MICHAEL–MICHAEL ALDOL REACTION OF CHALCONES WITH CYANOACETYLUREA AND CYANOACETYLPIPERIDINE

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Several highly substituted cyclohexane derivatives were synthesized by the one-pot condensation of chalcones and cyanoacetylurea (2 : 1) using sodium alkoxide in anhydrous alcohol at room temperature. The structure of the reaction products was established by infrared, ^{1}H and ^{13}C NMR spectroscopy as well as by their elemental analysis. Single crystal X-ray crystallography shows the presence of the cyclohexane moiety.

Until recently, many authors have published on the condensation reaction of chalcones with active methylene compounds[1–5], but less is known about the reaction of chalcones with *N*-cyanoacetyl derivatives. For example, the reaction of chalcones with malononitrile[6] and ethyl cyanoacetate[7] in presence of ammonium acetate gives substituted pyridines. Chalcones also react with thioacetamide[8] to give 2-pyridinethiones, with cyanoacetamide[9] to give 2-pyridones, with ethyl cyanoacetate[10] in presence of sodium ethoxide to give 8-oxaquinolines and with ethyl phenylacetate[11] to give the corresponding ethyl 3-aryl-4-benzoyl-2-phenylbutyrates. In addition, chalcones were also condensed with guanidine hydrochloride[12] to give 4,6-diaryl-3,4-dihydro-1*H*-pyrimidine-2-thione. Furthermore, the preparative value of one-pot multicomponent, sequential Michael–Michael ring closure reactions followed by aromatization is illustrated by a total synthesis of juncunol[13], an unusual dihydrophenanthrene. Recently, we reported that the base catalyzed reaction of chalcones with benzyl cyanides[14] in 1 : 1 molar ratio using a suspension of sodium ethoxide in ether affords a single diastereoisomer of 2,3-diaryl-4-aroylbutyronitriles.

More recently, we reported[15] that the base catalyzed condensation of benzyl cyanides and chalcones in a 1 : 2 molar ratio using sodium ethoxide in anhydrous ether at room temperature gives only 1,3,4,5-tetraaryl-2-aroyl-4-cyanocyclohexanols. An ambiguous structural assignment was achieved from analytical and infrared, proton and ^{13}C nuclear

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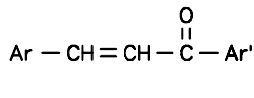
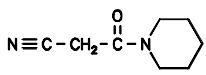
magnetic resonance spectral data as well as single crystal X-ray crystallography. In view of our continued interest in the chemistry of active methylene nitriles and other related compounds[16–22], we report herein the synthesis of a series of highly substituted



I



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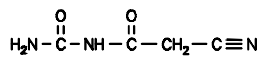

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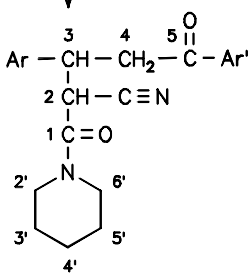

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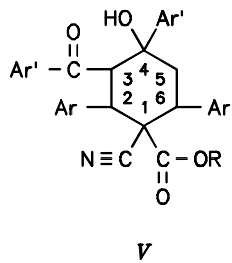

V

IV	Ar	Ar'
a	4-tolyl	4-tolyl
b	4-tolyl	2-naphthyl
c	4-tolyl	phenyl
d	phenyl	phenyl
e	4-anisyl	4-tolyl

V	Ar	Ar'	R
a	3-bromophenyl	2-naphthyl	ethyl
b	3-bromophenyl	4-chlorophenyl	ethyl
c	4-anisyl	2-naphthyl	ethyl
d	phenyl	phenyl	ethyl
e	4-tolyl	2-naphthyl	methyl
f	3,4-dichlorophenyl	2-naphthyl	methyl
g	3-bromophenyl	2-naphthyl	methyl
h	4-tolyl	phenyl	methyl
i	3,4-dimethoxyphenyl	2-naphthyl	methyl

SCHEME 1

TABLE I
Selected bond angles (°) for the compound *Vd*

Atoms	Angles	Atoms	Angles
C5–C1–C2	109.3(7)	C6–C1–C2	106.0(17)
C6–C1–C5	111.4(7)	C9–C1–C2	110.0(7)
C9–C1–C5	108.1(8)	C9–C1–C6	111.9(8)
C3–C2–C1	108.4(7)	C21–C2–C1	112.2(3)
C21–C2–C3	115.2(7)	C4–C3–C2	109.2(6)
C31–C3–C2	108.7(7)	C31–C3–C4	109.6(7)
C10–C4–C3	109.5(7)	O3–C4–C3	110.4(7)
O3–C4–C10	105.3(6)	C41–C4–C3	113.1(7)
C41–C4–C10	106.0(7)	C41–C4–O3	112.2(2)
C10–C5–C1	109.0(6)	C51–C5–C1	113.1(7)
C51–C5–C10	113.7(7)	O1–C6–C1	121.8(9)
O2–C6–C1	114.7(8)	O2–C6–O1	123.5(8)
O2–C7–C8	116.2(12)	N1–C9–C1	173.0(11)
C5–C10–C4	112.8(7)	C7–O2–C6	115.7(8)
C22–C21–C2	125.9(8)	C26–C21–C2	117.5(9)
C26–C21–C22	116.6(9)	C23–C22–C21	120.8(9)
C24–C23–C22	121.6(11)	C25–C24–C23	119.3(12)
C26–C25–C24	119.5(11)	C25–C26–C21	122.2(10)
O4–C31–C3	119.6(8)	C32–C31–C3	121.6(8)
C32–C31–O4	118.8(8)	C33–C32–C31	122.4(8)
C37–C32–C31	118.7(8)	C37–C32–C33	118.9(8)
C34–C33–C32	120.1(9)	C35–C34–C33	121.2(10)
C36–C35–C34	117.8(10)	C37–C36–C35	122.1(11)
C36–C37–C32	119.9(10)	C42–C41–C4	124.0(11)
C46–C41–C4	118.2(10)	C46–C41–C42	117.7(9)
C43–C42–C41	122.2(12)	C44–C43–C42	119.5(13)
C45–C44–C43	121.3(14)	C46–C45–C44	119.3(16)
C45–C46–C41	119.9(12)	C52–C51–C5	119.5(8)
C56–C51–C5	121.9(7)	C56–C51–C52	118.5(9)
C53–C52–C51	121.1(9)	C54–C53–C52	119.7(9)
C55–C54–C53	120.1(10)	C56–C55–C54	120.2(10)
C55–C56–C51	120.3(8)		

cyclohexanol derivatives by a base catalyzed cyclocondensation reaction of chalcones *I* and *N*-cyanoacetylpiperidine *II* or chalcones *I* and cyanoacetylurea *III* in 2 : 1 molar ratios (Scheme 1). This simple one-pot reaction is carried out by stirring a mixture of chalcones and the cyanoacetyl derivatives using sodium alkoxide in anhydrous alcohol. In each condensation, a heavy precipitate is formed which upon workup and recrystallization from glacial acetic acid provides piperidine derivatives *IV* or the cyclohexanol derivatives *V*. When cyanoacetylurea was used as a Michael donor, the cyclohexanol derivatives *V* isolated as a result of this reaction indicated that the reaction could involve double Michael addition of cyanoacetylurea carbanion to two moles of chalcones

TABLE II
Selected bond lengths (Å) for the compound *Vd*

Atoms	Distances	Atoms	Distances
C1–C2	1.553(12)	C1–C5	1.534(13)
C1–C6	1.484(12)	C1–C9	1.424(12)
C2–C3	1.518(11)	C2–C21	1.486(13)
C3–C4	1.539(13)	C3–C31	1.499(12)
C4–C10	1.508(11)	C4–O3	1.412(10)
C4–C41	1.487(12)	C5–C10	1.515(10)
C5–C51	1.489(12)	C6–O1	1.188(11)
C6–O2	1.302(12)	C7–C8	1.219(21)
C7–O2	1.440(14)	C9–N1	1.138(12)
O4–C31	1.192(11)	C21–C22	1.356(14)
C21–C26	1.358(14)	C22–C23	1.347(16)
C23–C24	1.331(17)	C24–C25	1.332(19)
C25–C26	1.360(17)	C31–C32	1.470(12)
C32–C33	1.334(13)	C32–C37	1.334(14)
C33–C34	1.354(13)	C34–C35	1.337(16)
C35–C36	1.326(18)	C36–C37	1.341(15)
C41–C42	1.337(16)	C41–C46	1.361(17)
C42–C43	1.360(16)	C43–C44	1.299(25)
C44–C45	1.344(25)	C45–C46	1.376(17)
C51–C52	1.354(11)	C51–C56	1.354(13)
C52–C53	1.367(14)	C53–C54	1.328(16)
C54–C55	1.349(14)	C55–C56	1.358(15)

followed by cyclization with the elimination of urea molecule and then introduction of ethoxy or methoxy group instead. However, when *N*-cyanoacetylpiperidine was used as the Michael donor, a simple Michael adduct *IV* resulting from the addition of the acetylpiperidine carbanion to the chalcone was obtained and none of the cyclohexanol derivative analogs were isolated. The reason could also be that the cyanoacetylurea is more acidic than the piperidine derivative and hence a second Michael addition is possible.

The structural identification of the cyclohexanol derivatives *Va – Vi* was based on the spectral data, elemental analysis and X-ray crystallography. The infrared spectra of these cyclohexanol derivatives show a peak at 3 410 – 3 500 cm^{-1} corresponding to the stretching frequency of the hydroxyl group in addition to a very intense band at about 1 650 cm^{-1} due to the benzoyl carbonyl stretching frequency and a band at around 2 220 cm^{-1} characteristic for the cyano group. The carbalkoxy carbonyl stretching frequency shows a strong peak at around 1 735 cm^{-1}. The nuclear magnetic resonance spectral data of the cyclohexanol derivatives were also consistent with the suggested structure. As a representative example, in the ^1H NMR spectrum of the compound *Vd* the hydroxyl proton signal is a doublet (J = 2.5 Hz) as a result of a long range coupling with H-5. The intramolecular hydrogen bonding between OH and H-5 makes the molecule rigid enough in a W geometry and this makes the long range coupling possible.

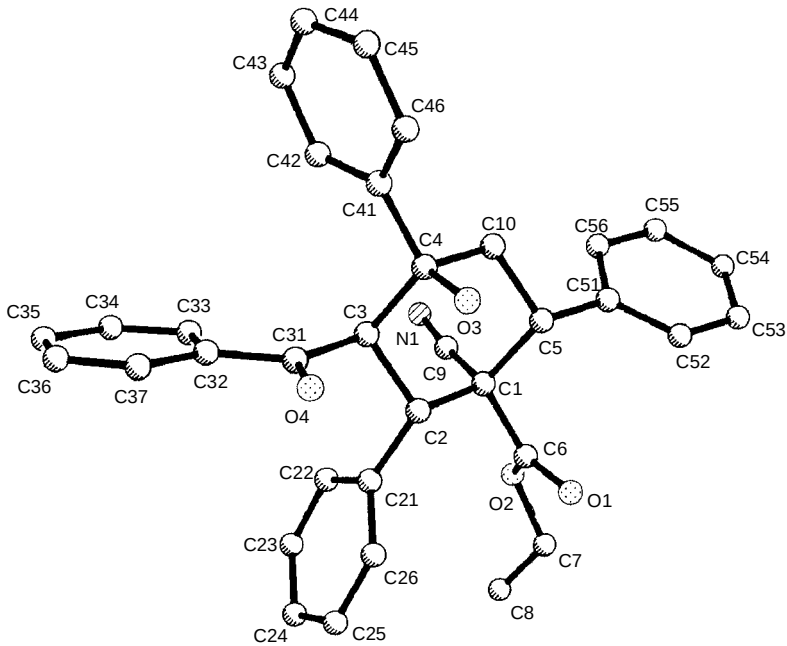

FIG. 1
The crystal structure of the compound *Vd*

TABLE III
Atomic coordinates (. 10^4) and equivalent isotropic displacement parameters ($Å^2$. 10^3) for the compound *Vd*

Atom	*x*	*y*	*z*	U_{eq}
C1	4219(9)	11304(8)	3897(3)	36(3)
C2	4698(8)	10045(3)	3941(3)	39(4)
C3	3536(8)	9038(7)	3807(3)	35(3)
C4	2575(9)	9189(7)	4240(4)	39(4)
C5	3271(8)	11479(7)	4331(3)	40(4)
C6	5396(9)	12247(8)	4008(4)	46(4)
C7	6891(14)	13658(11)	3659(5)	118(7)
C8	7673(15)	13453(17)	3335(8)	357(18)
C9	3551(9)	11334(8)	3355(4)	46(4)
C10	2147(8)	10433(7)	4225(3)	42(4)
N1	2903(8)	11324(7)	2946(3)	62(4)
O1	5999(6)	12459(5)	4459(3)	56(3)
O2	5677(6)	12839(6)	3581(3)	65(3)
O3	3197(6)	9187(5)	4788(2)	55(3)
O4	4468(6)	7553(5)	4270(3)	59(3)
C21	5696(9)	9910(7)	3609(4)	39(4)
C22	5762(10)	9926(8)	3046(4)	57(4)
C23	6839(12)	9860(10)	2792(4)	73(5)
C24	7966(11)	9757(10)	3079(6)	85(6)
C25	8033(11)	9721(11)	3632(5)	95(6)
C26	6952(10)	9791(8)	3892(4)	64(5)
C31	3996(9)	7819(8)	3834(4)	45(4)
C32	3856(8)	6923(8)	3338(4)	41(4)
C33	3451(9)	7192(8)	2826(4)	56(4)
C34	3340(10)	6334(9)	2382(4)	71(5)
C35	3595(11)	5200(10)	2447(5)	82(6)
C36	3988(12)	4948(10)	2959(5)	95(6)
C37	4132(10)	5790(9)	3404(4)	74(5)
C41	1370(10)	8277(8)	4126(5)	53(4)
C42	813(11)	7823(10)	3613(5)	73(5)
C43	–345(13)	7075(12)	3525(6)	92(6)
C44	–908(15)	6718(12)	3952(9)	115(9)
C45	–392(14)	7118(12)	4480(8)	110(8)
C46	773(12)	7895(9)	4560(5)	73(5)
C51	2865(9)	12717(8)	4360(4)	43(4)
C52	3466(9)	13595(8)	4758(4)	52(4)
C53	3098(10)	14734(9)	4806(5)	67(5)
C54	2139(12)	14986(9)	4451(5)	73(6)
C55	1503(11)	14110(10)	4057(5)	58(4)

TABLE IV
Characteristic data of the compounds *IV* and *V*

Compound	M.p., °C Yield, %	Formula (M.w.)	IR ν, cm^{-1}	Calculated/Found			
				% C	% H	% N	% X
IVa	145 – 146	$C_{25}H_{28}N_2O_2$	2 240 (C≡N)	77.28	7.27	7.21	–
	65	(388.2)	1 680 (C=O)	77.42	7.25	7.25	
			1 640 (C=O)				
IVb	155 – 157	$C_{28}H_{28}N_2O_2$	2 245 (C≡N)	79.21	6.65	6.60	–
	67	(424.2)	1 670 (C=O)	79.85	6.70	6.50	
			1 640 (C=O)				
IVc	154 – 156	$C_{24}H_{28}N_2O_2$	2 240 (C≡N)	76.55	7.50	7.44	–
	72	(376.2)	1 680 (C=O)	76.45	7.54	7.49	
			1 640 (C=O)				
IVd	148 – 149	$C_{23}H_{24}N_2O_2$	2 240 (C≡N)	76.63	6.72	7.78	–
	75	(360.2)	1 670 (C=O)	76.85	6.60	7.81	
			1 645 (C=O)				
IVe	168 – 170	$C_{25}H_{28}N_2O_2$	2 245 (C≡N)	77.28	6.72	7.21	–
	68	(388.2)	1 680 (C=O)	77.78	7.07	6.93	
			1 640 (C=O)				
Va	234 – 235	$C_{43}H_{33}Br_2NO_4$	3 480 (OH)	65.73	4.24	1.78	20.10
	65	(787.6)	2 250 (C≡N)	65.40	4.08	2.08	19.97
			1 740 (C=O)				
			1 660 (C=O)				
Vb	216 – 218	$C_{35}H_{27}Br_2Cl_2NO_4$	3 490 (OH)	55.78	3.61	1.68	–
	71	(753.0)	2 245 (C≡N)	55.62	3.59	1.66	
			1 740 (C=O)				
			1 665 (C=O)				
Vc	160 – 161	$C_{45}H_{39}NO_6$	3 480 (OH)	78.34	5.70	2.03	–
	69	(689.3)	2 250 (C≡N)	78.17	5.77	1.68	
			1 740 (C=O)				
			1 660 (C=O)				
Vd	193 – 195	$C_{35}H_{31}NO_4$	3 485 (OH)	79.36	5.90	2.65	–
	70	(529.2)	2 245 (C≡N)	79.66	6.01	2.65	
			1 745 (C=O)				
			1 660 (C=O)				
Ve	207 – 208	$C_{44}H_{37}NO_4$	3 485 (OH)	82.08	5.80	2.18	–
	68	(643.3)	2 240 (C≡N)	82.53	5.96	2.07	
			1 740 (C=O)				
			1 655 (C=O)				

TABLE IV
(*Continued*)

Compound	M.p., °C Yield, %	Formula (M.w.)	IR ν, cm^{-1}	Calculated/Found			
				% C	% H	% N	% X
Vf	243 – 245 72	$C_{42}H_{29}Cl_4NO_4$ (751.1)	3 485 (OH) 2 245 (C≡N) 1 745 (C=O) 1 660 (C=O)	67.10 67.23	3.89 4.09	1.86 1.76	18.62 18.68
Vg	221 – 223 69	$C_{42}H_{31}Br_2NO_4$ (771.1)	3 480 (OH) 2 240 (C≡N) 1 740 (C=O) 1 665 (C=O)	65.36 65.84	4.05 4.16	1.82 1.80	20.47 20.75
Vh	208 – 209 70	$C_{36}H_{33}NO_4$ (543.2)	3 485 (OH) 2 240 (C≡N) 1 740 (C=O) 1 665 (C=O)	79.52 79.43	6.12 6.29	2.58 2.60	–
Vi	223 – 224 68	$C_{46}H_{41}NO_8$ (735.3)	3 480 (OH) 2 245 (C≡N) 1 745 (C=O) 1 660 (C=O)	75.09 75.64	5.62 5.73	1.90 1.91	–

The ^{13}C NMR spectra exhibit a signal at δ 203 – 205 ppm due to the aroyl carbonyl group. The cyano carbon appeared at around δ 117.0 ppm. Another signal at δ 166.0 ppm appeared due to the carbethoxy carbonyl group carbon in addition to the remaining aliphatic carbons at δ (ppm) 74.9, 62.1, 52.7, 49.5, 45.4, and 42.0.

EXPERIMENTAL

Cyanoacetylurea and cyanoacetylpiperidine were purchased from Aldrich Chemical Co. Reagent quality solvents were used without further purification. IR spectra were recorded as a KBr disc using a Pye–Unicam SP3-100 instrument. ^1H and ^{13}C NMR spectra (δ, ppm; *J*, Hz) were run in $CDCl_3$ using a Bruker WP 80-SY instrument with TMS as internal standard. Compounds were analyzed at M-H-W Laboratories, Phoenix, Arizona, U.S.A. Melting points were determined on an Electrothermal melting point apparatus and were uncorrected.

The single crystal X-ray crystallographic analysis of the compound *Vd* was carried out on a Nicolet R3 m/V 4-circle X-ray diffractometer. The crystal system was triclinic with space group P_1, a = 10.4835 Å, b = 11.0533 Å, c = 24.1180 Å; α = 94.767°, β = 95.853°, γ = 98.144°. The selected

TABLE V
^{1}H NMR spectra (δ, ppm; *J*, Hz) of the compound *IV* and *V*

Compound	^{1}H NMR spectrum
IVa	1.59 m, 6 H (H′-3,H-4′,H-5′); 2.28 s, 3 H ($ArCH_3$); 2.38 s, 3 H ($ArCH_3$); 3.45 m, 4 H (H-2′,H-6′); 3.76 m, 2 H (H-2,H-3); 4.06 dd, 2 H, *J* = 3.6 (H-4); 7.25 – 8.05 m, 8 H (ArH)
IVb	1.58 m, 6 H (H-3′,H-4′,H-5′); 2.27 s, 3 H ($ArCH_3$); 3.46 m, 4 H (H-2′,H-6′); 3.90 m, 2 H (H-2,H-3); 4.10 dd, 2 H, *J* = 4.0 (H-4); 7.15 – 8.50 m, 11 H (ArH)
IVc	1.65 m, 6 H (H-3′,H-4′,H-5′); 2.31 s, 3 H ($ArCH_3$); 3.36 m, 4 H (H-2′,H-6′); 3.80 m, 2 H (H-2,H-3); 4.44 dd, 2 H, *J* = 3.6 (H-4); 7.10 – 8.00 m, 9 H (ArH)
IVd	1.58 m, 6 H (H-3′,H-4′,H-5′); 3.50 m, 4 H (H-2′,H-6′); 3.78 m, 2 H (H-2,H-3); 4.09 m, 2 H (H-4); 7.05 – 8.10 m, 10 H (ArH)
IVe	1.59 m, 6 H (H-3′,H-4′,H-5′); 2.28 s, 3 H ($ArCH_3$); 3.46 m, 4 H (H-2′,H-6′); 3.73 m, 2 H (H-2,H-3); 3.85 s, 3 H ($ArCH_3$); 4.06 dd, 2 H, *J* = 3.8 (H-4); 6.80 – 8.00 m, 8 H (ArH)
Va	0.85 t, 3 H, *J* = 7.2 (CH_3); 2.25 dd, 1 H, *J* = 13.3, *J* = 3.2 (H-5); 3.02 dt, 1 H, *J* = 14.4, *J* = 2.5, *J* = 13.0 (H-5); 3.86 q, 2 H, *J* = 7.2 (CH_2); 4.38 dd, 2 H, *J* = 14.2, *J* = 13.2, *J* = 2.6 (H-2,H-6); 5.13 d, 1 H, *J* = 12.3 (H-3); 5.51 d, 1 H (OH); 6.69 – 8.06 m, 22 H (ArH)
Vb	0.95 t, 3 H, *J* = 7.2 (CH_3); 2.02 dd, 1 H, *J* = 13.6, *J* = 3.3 (H-5); 3.09 dt, 1 H, *J* = 14.3, *J* = 2.6, *J* = 13.1 (H-5); 3.95 q, 2 H, *J* = 7.1 (CH_2); 4.22 dd, 2 H, *J* = 14.2, *J* = 13.1, *J* = 2.6 (H-2,H-6); 5.60 d, 1 H, *J* = 12.2 (H-3); 5.85 d, 1 H (OH); 7.08 – 7.70 m, 16 H (ArH)
Vc	0.80 t, 3 H, *J* = 7.2 (CH_3); 2.21 dd, 1 H, *J* = 13.2, *J* = 3.2 (H-5); 3.06 dt, 1 H, *J* = 14.3, *J* = 2.5, *J* = 13.1 (H-5); 3.40 s, 3 H (OCH_3); 3.75 s, 3 H (OCH_3); 3.80 q, 2 H, *J* = 7.2 (CH_2); 4.37 dd, 2 H, *J* = 13.2, *J* = 2.5 (H-2,H-6); 5.14 d, 1 H, *J* = 12.2 (H-3); 5.55 d, 1 H (OH); 6.39 – 8.06 m, 22 H (ArH)
Vd	0.75 t, 3 H, *J* = 7.1 (CH_3); 2.17 dd, 1 H, *J* = 13.2, *J* = 3.1 (H-5); 2.97 dt, 2 H, *J* = 14.3, *J* = 2.5, *J* = 13.1 (H-5); 3.73 q, 2 H, *J* = 7.1 (CH_2); 4.29 dd, 2 H, *J* = 14.2, *J* = 13.2, *J* = 2.6 (H-2,H-6); 4.92 d, 1 H, *J* = 12.3 (H-3); 5.32 d, 1 H (OH); 7.34 – 8.20 m, 20 H (ArH)
Ve	1.97 s, 3 H ($ArCH_3$); 2.24 dd, 1 H, *J* = 13.1, *J* = 3.2 (H-5); 2.30 s, 3 H ($ArCH_3$); 3.10 dt, 1 H, *J* = 14.3, *J* = 2.6, *J* = 13.1 (H-5); 3.32 s, 3 H (OCH_3); 4.42 dd, 2 H, *J* = 14.1, *J* = 13.1, *J* = 2.5 (H-2,H-6); 5.16 d, 1 H, *J* = 12.2 (H-3); 5.48 d, 1 H, *J* = 2.5 (OH); 6.69 – 8.06 m, 22 H (ArH)

TABLE V
(*Continued*)

Compound	^{1}H NMR spectrum
Vf	2.24 dd, 1 H, *J* = 13.1, *J* = 3.2 (H-5); 3.10 dt, 1 H, *J* = 14.3, *J* = 2.5, *J* = 13.1 (H-5); 3.30 s, 3 H (OCH_3); 4.42 dd, 2 H, *J* = 14.1, *J* = 13.2, *J* = 2.6 (H-2,H-6); 5.18 d, 1 H, *J* = 12.1 (H-3); 5.46 d, 1 H, *J* = 2.5 (OH); 6.70 – 8.10 m, 20 H (ArH)
Vg	2.22 dd, 1 H, *J* = 13.2, *J* = 3.2 (H-5); 3.11 dt, 1 H, *J* = 14.3, *J* = 2.6, *J* = 13.1 (H-5); 3.33 s, 3 H (OCH_3); 4.45 dd, 2 H, *J* = 14.1, *J* = 13.2, *J* = 2.6 (H-2,H-6); 5.19 d, 1 H, *J* = 12.1 (H-3); 5.52 d, 1 H, *J* = 2.5 (OH); 6.90 – 8.20 m, 22 H (ArH)
Vh	2.24 dd, 1 H, *J* = 13.1, *J* = 3.2 (H-5); 2.09 s, 3 H ($ArCH_3$); 2.35 s, 3 H ($ArCH_3$); 3.12 dt, 1 H, *J* = 14.2, *J* = 2.5, *J* = 13.2 (H-5); 3.32 s, 3 H (OCH_3); 4.40 dd, 2 H, *J* = 14.3, *J* = 13.2, *J* = 2.5 (H-2,H-6); 5.16 d, 1 H, *J* = 12.1 (H-3); 5.48 d, 1 H, *J* = 2.5 (OH); 6.65 – 8.25 m, 18 H (ArH)
Vi	2.25 dd, 1 H, *J* = 3.3, *J* = 3.1 (H-5); 3.10 dt, 1 H, *J* = 14.1, *J* = 2.6, *J* = 13.2 (H-5); 3.34 s, 3 H (OCH_3); 3.57 s, 6 H (OCH_3); 3.85 s, 3 H (OCH_3); 3.91 s, 3 H (OCH_3); 4.45 dd, 2 H, *J* = 14.1, *J* = 13.2, *J* = 2.5 (H-2,H-6); 5.10 d, 1 H, *J* = 12.2 (H-3); 5.40 d, 1 H, *J* = 2.6 (OH); 6.75 – 8.20 m, 20 H (ArH)

bond angles are listed in Table I. Figure 1 shows X-ray plot of the compound *Vd*. The bond lengths, the atomic coordinates and equivalent isotropic displacement parameters are listed in Tables II and III. The relative configuration of each of the five chiral carbons in the cyclohexane ring was also determined from the X-ray molecular picture.

(1*S**, 2*S**, 3*R**, 4*S**, 5*R**)-(±)-3-Aroyl-2,4,6-triaryl-1-cyano-4-hydroxycyclohexane Carboxylates *V* and *N*-(3,5-Diaryl-2-cyano-5-oxopentanoyl)piperidine *IV*. General Procedure

To a solution of the appropriate sodium alkoxide (0.01 mol) in anhydrous alcohol (150 ml) containing cyanoacetylurea or cyanoacetylpiperidine (0.01 mol) was added chalcone (0.02 mol for the urea derivative or 0.01 mol for the piperidine derivative). The mixture was stirred at room temperature for 12 – 18 h, the solid formed was filtered and crystallized from glacial acetic acid. The filtrate was poured into water (150 ml), the organic layer separated, dried by anhydrous sodium sulfate and evaporated to give the crude unreacted starting materials.

The characteristic data of the compounds *IV* and *V* are given in Table IV. For the ^{1}H NMR spectra see Table V.

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