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Fast, Facile and Convenient Synthesis of α, α-Bis(substituted-arylidene) Cycloalkanones: An improved Protocol

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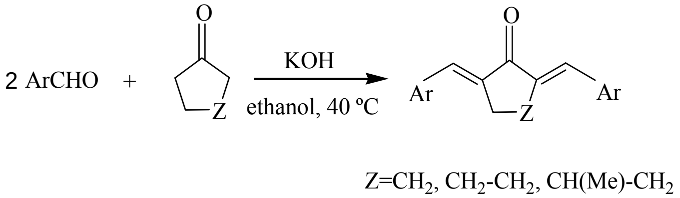
Abstract: Commercially available potassium hydroxide (KOH), a simple base, was found to be a catalyst for tandem cross-aldol condensation between cyclic ketones and aromatic aldehydes leading to a fast and easy synthesis of α,α-bis (substituted-arylidene) cycloalkanones in the shortest times of all previous methods. The reaction of aryl aldehydes with five and six-membered cyclic ketones afforded excellent yields after few seconds in most cases. The reaction conditions were compatible with various electron withdrawing and electron donating substituents, *e.g.* Cl, F, NO_2, OMe, Me and NMe_2.

Keywords: Aromatic aldehyde, Cyclic ketone, α,α-Bis(substituted-arylidene) cycloalkanones

Introduction

Arylidene cycloalkanones are useful intermediates for synthesis of bioactive pyrimidine[1], liquid crystalline polymers[2], perfumes[3], bis-spiropyrrolidines[4] and bis-spiroisoxazolines[5].

Usually, the aldol reaction is catalyzed by strong acid or base, however, the yield is low in common due to reverse and side-reactions[6]. Therefore, much effort on improvement of the reaction yield were performed, the main progress was the following: (1) Using Lewis[7] acid such as $RuCl_3$, SmI_3[8], BF_3/Et_2O[9,10], $FeCl_3/6H_2O$[11], $Mg(HSO4)_2$[12], $Yb(OTf)_3$[13], $InCl_3/4H_2O$[14], $LiClO_4$[15], SSA[16], NKC-9 (polymer-supported sulfonic acid)[17], SiO_2–Pr–SO_3H[18], CH_3CO_2Na/CH_3CO_2H[19] as catalyst promoted the aldol reaction. (2) The aldol reaction was catalyzed by organometallic complexes but the yields were not satisfactory[20] or required long reaction time[21]. And (3) other catalyst such as I_2[22], TMSCl/NaI[23], LiOH[24], KF/Al_2O_3[25], BMPTO[26], NaOH in micellar medium[27], KOH[28] and Na-HAP[29] were also found to accelerate this type reaction. However, the use of toxic reagents, long reaction time, low yields, formation of a mixture of products and tedious separation procedures are among the drawbacks of the reported methods. In continuation[30] of synthesis of these compounds in our research group, here we want to report an easy efficient and selective method for condensation of cyclic ketones with aromatic aldehydes under soft conditions in the presence of KOH as a cheap and commercially available catalyst (Scheme 1).



Scheme 1

Experimental

To a 10 mL solution of KOH (0.11 g) in ethanol at 40 ºC in a round bottom flask, cyclic ketone (5 mmol) and aromatic aldehyde (10 mmol) was added and the mixture was stirred for 5-120 s. The resulting product was then isolated by simple filtration from the reaction mixture and given washings with water to remove any trace of KOH remaining on the product.

Results and Discussion

Synthesis of *α,ά*-bis(substituted benzylidene)cycloalkanones using KOH as catalyst already reported in chemistry literature[24,28] but in all of them time is long (Table 1), yield in some cases is moderate or low (Table 1) and in all of reported paper for synthesis of *α,ά*-bis(substituted benzylidene)cycloalkanones number of synthesized products are limited. It is therefore important to find a more convenient method for improve this protocol for the synthesis of cyclic chalcones.

Table 1. Comparison of our results with results obtained by other groups

Catalyst	Conditions	No. of Cyclic Chalcon	Time	Yield, %[c]	Ref.
SiO_2–Pr–SO_3H	Oven/80 ºC/ heat	15	10-180 min	66-100	[18]
KF/Al_2O_3	MW/450 Watt	11	2-5 min	75-90	[25]
SmI_3	N_2 atmosphere/Sm/ I_2/r.t-60 ºC/ stirring	12	3-4.5 h	80-98	[8]
NKC-9	$CHCl_3$/80 ºC/ stirring	15	4-6 h	75-94	[17]
NaOH	CTAB/40 ºC/ stirring	8	6-8 h	80-98	[27]
$LiClO_4$	$TMSNEt_2$/r.t/ stirring in inert atmosphere	16	7 min	86-99	[15]
I_2	CH_2Cl_2/r.t/ Stirring	21	4.5-9.5 h	89-95	[22]
SSA	Solvent-free/80 ºC/heat	15	2-24 h	10-97	[16]
LiOH	EtOH/30 ºC/ stirring	30	2-75 min	75-98	[24]
KOH	EtOH/30 ºC/ stirring	Tested	1 h	Trace	[24]
KOH	EtOH/30 ºC/ stirring	17	6-12 h	65-92	[28]
KOH	EtOH/40 ºC/ stirring	40	5-120 s	86-99	This work

The model reaction studied was the condensation of cyclopentanone with 4-methoxybenzaldehyde to yield a bisubstituted cyclohexanone (Scheme 1). When the mixture of cyclopentanone and 4-methoxybenzaldehyde was stirred at room temperature in presence of KOH in water (5, 10, 15 mL), only a trace amount of the product was observed. However, when solvent changed to ethanol, the yield of the product increased to 20, 45 and 30 % for 5, 10, 15 mL ethanol sequentially. The same reaction was also carried out in presence of 10 mL ethanol which gave 98% yield at 40 ºC in 5 s (Table 2, Entry 2). Thus, it can be said that amount of solvent can be improved yield of product and 10 mL of ethanol is the best.

Next, we also examined scope of the reaction by using various aryl aldehydes (electron-donating and electron-withdrawing groups) and cyclic ketones (cyclopentanone, cyclohexanone, 4-methyl cyclohexanone). The results are summarized in Table 2. In general, high yields of cross-aldol products were obtained in ethanol at 40 ºC for 5-120 s.

Table 2. Preparation of *α,ά*-bis(substituted arylidene)cycloalkanones catalyzed by KOH at 40 °C[a,b]

Entry	Ar		Time, s	Yield, %	mp, ºC	Lit. mp, ºC
1	C_6H_5	CH_2	5	98	188-191	188-191
2	4-MeO-C_6H_4	CH_2	5	98	213-215	212-215
3	3-MeO-C_6H_4	CH_2	5	96	145-147	Not reported
4	4-Me-C_6H_4	CH_2	5	98	245-247	242-244
5	2-Cl-C_6H_4	CH_2	5	99	157-160	154-156
6	4-Cl-C_6H_4	CH_2	5	99	223-226	228-229
7	2,4-Cl_2-C_6H_3	CH_2	5	98	219-220	206-207
8	2,3-Cl_2-C_6H_3	CH_2	5	95	210-212	Not reported
9	2,6-Cl_2-C_6H_3	CH_2	5	96	189-191	Not reported
10	3-NO_2-C_6H_4	CH_2	5	93	224-226	Not reported
11	PhCH=CH_2	CH_2	5	94	228-230	226-228
12	4-Me_2N-C_6H_4	CH_2	5	96	270-272	271-273
13	C_6H_5	CH_2CH_2	20	97	115-118	115-118
14	4-MeO-C_6H_4	CH_2CH_2	20	98	160-161	162-164
15	3-MeO-C_6H_4	CH_2CH_2	60	97	80-84	Not reported
16	4-Me-C_6H_4	CH_2CH_2	30	92	168-171	170-171
17	3-Me-C_6H_4	CH_2CH_2	120	90	75-78	Not reported
18	2-Cl-C_6H_4	CH_2CH_2	60	93	108-110	102-104
19	4-Cl-C_6H_4	CH_2CH_2	60	94	147-149	147-148
20	4-Br-C_6H_4	CH_2CH_2	5	95	165-167	164-165
21	2,4-Cl_2-C_6H_3	CH_2CH_2	5	93	164-165	163-164
22	2,3-Cl_2-C_6H_3	CH_2CH_2	5	98	183-186	Not reported
23	2,6-Cl_2-C_6H_3	CH_2CH_2	120	97	182-185	Not reported
24	2-Cl-6-F-C_6H_3	CH_2CH_2	120	97	122-124	Not reported
25	4-NO_2-C_6H_4	CH_2CH_2	20	96	208-210	160-163
26	3-NO_2-C_6H_4	CH_2CH_2	60	90	193-196	195-197
27	2-NO_2-C_6H_4	CH_2CH_2	120	90	151-154	158-159
28	PhCH=CH_2	CH_2CH_2	5	90	188-190	182-184
29	2-Naphthyl	CH_2CH_2	120	89	177-179	199-202

Contd...

30	4-Me_2N-C_6H_4	CH_2CH_2	10	95	252-254	250-252
31	C_6H_5	CH(Me)CH_2	120	88	90-95	97-99
32	4-MeO-C_6H_4	CH(Me)CH_2	120	90	144-146	137-139
33	4-Me-C_6H_4	CH(Me)CH_2	120	88	130-132	125-127
34	2-Cl-C_6H_4	CH(Me)CH_2	120	97	121-123	Not reported
35	4-Cl-C_6H_4	CH(Me)CH_2	30	96	151-157	156-160
36	2,4-Cl_2-C_6H_3	CH(Me)CH_2	30	97	144-146	219-222
37	2-Cl-6-F-C_6H_3	CH(Me)CH_2	30	98	112-114	Not reported
38	3-NO_2-C_6H_4	CH(Me)CH_2	120	92	164-167	Not reported
39	PhCH=CH_2	CH(Me)CH_2	120	86	153-157	Not reported
40	2-Naphthyl	CH(Me)CH_2	120	87	155-160	Not reported

[a]Stirred at 40 °C for the time given. [b]All the known products were characterized from melting point and spectral (IR, ^{1}H NMR) data also all the unknown products were characterized from spectral (IR, ^{1}H NMR, ^{13}C NMR) and analytical data

Spectral data for unknown compounds

Entry 3: IR (KBr): 3065, 2934, 2834, 1692, 1626, 1606, 1572, 1491, 1271, 1223, 1185 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.56 (2H, sbr, 2CH=), 7.35-7.38 (2H, Ar), 7.12-7.21 (4H, Ar), 6.94-6.96 (2H, Ar), 3.85 (6H, s, OMe), 3.09 (4H, s, $2CH_2$); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 26.53, 55.30, 115.10, 116.02, 123.31, 129.73, 133.78, 137.12, 137.54, 159.72, 196.24; Anal. Calcd for $C_{21}H_{20}O_3$: C, 78.73; H, 6.29; Found: C, 78.04; H, 6.57. **Entry 8**: IR (KBr): 3033, 2922, 2881, 1682, 1597, 1555, 1451, 1406, 1151, 777, 681 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.91 (2H, sbr, 2CH=), 7.49-7.52 (2H, Ar), 7.44-7.46 (2H, Ar), 7.27-7.31 (2H, Ar), 2.99 (4H, s, $2CH_2$); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 26.46, 127.02, 128.11, 130.31, 130.85, 133.95, 134.00, 136.02, 140.01, 195.17; Anal. Calcd for $C_{19}H_{12}Cl_4O$: C, 57.32; H, 3.04; Found: C, 57.83; H, 3.31. **Entry 9**: IR (KBr): 3063, 2931, 2856, 1704, 1633, 1554, 1426, 1237, 1177, 770 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.53 (2H, sbr, 2CH=), 7.35-7.38 (4H, Ar), 7.22-7.28 (2H, Ar), 2.64 (4H, s, $2CH_2$); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 25.60, 128.16, 128.80, 129.77, 133.74, 134.51, 143.12, 194.20; Anal. Calcd for $C_{19}H_{12}Cl_4O$: C, 57.32; H, 3.04; Found: C, 58.12; H, 3.46. **Entry 10**: IR (KBr): 3063, 2923, 2884, 1687, 1604, 1529, 1349, 1187 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.49 (2H, sbr, 2CH=), 8.20-8.27 (2H, Ar), 7.85-7.93 (2H, Ar), 7.68-7.78 (4H, Ar), 3.26 (4H, s, $2CH_2$); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 26.39, 123.83, 123.96, 124.26, 124.54, 129.94, 131.76, 136.48, 137.12, 139.24, 148.59; Anal. Calcd for $C_{19}H_{14}N_2O_5$: C, 65.14; H, 4.03; Found: C, 65.67; H, 4.43. **Entry 15**: IR (KBr): 3065, 2937, 2835, 1667, 1597, 1487, 1430, 1291, 1242, 1166, 1047 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.79 (2H, sbr, 2CH=), 6.90-7.36 (8H, Ar), 3.85 (6H, s, OMe), 2.94 (4H, m, $2CH_2$), 1.80 (2H, m, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 22.97, 22.52, 55.29, 114.25, 115.81, 122.88, 129.40, 136.45, 136.85, 137.30, 159.47, 190.22; Anal. Calcd for $C_{22}H_{22}O_3$: C, 79.02; H, 6.63; Found: C, 78.83; H, 6.83. **Entry 17**: IR (KBr): 3054, 2924, 2863, 1667, 1484, 1279, 1227,1157 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.82 (2H, sbr, 2CH=), 7.29-7.35 (6H, Ar), 7.18-7.20 (2H, Ar), 2.96 (4H, m, $2CH_2$), 2.43 (6H, s, Me), 1.82 (2H, m, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 21.50, 23.08, 28.56, 127.47, 128.29, 129.43, 131.15, 136.00, 136.11, 137.10, 138.01, 190.42; Anal. Calcd for $C_{22}H_{22}O$: C, 87.38; H, 7.33; Found: C, 88.08; H, 7.22. **Entry 22**: ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.87 (2H, sbr, 2CH=), 7.45-7.49 (2H, Ar), 7.24-7.29 (4H, Ar), 2.75 (4H, m, $2CH_2$), 1.77 (2H, m, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 23.03, 28.31, 126.84, 128.58, 130.22, 132.99, 133.62 134.10, 136.60, 138.20, 189.40; Anal. Calcd for $C_{20}H_{14}Cl_4O$: C, 58.29; H, 3.42; Found: C, 58.18; H, 3.81. **Entry 23**: IR (KBr): 3056, 2943, 2895, 1678, 1623, 1605, 1555, 1427, 1302, 1265, 1141, 782 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.61 (2H, sbr, 2CH=), 7.20-7.37 (6H, Ar), 2.47 (4H, m, 2CH$_2$), 1.74 (2H, m, CH$_2$); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 21.97, 28.24, 127.98, 129.48, 132.29, 134.17, 134.45, 140.16, 188.04; Anal. Calcd for $C_{20}H_{14}Cl_4O$: C, 58.29; H, 3.42; Found: C, 59.08; H, 3.63. **Entry 24**: IR (KBr): 3045, 2936, 2915, 1679, 1621, 1568, 1444, 1307, 1265, 913, 782 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.60 (2H, sbr, 2CH=), 7.03-7-39 (6H, Ar), 2.54 (4H, m, 2CH$_2$), 1.73 (2H, m, CH$_2$); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 22.41, 28.70, 28.75, 114.16, 114.39, 123.34, 123.53, 125.30, 125.33, 128.21, 130.03, 130.12, 135.29, 135.34, 140.69, 158.51, 161.01, 188.24; Anal. Calcd for $C_{20}H_{14}Cl_2F_2O$: C, 63.34; H, 3.72; Found: C, 63.56; H, 3.38. **Entry 34**: IR (KBr): 3065, 2955, 2926, 1673, 1613, 1468, 1435, 1298, 1238, 1145, 1051, 755 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.95 (2H, sbr, 2CH=), 7.29-7.46 (8H, Ar), 2.86 (2H, m, 2H of CH$_2$), 2.42 (2H, m, 2H of CH$_2$), 1.89 (1H, m, CH), 1.02 (3H, d, J = 6.4 Hz, Me); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 21.50, 29.64, 36.42, 126.38, 129.69, 129.77, 130.62, 134.35, 135.06, 136.90, 189.45; Anal. Calcd for $C_{21}H_{18}Cl_2O$: C, 70.60; H, 5.08; Found: C, 71.10; H, 5.73. **Entry 37**: IR (KBr): 3065, 2956, 2926, 1679, 1621, 1567, 1445, 1299, 1246, 1143, 911, 782 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.61 (2H, sbr, CH=), 7.25-7.32 (4H, Ar), 7.04-7.08 (2H, Ar), 2.55 (2H, m, 2H of CH$_2$), 2.30 (2H, m, 2H of CH$_2$), 1.91 (1H, m, CH), 0.97 (3H, d, J= 6.4 Hz, Me); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 21.24, 28.76, 36.63, 36.68, 114.20, 114.43, 123.34, 123.53, 125.31, 125.35, 128.52, 130.05, 130.15, 135.29, 135.33, 139.85, 158.50, 161.00, 188.02; Anal. Calcd for $C_{21}H_{16}Cl_2F_2O$: C, 64.14; H, 4.10; Found: C, 64.03; H, 4.25. **Entry 38**: IR (KBr): 3085, 2925, 2884, 1672, 1611, 1528, 1349, 1299, 1196, 1147 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 8.29 (2H, sbr, 2CH=), 8.20-8.22 (2H, Ar), 7.61-7.81 (6H, Ar), 3.05 (2H, dd, J = 3.6, 16.0 Hz, 2H of 2CH$_2$), 2.62 (2H, ddd, J = 2.4, 12.0, 16.0 Hz, 2H of 2CH$_2$), 1.98 (1H, m, CH), 1.12 (3H, d, J = 6.8 Hz, Me); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 21.45, 29.10, 36.08, 123.29, 124.55, 129.57, 134.90, 136.08, 137.20, 137.29, 148.25, 188.95; Anal. Calcd for $C_{21}H_{18}N_2O_5$: C, 66.66; H, 4.79; Found: C, 67.13; H, 4.54. **Entry 39**: IR (KBr): 3031, 2927, 2879, 1655, 1609, 1582, 1295, 1212, 1148, 971 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 6.98-8.55 (16H, Ar & 6CH=), 3.02 (2H, dd, J = 3.2, 15.8 Hz, 2H of 2CH$_2$), 2.35 (2H, m, 2H of CH$_2$), 2.01 (1H, m, CH), 1.21 (3H, d, J = 6.4 Hz, Me); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 21.75, 28.45, 34.79, 123.71, 127.24, 128.84, 128.90, 134.70, 136.56, 136.77, 140.83, 188.74; Anal. Calcd for $C_{25}H_{24}O$: C, 88.20; H, 7.11; Found: C, 87.89; H, 7.04. **Entry 40**: IR (KBr): 3054, 2953, 2923, 1665, 1605, 1503, 1296, 1227, 1145, 1004 cm^{-1}; ^{1}H NMR (400 MHz, CDCl$_3$): δ 8.09 (2H, sbr, CH=), 7.54-8.04 (14H, Ar), 3.22 (2H, dd, J = 3.6, 15.8 Hz, 2H of CH$_2$), 2.68 (2H, ddd, J = 2.4, 11.4, 15.2 Hz, 2H of CH$_2$), 1.98 (1H, m, CH), 1.13 (3H, d, J = 6.8 Hz, Me); ^{13}C NMR (100 MHz, CDCl$_3$): δ= 21.69, 29.52, 36.66, 126.49, 126.92, 127.70, 127.72, 127.86, 127.99, 128.50, 130.32, 133.11, 133.14, 133.59, 135.69, 137.36, 190.08; Anal. Calcd for $C_{29}H_{24}O$: C, 89.66; H, 6.23; Found: C, 90.11; H, 6.36.

Conclusion

In conclusion, we have developed a easy, effective, quick and cost-effective procedure for the synthesis of α,α-bis(substituted-arylidene) cycloalkanones using commercially available KOH under mild conditions. Moreover, the high yield of products, no work-up need, ready availability of the catalyst, compatibility with various functional groups and the clean procedure, will make the present method a useful and important addition to the present methodologies for this synthesis.

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