143.9 (d, 1C), 151.6 (s, 1C), 111.4 (s, 1C), 112.7 (d, 1C), 145.9 (d, 1C).			
L ₂ B ₂	118.7 (d, 1C), 139.0 (d, 1C), 155.6 (s, 1C), 133.2 (s, 1C), 129.2 (d, 2C), 131.1 (d, 2C), 133.3 (s, 1C), 127.8 (d, 2C), 128.8 (d, 2C), 143.1 (s, 1C), 116.3 (d, 1C), 129.6 (d, 1C), 118.8 (d, 1C), 129.8 (d, 1C), 116.3 (d, 1C).			
L ₃ B ₃	118.7 (d, 1C), 139 (d, 1C), 155.6 (s, 1C), 133.2 (s, 1C), 129.2 (d, 2C), 128.9 (d, 2C), 131.9 (d, 1C), 127.5 (s, 1C), 127.4 (d, 2C), 114.2 (d, 2C), 159.9 (s, 1C), 55.9 (q, 1C), 143.1 (s, 1C), 116.3 (d, 1C), 129.6 (d, 1C), 118.8 (d, 1C), 129.8 (d, 1C), 116.3 (d, 1C).			

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Compd.	¹³ C NMR (ppm)
L_4B_4	118.7 (d, 1C), 139 (d, 1C), 155.6 (s, 1C), 143. (s, 1C), 109.5 (d, 1C), 109.9 (d, 1C), 143.9 (d, 1C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128 (d, 1C), 143.1 (s, 1C), 116.3 (d, 1C), 129.6 (d, 1C), 118.8 (d, 1C), 129.8 (d, 1C), 116.3 (d, 1C).
L ₅ B ₅	118.7 (d, 1C), 139.9 (d, 1C), 155.6 (s, 1C), 118.4 (s, 1C), 161.1 (s, 1C), 116.0 (d, C), 132.5 (d, 1C), 121.5 (s, 1C), 130.6 (d, 2C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128 (d, 1C), 143.1 (s, 1C), 116.3 (d, 2C), 129.6 (d, 2C), 118.8 (d, 1C).
L ₁ C ₁	124.7 (d, 1C), 125.3 (d, 1C), 155.6 (s, 1C), 143.2 (s, 1C), 109.9 (d, 2C), 147.9 (s, 1C), 118.1 (d, 1C), 128.0 (d, 1C), 139.3 (s, 1C), 119.5 (d, 1C), 135.9 (s, 1C).
L ₂ C ₂	118.7 (d, 1C), 139.0 (d, 1C), 155.6 (s, 1C), 133.2 (s, 1C), 129.2 (d, 2C), 131.1 (d, 2C), 133.3 (s, 1C), 127.8 (d, 2C), 128.8 (d, 2C), 147.9 (s, 1C), 135.9 (s, 1C), 119.5 (d, 1C), 139.3 (s, 1C), 128 (d, 1C), 118 (d, 1C).
L ₃ C ₃	118.7 (d, 1C), 139 (d, 1C), 155.6 (s, 1C), 133.2 (s, 1C), 129.2 (d, 2C), 128.9 (d, 2C), 131.9 (d, 1C), 127.5 (s, 1C), 127.4 (d, 2C), 114.2 (d, 2C), 159.9 (s, 1C), 55.9 (q, 1C), 147.9 (s, 1C), 135.9 (s, 1C), 119.5 (d, 1C), 139.3 (s, 1C), 128 (d, 1C), 118 (d, 1C)
L ₄ C ₄	118.7 (d, 1C), 139 (d, 1C), 155.6 (s, 1C), 143. (s, 1C), 109.5 (d, 1C), 109.9 (d, 1C), 143.9 (d, 1C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128 (d, 1C), 147.9 (s, 1C), 135.9 (s, 1C), 119.5 (d, 1C), 139.3 (s, 1C), 128 (d, 1C), 118 (d, 1C).
L_5C_5	118.7 (d, 1C), 139.9 (d, 1C), 155.6 (s, 1C), 118.4 (s, 1C), 161.1 (s, 1C), 116.0 (d, C), 132.5 (d, 1C), 121.5 (s, 1C), 130.6 (d, 2C), 135.2 (s, 1C), 126.4 (d, 2C), 128.7 (d, 2C), 128 (d, 1C), 147.9 (s, 1C), 135.9 (s, 1C), 119.5 (d, 1C), 139.3 (s, 1C), 128.0 (d, 1C), 118.1 (d, 1C).

TABLE-4 FTIR AND EIMS DATA OF SYNTHESIZED COMPOUNDS L_1 - L_5C_5

Compound	FTIR (KBr, cm ⁻¹)	Mass (m/z)
L_1	1637, 1563	188
L_2	1676, 1528	242
L_3	1618, 1569	239
\mathbf{L}_{4}	1670, 1535	233
L_5	3457, 1670, 1535	224
L_1A_1	1583, 1569	155
L_2A_2	1560, 1559	257
L_3A_3	1575, 1559	253

Compound	FTIR (KBr, cm ⁻¹)	Mass (m/z)
L_4A_4	1585, 1559	213
L_5A_5	3465, 1585, 1559	223
L_1B_1	1587, 1559	278
L_2B_2	1583, 1559	332
L_3B_3	1585, 1559	328
L_4B_4	1579, 1559	288
L_5B_5	3467, 1579, 1559	314
L_1C_1	1585, 1559	368
L_2C_2	1585, 1559	422
L_3C_3	1585, 1559	418
L_4C_4	1585, 1559	378
L ₅ C ₅	3476, 1585, 1559	404

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