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Atom-efficient cross-coupling reactions of triarylbismuths with acyl chlorides under Pd(0) catalysis

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Abstract—The atom-efficient cross-coupling reaction of triarylbismuths with a variety of aliphatic, aromatic, and hetero-aromatic acyl chlorides was demonstrated to afford high yields of cross-coupled ketones under palladium catalysis. The corresponding cross-coupling reaction with diacid chlorides also furnished bis-coupled ketones in good yields.

1. Introduction

The importance of metal catalyzed reactions in organic synthesis is unparalleled by numerous applications of these methods in organic synthesis.[1–4] Ketones in general are important functionality found in several natural products and in various pharmaceutical products. Straightforward synthesis of isomeric ketones is often problematic under Friedel–Crafts acylation procedures.[5] In this context, the metal catalyzed organometallic coupling methodology outweighs other approaches for its flexibility to synthesize isomeric ketones under mild conditions. Organometallic reagents such as organo-boron, -tin, -zinc, -indium, -magnesium, and -lithiums are known to furnish ketones with acyl halides under metal catalysis.[6,7] The carbonylative cross-coupling of organometallic reagents with electrophilic partners is another alternative to produce ketones.[8] However, the drawback associated with this methodology is that the carbonyl insertion into ArPdX intermediate is often dictated by electronics of the Ar group, thus limiting its utility for the synthesis of a variety of divergent functionalized ketones.

Hence, the cross-coupling reaction of acyl chlorides with organometallic reagents is undoubtedly a popular route for the synthesis of functionalized ketones. The high reactive nature of acyl chlorides compared to other acyl donors is an additional advantage for efficient coupling with organometallic nucleophilic partner in the presence of metal catalyst. However, reactivity studies utilizing alternative acyl donors were also been reported recently for cross-coupling reactions with organometallic reagents under different conditions.[6b,c,9]

The recent interest in organobismuths is due to its non-toxicity and high reactivity that led to several applications of these reagents in organic synthesis.[10–14] As part of the pioneering efforts, Barton et al. have reported studies involving organobismuth reagent under metal catalyzed or mediated conditions. Importantly, organobismuth compounds can be easily accessed through standard procedures.[10] Notably, many studies were recently reported involving organobismuths for C–N and C–O bond formations, while the corresponding studies for C–C bond formations are scarce. However, use of organobismuths for various other applications in organic synthesis and in material applications are also reported.[10a] In the present context, Barton et al. in their seminal studies first reported the cross-coupling reaction of triphenylbismuth with acyl chlorides in the presence of $Pd(OAc)_2$ catalyst precursor.[12a] As part of our interest in the development of reactions involving organobismuth reagents for organic synthesis,[13] we recently have demonstrated the high cross-coupling reactivity of various triarylbismuths with a variety of acyl chlorides under palladium catalyzed conditions.[13a] During these studies and also from the studies reported by Barton et al., it was realized that the triethylamine base is beneficial for effective cross-coupling of acyl chlorides with triarylbismuths under palladium catalysis. Barton et al. also proposed the catalytic cycle for this cross-coupling reaction involving Pd(0), which is expected to form during in situ reduction of Pd(II).[12a] Thus, the subsequent oxidative addition of acyl chlorides to Pd(0) generates acyl palladium(II) species. Further, ligand exchange with triarylbismuth followed by reductive elimination delivers ketone with the regeneration of Pd(0). Further, during our studies, we found the superior cross-coupling reactivity of triarylbismuths with electrophilic coupling partners with the catalytic system generated from Pd(II) salts[13b,c] when compared to using directly Pd(0) catalysts[14a] with appropriate base and solvent combinations. However, it

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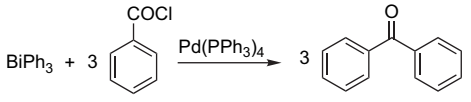
is difficult at this stage to reason out for this difference in reactivity with different catalyst precursors as not many protocols are at presently available using triarylbismuths for C–C bond formations in combination with various electrophilic coupling partners. This prompted us to further explore the cross-coupling reactivity of triarylbismuths with acyl chlorides in the presence of Pd(0) complex directly and hence a systematic study has been carried out in this direction. So, herein, we report our detailed study on $Pd(PPh_3)_4$ catalyzed atom-efficient cross-coupling reaction of triarylbismuths with a variety of acyl chlorides for efficient synthesis of a library of functionalized ketones.

2. Results and discussion

In our focused efforts to establish the cross-coupling reactivity with $Pd(PPh_3)_4$ complex, initially the cross-coupling reaction of $BiPh_3$ (1 equiv) and benzoyl chloride (3.3 equiv) was studied in different solvents and reaction conditions. As given in Table 1, the cross-coupling reaction with $Pd(PPh_3)_4$ produced poor conversion in DMF, NMP and DMA solvents (entries 1.1–1.3). Further studies revealed good cross-coupling conversion in solvents such as CH_3CN, THF, DME, and 1,4-dioxane (entries 1.4–1.7). Additional experiments carried out involving lowering the reaction temperature to 40 and 60 °C produced moderate conversions, thus revealing 80 °C is suitable for facile cross-coupling delivering high conversion to benzophenone product (entries 1.8–1.11). As shown, absence of base also produced reasonable conversion up to 71% leaving roughly one-third of acid chloride unreacted (entry 1.12). Reactions carried out to further understand the effectiveness of the other organic and inorganic bases revealed that pyridine, potassium carbonate, and cesium carbonate were ineffective to provide high conversion (entries 1.13–1.15). Notably, it was also found that the cross-coupling conversion was equally good in THF (81%), DME (90%), and with a relative high conversion in 1,4-dioxane (93%) solvent system. So, for our further study with $Pd(PPh_3)_4$ catalyst, 1,4-dioxane solvent in combination with triethylamine base was used to cross-couple 1 equiv triarylbismuth with 3 equiv of acyl chlorides. This study also revealed that the catalytic reactivity of $Pd(PPh_3)_4$ is on par with the Pd(II) catalyst precursor for cross-coupling reaction of triarylbismuths with acyl chlorides. It is worth mentioning that the cross-coupling reaction involving triarylbismuths as organometallic coupling partner is facile and more atom-efficient when compared to the similar reactivity known with organo-boron, tin, and other reagents.[7]

Table 1. Screening conditions[a]



Entry	Solvent	Base	Temp (°C)	Conv[b] (%)
1.1	DMF	Triethylamine	80	22
1.2	NMP	Triethylamine	80	32
1.3	DMA	Triethylamine	80	40
1.4	CH_3CN	Triethylamine	80	73
1.5	THF	Triethylamine	80	81
1.6	DME	Triethylamine	80	90
1.7	1,4-Dioxane	Triethylamine	80	93
1.8	1,4-Dioxane	Triethylamine	60	84
1.9	DME	Triethylamine	60	77
1.10	1,4-Dioxane	Triethylamine	40	64
1.11	1,4-Dioxane	Triethylamine	rt	54
1.12	1,4-Dioxane	None	80	71
1.13	1,4-Dioxane	Pyridine	80	62
1.14	1,4-Dioxane	K_2CO_3	80	74
1.15	1,4-Dioxane	Cs_2CO_3	80	71

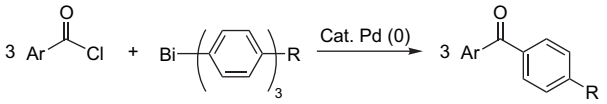
[a] Conditions, equivalent ratios with respect to $BiPh_3$: $BiPh_3$ (1 equiv), $Pd(PPh_3)_4$ (0.09 equiv), PhCOCl (3.3 equiv), base (1 equiv), 3 h.
[b] Based on GC analysis.

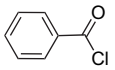
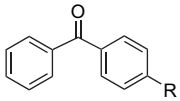
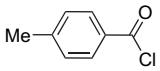
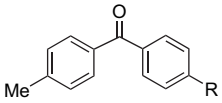
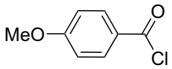
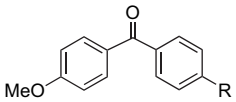
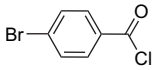
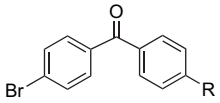
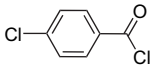
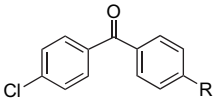
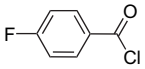
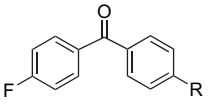
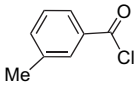
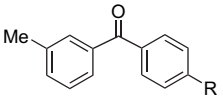
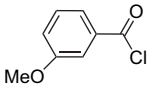
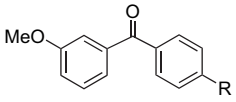
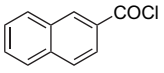
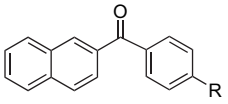
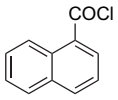
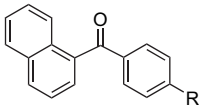
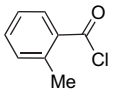
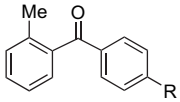
Hence, we have explored the scope and general reactivity of various other aromatic acyl chlorides under the present reaction conditions. Thus, a variety of aromatic acyl chlorides were subjected to optimized conditions with different triarylbismuths as given in Table 2. This study clearly established a general and high atom-efficient cross-coupling reactivity of triarylbismuths with a plethora of aromatic acyl chlorides. In detail, the reactions of various electronically different *ortho*-, *meta*-, and *para*-substituted aromatic acyl chlorides were found to be efficient giving high yields of the cross-coupled functionalized diaryl ketones. For example, the reactivity of *para*-substituted benzoyl chlorides with methyl, methoxy, bromo, chloro, and fluoro groups produced high yields of functionalized diaryl ketones with electronically different triarylbismuths (entries 2.1–2.18). The cross-coupling reactivity found with *p*-bromo-benzoyl chloride was chemoselective, as we have not observed the competitive cross-coupling product arising from C–Br terminus of *p*-bromobenzoyl chloride (entries 2.10–2.12). The efficient cross-coupling reactivity was also found with *meta*-substituted aromatic acyl chlorides with methyl and methoxy groups furnishing high yields of the ketones with triarylbismuths (entries 2.19–2.24). The reactivity of 1-naphthoyl chloride and 2-naphthoyl chloride with different triarylbismuths was effective giving naphthyl phenyl ketones in high yields (entries 2.25–2.30). The corresponding cross-coupling reactivity of *o*-toluoyl chloride with different $BiAr_3$ furnished moderate yields of the products probably due to sterics associated with *ortho* substitution (entries 2.31 and 2.32).

Further, we have elaborated our study with hetero-aroyl chlorides to establish their reactivity with different triarylbismuth reagents (Table 3). This study disclosed the efficient reactivity of these acyl chlorides furnishing high yields of the cross-coupled products. For instance, the coupling reaction of furan-2-carbonyl chloride with a variety of electronically different triarylbismuths substituted with *p*-methyl, *p*-methoxy, and *p*-fluoro groups produced cross-coupling products in 81–92% isolated yields (entries 3.1–3.4). The reactivity of thiophene-2-carbonyl chloride with different triarylbismuths also furnished moderate to high yields of the cross-coupled ketones (entries 3.5–3.9). Thus, the cross-coupling reactivity of hetero-aroyl chlorides with electronically divergent triarylbismuths proved to be facile and efficient under the present palladium catalyzed protocol.

To broaden the scope of this reaction further, the cross-coupling reactions of di-aroyl chlorides such as isophthaloyl dichloride and terephthaloyl dichloride were studied (Table 3). As given, the cross-coupling reactions with di-aroyl chlorides were found to be equally efficient with different triarylbismuth reagents. Thus, the reaction of isophthaloyl

Table 2. Cross-coupling reactivity of acyl chlorides with triarylbismuths[a]



Entry	ArCOCl	Ketone	R	Yield[b] (%)
2.1			**2.1a**, R=–H	91
2.2			**2.2a**, R=–CH_3	94
2.3			**2.3a**, R=–OCH_3	86
2.4			**2.4a**, R=–H	97
2.5			**2.5a**, R=–CH_3	95
2.6			**2.6a**, R=–OCH_3	79
2.7			**2.7a**, R=–H	86
2.8			**2.8a**, R=–CH_3	81
2.9			**2.9a**, R=–OCH_3	77
2.10			**2.10a**, R=–H	81
2.11			**2.11a**, R=–CH_3	88
2.12			**2.12a**, R=–OCH_3	79
2.13			**2.13a**, R=–H	94
2.14			**2.14a**, R=–CH_3	96
2.15			**2.15a**, R=–OCH_3	90
2.16			**2.16a**, R=–H	95
2.17			**2.17a**, R=–CH_3	93
2.18			**2.18a**, R=–OCH_3	85
2.19			**2.19a**, R=–H	90
2.20			**2.20a**, R=–CH_3	97
2.21			**2.21a**, R=–OCH_3	84
2.22			**2.22a**, R=–H	91
2.23			**2.23a**, R=–CH_3	93
2.24			**2.24a**, R=–OCH_3	86
2.25			**2.25a**, R=–H	98
2.26			**2.26a**, R=–CH_3	97
2.27			**2.27a**, R=–OCH_3	90
2.28			**2.28a**, R=–H	74
2.29			**2.29a**, R=–CH_3	83
2.30			**2.30a**, R=–OCH_3	72
2.31			**2.31a**, R=–H	60
2.32			**2.32a**, R=–CH_3	74

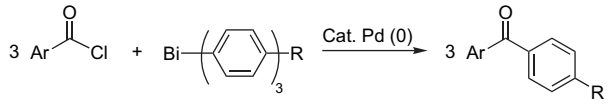
[a] Conditions for ArCOCl reactions, equivalent ratios with respect to $BiAr_3$: $BiAr_3$ (1 equiv), ArCOCl (3.3 equiv), Pd $(PPh_3)_4$ (0.09 equiv), Et_3N (1 equiv), 1,4-dioxane (3 mL), 80 °C, 3 h.

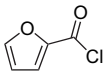
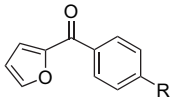
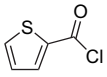
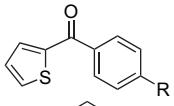
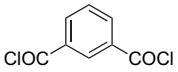
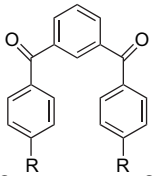
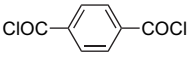
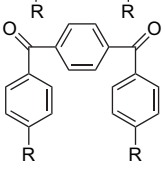
[b] Isolated yields after column chromatography. All the products were characterized by ^1H, ^{13}C NMR, IR, and mass spectral analyses.

dichloride with different triarylbismuths produced the corresponding bis-ketones in good yields (entries 3.10–3.13). The reactivity with terephthaloyl dichloride was also found to be efficient furnishing the corresponding cross-coupled bis-ketones in good yields (3.14–3.16). This study further proved the effective atom-efficient coupling reactivity of different triarylbismuths with di-aroyl chlorides for bis-couplings in one-pot operation. Importantly, the bis-coupled products are known to be very useful precursors for the synthesis of chiral cyclophanes, annularly functionalized cyclophanes, and novel photochromic applications.[15]

To gain further insights and establish the reactivity with triarylbismuths, the cross-coupling reaction of aliphatic acyl chlorides was contemplated as these compounds are relatively more reactive than aroyl chlorides. In our attempts with aliphatic acyl chlorides, it was found that the presence of a little excess amount (2 equiv) of acyl chlorides is highly beneficial for atom-efficient cross-couplings with triarylbismuths to give mixed alkyl aryl ketones in good yields. The little excess amount of acid chlorides was necessary due to low boiling and high volatile nature of some of the acyl chlorides employed. So, in the reactions involving aliphatic acyl

Table 3. Cross-coupling reactivity of hetero-aroyl and di-aroyl chlorides with triarylbismuths[a,b]



Entry	ArCOCl	Ketone	R	Yield[c] (%)
3.1			**3.1a**, R=–H	81
3.2			**3.2a**, R=–CH_3	92
3.3			**3.3a**, R=–OCH_3	84
3.4			**3.4a**, R=–F	86
3.5			**3.5a**, R=–H	77
3.6			**3.6a**, R=–CH_3	91
3.7			**3.7a**, R=–OCH_3	85
3.8			**3.8a**, R=–F	82
3.9			**3.9a**, R=–Cl	56
3.10			**3.10a**, R=–H	85
3.11			**3.11a**, R=–CH_3	80
3.12			**3.12a**, R=–OCH_3	78
3.13			**3.13a**, R=–F	76
3.14			**3.14a**, R=–H	82
3.15			**3.15a**, R=–CH_3	75
3.16			**3.16a**, R=–F	73

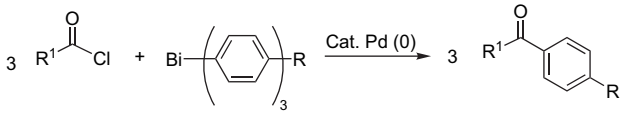
[a] Conditions for entries 3.1–3.9, equivalent ratios with respect to $BiAr_3$: ArCOCl (3.3 equiv), $BiAr_3$ (1 equiv), $Pd(PPh_3)_4$ (0.09 equiv), Et_3N (1 equiv), 1,4-dioxane (3 mL), 80 °C, 3 h.

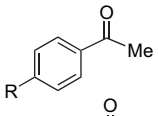
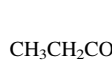
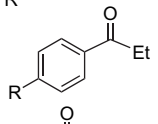
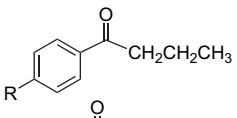
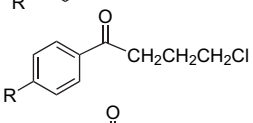
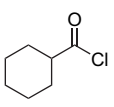
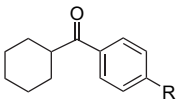
[b] Conditions for entries 3.10–3.16, equivalent ratios with respect to $BiAr_3$: ArCOCl (1 equiv), $BiAr_3$ (1 equiv), $Pd(PPh_3)_4$ (0.09 equiv), Et_3N (1 equiv), 1,4-dioxane (3 mL), 80 °C, 3 h.

[c] Isolated yields after column chromatography. All the products were characterized by ^1H, ^{13}C NMR, IR, and mass spectral analyses.

chlorides, 5 equiv of acyl chlorides were employed, out of which 3 equiv of acyl chlorides are necessary for atom-efficient coupling with three aryl groups from 1 equiv of triarylbismuth compound. Accordingly, several aliphatic acyl chlorides were studied for cross-coupling reaction with different triarylbismuths and the results are given in

Table 4. Cross-coupling reactivity of aliphatic acyl chlorides with triarylbismuths[a]



Entry	R^1COCl	Ketone	R	Yield[b] (%)
4.1			**4.1a**, R=–H	77
4.2	CH_3COCl		**4.2a**, R=–CH_3	81
4.3			**4.3a**, R=–OCH_3	78
4.4			**4.4a**, R=–H	80
4.5	CH_3CH_2COCl		**4.5a**, R=–CH_3	77
4.6			**4.6a**, R=–OCH_3	67
4.7			**4.7a**, R=–H	76
4.8	$CH_3CH_2CH_2COCl$		**4.8a**, R=–CH_3	73
4.9			**4.9a**, R=–OCH_3	65
4.10			**4.10a**, R=–H	89
4.11	$ClCH_2CH_2CH_2COCl$		**4.11a**, R=–CH_3	81
4.12			**4.12a**, R=–OCH_3	66
4.13			**4.13a**, R=–H	79
4.14			**4.14a**, R=–CH_3	89
4.15			**4.15a**, R=–OCH_3	80

[a] Conditions for R^1COCl, equivalent ratios with respect to $BiAr_3$: $BiAr_3$ (1 equiv), R^1COCl (5 equiv), $Pd(PPh_3)_4$ (0.15 equiv), Et_3N (5 equiv), 1,4-dioxane (3 mL), 80 °C, 3 h.

[b] Isolated yields after column chromatography. All the products were characterized by ^1H, ^{13}C NMR, IR, and mass spectral analyses.

Table 4. Triarylbismuths in combination with various aliphatic acyl chlorides reacted in a facile manner delivering good yields of the mixed alkyl aryl ketones. Thus, the acetyl chloride with various triarylbismuths furnished the cross-coupled functionalized acetophenones in good yields (entries 4.1–4.3). In addition, the reactivity of propionyl (entries 4.4–4.6), butyryl (entries 4.7–4.9), and 4-chlorobutyryl chlorides (entries 4.10–4.12) also produced efficient coupling with the different triarylbismuths and the corresponding mixed ketones were isolated in good yields. The reaction of cyclohexane carbonyl chloride with different triarylbismuths was also found to be efficient furnishing aryl cyclohexyl ketones in good to high yields (entries 4.13–4.15).

3. Conclusion

In this study, we have disclosed an atom-efficient cross-coupling reactions of triarylbismuths with a variety of aliphatic, aromatic, and hetero-aromatic acyl chlorides under palladium catalysis. The cross-coupling reactivity achieved using a variety of mono- and di-acyl chlorides with high functional group tolerance is another important aspect to note. This study also revealed the facile cross-coupling reactivity of electronically divergent acyl chlorides with equal ease with different triarylbismuths. In addition, the high atom-efficient reactivity and versatility associated with triarylbismuths under palladium catalyzed conditions for C–C bond formations were demonstrated. Further, this study opened up a new and atom-efficient route to access a variety of functionalized mixed ketones. It is expected that this study invariably strengthens the application of triarylbismuths as atom-efficient reagents for C–C bond formations, in addition to providing a new impetus for further elaboration of their use in organic synthesis.

4. Experimental section

4.1. General

All reactions were carried out in hot oven-dried Schlenk tubes under nitrogen atmosphere using anhydrous solvents. Anhydrous 1,4-dioxane was distilled over sodium prior to use and purged with nitrogen gas. All other solvents where ever necessary were purified by standard purification procedures. Analytical thin layer chromatography (TLC) was performed using silica gel 60 F_{254} pre-coated plates or using aluminum sheets pre-coated with silica gel (Merck). Visualization of TLC plate was accomplished with UV lamp or in an iodine chamber. The column chromatography was performed using silica gel 60–120 (Acme, Mumbai) mesh size with ethyl acetate/petroleum ether as an eluent system. Melting points were uncorrected and measured using JSGW melting point apparatus (Jain Scientific Glass Works, Ambala cantt, India). IR spectra were recorded on a Bruker vector 22 FT-IR spectrophotometer. ^{1}H and ^{13}C NMR spectra were recorded on a JEOL-Lambda (400 MHz) and Bruker (200 and 400 MHz) spectrometers using $CDCl_3$ as a solvent. Starting materials such as benzoyl chloride, 4-chlorobenzoyl chloride, acetyl chloride, propionyl chloride, butyryl chloride, 4-chlorobutyrylchloride, and 2-furoyl chloride were commercially available and other acid chlorides were prepared according to the standard procedure.[16] Triarylbismuths were prepared by standard protocols.[10a–c] GC analysis of the crude reaction mixtures and also of the pure isolated products was performed using Perkin Elmer (Clarus 500) system. FAB mass spectra were measured on JEOL SX 102/DA-6000 mass spectrometer. ESI-MS spectra measured on Waters HAB213 Q-Tof Premier Micromass spectrometer.

4.1.1. Representative procedure for aromatic acid chlorides. An oven-dried Schlenk tube was charged with PhCOCl (0.83 mmol, 0.116 g), $BiPh_3$ (0.25 mmol, 0.11 g), $Pd(PPh_3)_4$ (0.026 g, 0.0225 mmol), and Et_3N (0.25 mmol, 0.025 g) followed by anhydrous 1,4-dioxane (3 mL) under nitrogen atmosphere. The reaction mixture in Schlenk tube was stirred in an oil bath at 80 °C for 3 h. In the end, the reaction mixture was cooled to room temperature, quenched with water, and extracted with ethyl acetate (2×15 mL). The combined ethyl acetate extract was washed with dilute hydrochloric acid (5 mL), saturated sodium bicarbonate solution (5 mL), brine (2×5 mL), and dried over anhydrous magnesium sulfate. The solvent was removed and the crude product mixture was subjected to silica gel column chromatography to obtain the pure benzophenone (**2.1a**) in 91% isolated yield. All the products were characterized by ^{1}H, ^{13}C NMR, IR, and mass spectral analyses.

4.1.2. Reaction conditions for di-aroyl chlorides. The representative procedure given in Section 4.1.1 was followed with the following stoichiometric ratios of the reactants: diacid chloride (1 equiv), $BiAr_3$ (1 equiv), $Pd(PPh_3)_4$ (0.09 equiv), Et_3N (1 equiv), 1,4-dioxane (3 mL), 80 °C, 3 h. All the products were characterized by ^{1}H, ^{13}C NMR, IR, and mass spectra analyses.

4.1.3. Reaction conditions for aliphatic acid chlorides. The representative procedure given in Section 4.1.1 was followed with the following stoichiometric ratios of the reactants: RCOCl (1.25 mmol, 5 equiv), $BiPh_3$ (0.25 mmol, 1 equiv), Pd $(PPh_3)_4$ (0.15 equiv), Et_3N (1.25 mmol, 5 equiv), 1,4-dioxane (3 mL), 80 °C, 3 h. All products were characterized by ^{1}H, ^{13}C NMR, IR, and mass spectra analyses.

4.2. Spectral data

4.2.1. Diphenylmethanone[17a] (2.1a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.44–7.48 (t, 4H, J=7.8 Hz), 7.55–7.59 (t, 2H, J=7.3 Hz), 7.77–7.80 (d, 4H, J=8.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 128.2, 130.0, 132.3, 137.5, 196.7; IR (cm^{-1}): 1658; MS (FAB): 183 (M^++1).

4.2.2. (4-Methylphenyl)(phenyl)methanone[17a] (2.2a and 2.4a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.38 (s, 3H), 7.21–7.24 (d, 2H, J=8.3 Hz), 7.40–7.43 (t, 2H, J=7.7 Hz), 7.50–7.53 (t, 1H, J=7.1 Hz), 7.66–7.68 (d, 2H, J=8.3 Hz), 7.72–7.74 (d, 2H, J=7.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5, 128.1, 128.9, 129.8, 130.2, 132.1, 134.7, 137.8, 143.1, 196.4; IR (cm^{-1}): 1656; MS (FAB): 197 (M^++1).

4.2.3. (4-Methoxyphenyl)(phenyl)methanone[17a] (2.3a and 2.7a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.76 (s, 3H), 6.84–6.86 (d, 2H, J=8.8 Hz), 7.34–7.47 (m, 3H), 7.63–7.65 (d, 2H, J=8.3 Hz), 7.73–7.74 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 113.4, 128.1, 129.6,

130.0, 131.8, 132.4, 138.1, 163.1, 195.4; IR (cm^{-1}): 1650; MS (FAB): 213 (M^++1).

4.2.4. Di(4-methylphenyl)methanone[17b] (2.5a). Mp 89–92 °C (lit.[17c] 95–96 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 2.35 (s, 6H), 7.18–7.20 (d, 4H, J=7.8 Hz), 7.61–7.63 (d, 4H, J=8.0 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5, 128.8, 130.1, 135.1, 142.8, 196.2; IR (cm^{-1}): 1645; MS (FAB): 211 (M^++1).

4.2.5. (4-Methoxyphenyl)(4-methylphenyl)methanone[17d] (2.6a and 2.8a). Mp 85–87 °C (lit. 88–89 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 2.35 (s, 3H), 3.80 (s, 3H), 6.86–6.88 (d, 2H, J=6.8 Hz), 7.18–7.20 (d, 2H, J=7.8 Hz), 7.58–7.60 (d, 2H, J=8.3 Hz), 7.72–7.74 (d, 2H, J=6.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5, 55.3, 113.3, 128.8, 130.0, 132.4, 135.4, 142.5, 162.9, 195.3; IR (cm^{-1}): 1643; MS (FAB): 227 (M^++1).

4.2.6. Di(4-methoxyphenyl)methanone[17e] (2.9a). Mp 137–139 °C (lit.[6e] 138–139 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 3.85 (s, 6H), 6.92–6.94 (d, 4H, J=8.8 Hz), 7.74–7.77 (d, 4H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.3, 113.3, 130.6, 132.1, 162.7, 194.4; IR (cm^{-1}): 1635; MS (FAB): 243 (M^++1).

4.2.7. (4-Bromophenyl)(phenyl)methanone[17a] (2.10a). Mp 77–79 °C (lit.[17f] 75–76 °C); ^1H NMR (200 MHz, $CDCl_3$): δ 7.48–7.79 (m, 9H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 127.5, 128.4, 129.9, 131.5, 131.6, 132.6, 136.3, 137.1, 195.6; IR (cm^{-1}): 1650; MS (FAB): 261 (M^+) and 263 (M^++2).

4.2.8. (4-Bromophenyl)(4-methylphenyl)methanone[18a] (2.11a). Mp 123–125 °C; ^1H NMR (200 MHz, $CDCl_3$): δ 2.44 (s, 3H), 7.26–7.30 (d, 2H, J=8.8 Hz), 7.63–7.70 (m, 6H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 127.1, 129.1, 130.1, 131.4, 131.5, 134.4, 136.6, 143.5, 195.3; IR (cm^{-1}): 1644; MS (FAB): 275 (M^+) and 277 (M^++2).

4.2.9. (4-Bromophenyl)(4-methoxyphenyl)methanone[17d] (2.12a). Mp 147–149 °C (lit. 151–152 °C); ^1H NMR (200 MHz, $CDCl_3$): δ 3.89 (s, 3H), 6.94–6.98 (d, 2H, J=8.8 Hz), 7.62 (s, 4H), 7.77–7.81 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.5, 113.7, 126.8, 129.7, 131.2, 131.5, 132.4, 137.0, 163.4, 194.4; IR (cm^{-1}): 1639; MS (FAB): 290 (M^+) and 292 (M^++2).

4.2.10. (4-Chlorophenyl)(phenyl)methanone[18b] (2.13a). Mp 74–76 °C; ^1H NMR (400 MHz, $CDCl_3$): δ 7.36–7.42 (m, 4H), 7.50–7.53 (t, 1H, J=7.4 Hz), 7.65–7.70 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 128.3, 128.5, 129.8, 131.4, 132.6, 135.7, 137.1, 138.8, 195.4; IR (cm^{-1}): 1650; MS (FAB): 217 (M^++1).

4.2.11. (4-Chlorophenyl)(4-methylphenyl)methanone[18c] (2.14a). Mp 123–125 °C (lit. 128–129 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 2.36 (s, 3H), 7.20–7.22 (d, 2H, J=7.8 Hz), 7.36–7.39 (d, 2H, J=8.6 Hz), 7.60–7.67 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5, 128.4, 129.0, 130.1, 131.2, 134.4, 136.1, 138.5, 143.5, 195.2; IR (cm^{-1}): 1644; MS (FAB): 231 (M^++1).

4.2.12. (4-Chlorophenyl)(4-methoxyphenyl)methanone[6e] (2.15a). Mp 116–118 °C (lit. 119–120 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 3.86 (s, 3H), 6.93–6.95 (d, 2H, J=8.8 Hz), 7.41–7.43 (d, 2H, J=8.3 Hz), 7.67–7.69 (d, 2H, J=8.6 Hz), 7.76–7.78 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 113.6, 128.4, 129.7, 131.1, 132.4, 136.4, 138.2, 163.3, 194.2; IR (cm^{-1}): 1639; MS (FAB): 247 (M^++1).

4.2.13. (4-Fluorophenyl)(phenyl)methanone[17a] (2.16a). Mp 45–47 °C (lit.[18d] 49 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 7.05–7.10 (m, 2H), 7.39–7.53 (m, 3H), 7.67–7.78 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 115.3, 115.5, 128.3, 129.8, 130.0, 132.4, 132.5, 132.6, 133.7, 133.7, 137.4, 164.0, 166.6, 195.2; IR (cm^{-1}): 1647; MS (FAB): 201 (M^++1).

4.2.14. (4-Fluorophenyl)(4-methylphenyl)methanone[17d] (2.17a). Mp 92–94 °C (lit. 96–97 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 2.35 (s, 3H), 7.04–7.08 (t, 2H, J=8.3 Hz), 7.19–7.21 (d, 2H, J=7.6 Hz), 7.59–7.61 (d, 2H, J=7.8 Hz), 7.72–7.75 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5, 115.1, 115.4, 128.9, 130.0, 132.4, 134.0, 134.6, 143.2, 163.9, 166.4, 195.0; IR (cm^{-1}): 1648; MS (FAB): 215 (M^++1).

4.2.15. (4-Fluorophenyl)(4-methoxyphenyl)methanone[18e] (2.18a). Mp 89–91 °C (lit. 96 °C); ^1H NMR (200 MHz, $CDCl_3$): δ 3.89 (s, 3H), 6.94–6.99 (d, 2H, J=8.8 Hz), 7.15–7.26 (m, 2H), 7.77–7.81 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.5, 113.6, 115.2, 115.4, 130.0, 132.2, 132.3, 134.4, 163.2, 163.8, 166.3, 194.1; IR (cm^{-1}): 1640; MS (FAB): 231 (M^++1).

4.2.16. (3-Methylphenyl)(phenyl)methanone[9d] (2.19a). ^1H NMR (400 MHz, $CDCl_3$): δ 2.40 (s, 3H), 7.32–7.39 (m, 2H), 7.44–7.47 (t, 2H, J=7.6 Hz), 7.54–7.61 (m, 3H), 7.76–7.79 (d, 2H, J=7.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.3, 127.3, 128.0, 128.1, 129.9, 130.4, 132.3, 133.1, 137.5, 137.6, 138.0, 196.9; IR (cm^{-1}): 1658; MS (FAB): 197 (M^++1).

4.2.17. (3-Methylphenyl)(4-methylphenyl)methanone[17b] (2.20a). Mp lit. 82 °C; ^1H NMR (400 MHz, $CDCl_3$): δ 2.36 (s, 3H), 2.39 (s, 3H), 7.21–7.24 (d, 2H, J=8.3 Hz), 7.28–7.32 (m, 2H), 7.49–7.51 (d, 1H, J=6.8 Hz), 7.55 (s, 1H), 7.66–7.68 (d, 2H, J=8.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.3, 21.5, 127.1, 127.9, 128.8, 130.2, 130.2, 132.8, 134.9, 137.8, 137.9, 143.0, 196.7; IR (cm^{-1}): 1656; MS (FAB): 211 (M^++1).

4.2.18. (3-Methylphenyl)(4-methoxyphenyl)methanone (2.21a). ^1H NMR (400 MHz, $CDCl_3$): δ 2.39 (s, 3H), 3.86 (s, 3H), 6.92–6.95 (d, 2H, J=9.0 Hz), 7.30–7.36 (m, 2H), 7.49–7.51 (d, 1H, J=7.1 Hz), 7.55 (s, 1H), 7.78–7.82 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.3, 55.4, 113.3, 126.8, 127.8, 130.0, 130.1, 132.4, 132.5, 137.9, 138.2, 163.0, 195.7; IR (cm^{-1}): 1650; MS (FAB): 227 (M^++1).

4.2.19. (3-Methoxyphenyl)(phenyl)methanone[9d] (2.22a). ^1H NMR (400 MHz, $CDCl_3$): δ 3.84 (s, 3H), 7.10–7.12 (m, 1H), 7.30–7.38 (m, 3H), 7.44–7.47 (t, 2H, J=7.8 Hz), 7.54–7.58 (m, 1H), 7.77–7.80 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 114.2, 118.8, 122.8, 128.2, 129.1, 130.0, 132.4, 137.5, 138.8, 159.4, 196.5; IR (cm^{-1}): 1659; MS (FAB): 213 (M^++1).

4.2.20. (3-Methoxyphenyl)(4-methylphenyl)methanone (2.23a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.39 (s, 3H), 3.80 (s, 3H), 7.05–7.08 (m, 1H), 7.21–7.34 (m, 5H), 7.67–7.69 (d, 2H, J=7.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 55.3, 114.1, 118.5, 122.6, 128.9, 129.0, 130.2, 134.7, 139.1, 143.2, 159.4, 196.2; IR (cm^{-1}): 1656; MS (FAB): 227 (M^++1).

4.2.21. (3-Methoxyphenyl)(4-methoxyphenyl)methanone (2.24a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.77 (s, 3H), 3.80 (s, 3H), 6.86–6.89 (d, 2H, J=8.8 Hz), 7.01–7.04 (m, 1H), 7.18–7.30 (m, 3H), 7.74–7.76 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 55.4, 113.4, 114.1, 118.2, 122.3, 129.0, 130.0, 132.5, 139.5, 159.4, 163.2, 195.3; IR (cm^{-1}): 1651; MS (FAB): 243 (M^++1).

4.2.22. 2-Naphthyl(phenyl)methanone[9d] (2.25a). Mp 72–75 °C (lit.[19a] 80–81 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.46–7.65 (m, 5H), 7.84–7.93 (m, 6H), 8.25 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 125.7, 126.7, 127.7, 128.2, 128.2, 129.3, 130.0, 131.8, 132.1, 132.3, 134.7, 135.2, 137.8, 196.7; IR (cm^{-1}): 1655; MS (FAB): 233 (M^++1).

4.2.23. (4-Methylphenyl)(2-naphthyl)methanone[18a] (2.26a). Mp 83–85 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.38 (s, 3H), 7.22–7.24 (d, 2H, J=8.0 Hz), 7.44–7.54 (m, 2H), 7.68–7.70 (d, 2H, J=8.0 Hz), 7.81–7.87 (m, 4H), 8.16 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 125.7, 126.6, 127.7, 128.1, 128.9, 129.2, 130.2, 131.5, 132.1, 135.0, 143.1, 196.4; IR (cm^{-1}): 1652; MS (FAB): 247 (M^++1).

4.2.24. (4-Methoxyphenyl)(2-naphthyl)methanone (2.27a). Mp 87–89 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.82 (s, 3H), 6.90–6.92 (d, 2H, J=8.5 Hz), 7.44–7.53 (m, 2H), 7.79–7.86 (m, 6H), 8.14 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 113.5, 125.8, 126.6, 127.7, 127.9, 128.1, 129.2, 130.3, 131.0, 132.1, 132.5, 134.9, 135.4, 163.1, 195.5; IR (cm^{-1}): 1648; MS (FAB): 263 (M^++1).

4.2.25. 1-Naphthyl(phenyl)methanone[17a] (2.28a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.42–7.60 (m, 7H), 7.86–7.92 (m, 3H), 7.98–8.00 (d, 1H, J=8.1 Hz), 8.10–8.12 (d, 1H, J=7.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 124.2, 125.6, 126.4, 127.2, 127.7, 128.3, 128.3, 130.3, 130.8, 131.2, 133.1, 133.6, 136.2, 138.2, 197.9; IR (cm^{-1}): 1658; MS (FAB): 233 (M^++1).

4.2.26. (4-Methylphenyl)(1-naphthyl)methanone[19b] (2.29a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.27 (s, 3H), 7.08–7.11 (d, 2H, J=8.5 Hz), 7.31–7.44 (m, 4H), 7.63–7.65 (d, 2H, J=8.3 Hz), 7.75–7.77 (d, 1H, J=7.1 Hz), 7.82–7.84 (d, 1H, J=8.0 Hz), 7.92–7.94 (d, 1H, J=8.3 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.7, 124.3, 125.6, 126.3, 127.0, 127.2, 128.3, 129.1, 130.5, 130.9, 133.6, 135.6, 136.6, 144.1, 197.7; IR (cm^{-1}): 1655; MS (FAB): 247 (M^++1).

4.2.27. (4-Methoxyphenyl)(1-naphthyl)methanone[19c] (2.30a). ^{1}H NMR (200 MHz, $CDCl_3$): δ 3.87 (s, 3H), 6.90–6.94 (d, 2H, J=8.8 Hz), 7.47–7.54 (m, 4H), 7.83–8.00 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.5, 113.7, 124.4, 125.7, 126.3, 126.8, 127.0, 128.3, 130.6, 130.9, 131.1, 132.8, 133.6, 137.0, 163.8, 196.7; IR (cm^{-1}): 1652; MS (FAB): 263 (M^++1).

4.2.28. (2-Methylphenyl)(phenyl)methanone[17a] (2.31a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.25 (s, 3H), 7.14–7.51 (m, 7H), 7.71–7.73 (d, 2H, J=7.3 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 19.9, 125.2, 128.4, 128.5, 130.1, 130.2, 131.0, 133.1, 136.7, 137.7, 138.6, 198.6; IR (cm^{-1}): 1663; MS (FAB): 197 (M^++1).

4.2.29. (2-Methylphenyl)(4-methylphenyl)methanone[17d] (2.32a). ^{1}H NMR (200 MHz, $CDCl_3$): δ 2.31 (s, 3H), 2.42 (s, 3H), 7.23–7.28 (m, 6H), 7.68–7.72 (d, 2H, J=7.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 19.8, 21.7, 125.1, 128.2, 129.1, 130.0, 130.8, 135.1, 136.4, 139.0, 144.0, 198.3; IR (cm^{-1}): 1660; MS (FAB): 211 (M^++1).

4.2.30. 2-Furanyl(phenyl)methanone[9d] (3.1a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 6.59–6.61 (m, 1H), 7.24–7.52 (m, 4H), 7.58–7.72 (m, 1H), 7.96–7.98 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 112.1, 120.6, 128.3, 129.2, 132.5, 137.1, 147.1, 152.1, 182.5; IR (cm^{-1}): 1647; MS (FAB): 173 (M^++1).

4.2.31. 2-Furanyl(4-methylphenyl)methanone[19b] (3.2a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.37 (s, 3H), 6.51–6.52 (m, 1H), 7.15–7.24 (m, 3H), 7.62–7.63 (m, 1H), 7.81–7.83 (d, 2H, J=8.3 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 112.1, 120.2, 129.1, 129.4, 134.5, 143.4, 146.9, 152.4, 182.3; IR (cm^{-1}): 1643; MS (FAB): 187 (M^++1).

4.2.32. 2-Furanyl(4-methoxyphenyl)methanone[6g] (3.3a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.89 (s, 3H), 6.58–6.59 (m, 1H), 6.97–7.00 (d, 2H, J=8.8 Hz), 7.23–7.24 (d, 1H, J=3.4 Hz), 7.69 (s, 1H), 8.02–8.04 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 112.0, 113.6, 119.6, 129.7, 131.6, 146.5, 152.4, 163.2, 181.1; IR (cm^{-1}): 1638; MS (FAB): 203 (M^++1).

4.2.33. 2-Furanyl(4-fluorophenyl)methanone (3.4a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 6.53–6.55 (m, 1H), 7.08–7.19 (m, 3H), 7.64 (s, 1H), 7.95–7.99 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 112.3, 115.5, 115.7, 120.4, 131.9, 132.0, 147.1, 152.2, 164.2, 166.7, 180.9; IR (cm^{-1}): 1645; MS (FAB): 191 (M^++1).

4.2.34. Phenyl(2-thienyl)methanone[17a,19d] (3.5a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.04–7.09 (m, 1H), 7.37–7.54 (m, 4H), 7.58–7.62 (m, 1H), 7.71–7.80 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 127.8, 128.3, 129.0, 132.1, 134.1, 134.7, 137.9, 143.4, 188.1; IR (cm^{-1}): 1627; MS (FAB): 189 (M^++1).

4.2.35. (4-Methylphenyl)(2-thienyl)methanone[19d] (3.6a). Mp 65–67 °C (lit.[19e] 70–71 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.33 (s, 3H), 7.03–7.05 (m, 1H), 7.17–7.19 (d, 2H, J=7.8 Hz), 7.52–7.59 (m, 2H), 7.67–7.69 (d, 2H, J=8.3 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5, 127.7, 128.9, 129.2, 133.7, 134.4, 135.2, 142.9, 143.6, 187.8; IR (cm^{-1}): 1632; MS (FAB): 203 (M^++1).

4.2.36. (4-Methoxyphenyl)(2-thienyl)methanone[19d] (3.7a). Mp 68–70 °C (lit.[19e] 72–74 °C); ^{1}H NMR

(400 MHz, $CDCl_3$): δ 3.78 (s, 3H), 6.87–6.89 (d, 2H, J=8.8 Hz), 7.03–7.06 (m, 1H), 7.53–7.59 (m, 2H), 7.79–7.81 (d, 2H, J=6.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.3, 113.5, 127.7, 130.4, 131.4, 133.3, 133.9, 143.6, 162.9, 186.7; IR (cm^{-1}): 1628; MS (FAB): 219 (M^++1).

4.2.37. (4-Fluorophenyl)(2-thienyl)methanone[19d] (3.8a). Mp 93–94 °C (lit.[19e] 95–96 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.07–7.12 (m, 3H), 7.54–7.66 (m, 2H), 7.81–7.84 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 115.4, 115.6, 127.9, 131.6, 131.7, 134.2, 134.6, 143.2, 163.9, 166.4, 186.6; IR (cm^{-1}): 1628; MS (FAB): 207 (M^++1).

4.2.38. (4-Chlorophenyl)(2-thienyl)methanone[6g] (3.9a). Mp 93–95 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.09–7.11 (t, 1H, J=4.9 Hz), 7.39–7.41 (d, 2H, J=8.6 Hz), 7.54–7.56 (d, 1H, J=3.7 Hz), 7.66–7.67 (d, 1H, J=4.9 Hz), 7.73–7.75 (d, 2H, J=8.3 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 128.0, 128.7, 130.5, 134.5, 134.7, 136.3, 138.6, 143.1, 186.9; IR (cm^{-1}): 1628; MS (FAB): 223 (M^++1).

4.2.39. 1,3-Phenylenebis(phenyl)methanone[15b] (3.10a). Mp 96–98 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.39–7.43 (t, 4H, J=7.1 Hz), 7.50–7.56 (m, 3H), 7.73–7.75 (d, 4H, J=8.3 Hz), 7.93–7.95 (d, 2H, J=7.6 Hz), 8.11 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 128.4, 128.4, 130.0, 131.1, 132.8, 133.4, 136.8, 137.7, 195.8; IR (cm^{-1}): 1655; MS (FAB): 287 (M^++1).

4.2.40. 1,3-Phenylenebis(4-methylphenyl)methanone (3.11a). Mp 118–120 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.37 (s, 6H), 7.21–7.24 (d, 4H, J=8.0 Hz), 7.53–7.56 (t, 1H, J=7.8 Hz), 7.66–7.68 (d, 4H, J=8.3 Hz), 7.91–7.94 (m, 2H), 8.07–8.08 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 128.3, 129.1, 130.2, 130.9, 133.1, 134.2, 138.0, 143.7, 195.6; IR (cm^{-1}): 1656; MS (FAB): 315 (M^++1).

4.2.41. 1,3-Phenylenebis(4-methoxyphenyl)methanone (3.12a). Mp 136–138 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 3.85 (s, 6H), 6.93–6.95 (d, 4H, J=7.1 Hz), 7.56–7.59 (t, 1H, J=7.8 Hz), 7.80–7.82 (d, 4H, J=7.3 Hz), 7.92–7.93 (d, 2H, J=7.6 Hz), 8.05 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 55.4, 113.6, 128.3, 129.5, 130.5, 132.5, 132.6, 138.3, 163.4, 194.6; IR (cm^{-1}): 1651. HRMS (ES^+) for (M+H) $C_{22}H_{19}O_4$, calcd: 347.1283; found: 347.1284.

4.2.42. 1,3-Phenylenebis(4-fluorophenyl)methanone[15b] (3.13a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.08–7.30 (m, 4H), 7.55–7.59 (t, 1H, J=7.6 Hz), 7.77–7.80 (m, 4H), 7.91–7.93 (d, 2H, J=7.6 Hz), 8.05 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 115.5, 115.8, 128.6, 130.7, 132.6, 132.7, 133.3, 137.7, 164.3, 166.8, 194.2; IR (cm^{-1}): 1655; MS (FAB): 323 (M^++1).

4.2.43. 1,4-Phenylenebis(phenyl)methanone[15b] (3.14a). Mp 158–160 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.47–7.51 (t, 4H, J=7.8 Hz), 7.58–7.62 (t, 2H, J=7.3 Hz), 7.80–7.82 (d, 4H, J=8.5 Hz), 7.86 (s, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 128.4, 129.7, 130.0, 132.9, 136.8, 140.5, 196.0; IR (cm^{-1}): 1653; MS (FAB): 287 (M^++1).

4.2.44. 1,4-Phenylenebis(4-methylphenyl)methanone (3.15a). Mp 173–176 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.38 (s, 6H), 7.22–7.24 (d, 4H, J=8.0 Hz), 7.67–7.69 (d, 4H, J=8.0 Hz), 7.78 (s, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 129.1, 129.5, 130.3, 134.2, 140.7, 143.8, 195.8; IR (cm^{-1}): 1646; MS (FAB): 315 (M^++1).

4.2.45. 1,4-Phenylenebis(4-fluorophenyl)methanone[15b] (3.16a). Mp 208–210 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.10–7.26 (m, 4H), 7.72–7.86 (m, 8H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 115.6, 115.8, 129.6, 132.7, 132.8, 140.5, 164.4, 166.9, 194.4; IR (cm^{-1}): 1647; MS (FAB): 323 (M^++1).

4.2.46. 1-Phenylethanone[17a] (4.1a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.58 (s, 3H), 7.41–7.56 (m, 3H), 7.92–7.94 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 26.6, 128.3, 128.5, 133.1, 137.1, 198.1; IR (cm^{-1}): 1684; MS (FAB): 121 (M^++1).

4.2.47. 1-(4-Methylphenyl)ethanone[20a] (4.2a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.38 (s, 3H), 2.55 (s, 3H), 7.22–7.24 (d, 2H, J=7.8 Hz), 7.82–7.84 (d, 2H, J=8.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 26.5, 128.4, 129.2, 134.7, 143.8, 197.8; IR (cm^{-1}): 1680; MS (FAB): 135 (M^++1).

4.2.48. 1-(4-Methoxyphenyl)ethanone[20b] (4.3a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.52 (s, 3H), 3.83 (s, 3H), 6.88–6.91 (d, 2H, J=6.8 Hz), 7.88–7.91 (d, 2H, J=6.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 26.3, 55.4, 113.6, 130.3, 130.5, 163.5, 196.7; IR (cm^{-1}): 1675; MS (FAB): 151 (M^++1).

4.2.49. 1-Phenyl-1-propanone[9d] (4.4a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 1.20 (t, 3H, J=7.3 Hz), 2.98 (q, 2H, J=7.1 Hz), 7.41–7.45 (t, 2H, 8.0 Hz), 7.50–7.54 (t, 1H, J=7.3 Hz), 7.93–7.95 (d, 2H, J=7.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 8.1, 31.7, 127.9, 128.5, 132.8, 136.8, 200.8; IR (cm^{-1}): 1687; MS (FAB): 135 (M^++1).

4.2.50. 1-(4-Methylphenyl)-1-propanone[19b] (4.5a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 1.19 (t, 3H, J=7.1 Hz), 2.38 (s, 3H), 2.98 (q, 2H, J=7.3 Hz), 7.21–7.24 (d, 2H, J=8.3 Hz), 7.83–7.85 (d, 2H, J=8.0 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 8.2, 21.5, 31.6, 128.0, 129.1, 134.3, 143.5, 200.5; IR (cm^{-1}): 1685; MS (FAB): 149 (M^++1).

4.2.51. 1-(4-Methoxyphenyl)-1-propanone[20c] (4.6a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 1.18 (t, 3H, J=7.3 Hz), 2.92 (q, 2H, J=7.3 Hz), 3.83 (s, 3H), 6.88–6.90 (d, 2H, J=8.8 Hz), 7.90–7.92 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 8.4, 31.4, 55.4, 113.6, 130.0, 130.2, 163.3, 199.4; IR (cm^{-1}): 1679; MS (FAB): 165 (M^++1).

4.2.52. 1-Phenyl-1-butanone[17f] (4.7a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 0.98 (t, 3H, J=7.6 Hz), 1.75 (q, 2H, J=7.3 Hz), 2.92 (t, 2H, J=7.4 Hz), 7.41–7.45 (t, 2H, J=7.6 Hz), 7.50–7.55 (t, 1H, J=7.3 Hz), 7.92–7.95 (d, 2H, J=7.6 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 13.8, 17.7, 40.4, 127.9, 128.4, 132.8, 137.0, 200.4; IR (cm^{-1}): 1685; MS (FAB): 149 (M^++1).

4.2.53. 1-(4-Methylphenyl)-1-butanone[20d] (4.8a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 0.92 (t, 3H, J=7.6 Hz), 1.65–1.71 (m, 2H), 2.33 (s, 3H), 2.84 (t, 2H, J=7.1 Hz), 7.16–7.18 (d, 2H, J=8.3 Hz), 7.77–7.79 (d, 2H, J=8.3 Hz);

^{13}C NMR (100 MHz, $CDCl_3$): δ 13.8, 17.8, 21.6, 40.3, 128.1, 129.1, 134.5, 143.5, 200.1; IR (cm^{-1}): 1681; MS (FAB): 163 (M^++1).

4.2.54. 1-(4-Methoxyphenyl)-1-butanone[20e] (4.9a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 0.92 (t, 3H, J=7.3 Hz), 1.16–1.71 (m, 2H), 2.82 (t, 2H, J=7.3 Hz), 3.80 (s, 3H), 6.85–6.88 (d, 2H, J=7.1 Hz), 7.86–7.89 (d, 2H, J=7.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 13.9, 17.9, 40.1, 55.4, 113.6, 130.1, 130.2, 163.2, 199.1; IR (cm^{-1}): 1675; MS (FAB): 179 (M^++1).

4.2.55. 4-Chloro-1-phenyl-1-butanone[17f] (4.10a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.17–2.24 (m, 2H), 3.16 (t, 2H, J=7.1 Hz), 3.66 (t, 2H, J=6.1 Hz), 7.43–7.47(t, 2H, J=7.8 Hz), 7.53–7.57 (t, 1H, J=7.1 Hz), 7.95–7.97 (d, 2H, J= 8.0 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 26.6, 35.2, 44.6, 127.9, 128.6, 133.1, 136.6, 198.9; IR (cm^{-1}): 1683; MS (FAB): 183 (M^++1).

4.2.56. 4-Chloro-1-(4-methylphenyl)-1-butanone (4.11a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.17–2.22 (m, 2H), 2.37 (s, 3H), 3.12 (t, 2H, J=6.8 Hz), 3.64 (t, 2H, J=6.3 Hz), 7.22–7.24 (d, 2H, J=7.8 Hz), 7.83–7.85 (d, 2H, J=8.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 26.7, 35.1, 44.7, 128.0, 129.2, 134.2, 143.9, 198.5; IR (cm^{-1}): 1681; MS (FAB): 197 (M^++1).

4.2.57. 4-Chloro-1-(4-methoxyphenyl)-1-butanone (4.12a). ^{1}H NMR (400 MHz, $CDCl_3$): δ 2.17–2.21 (m, 2H), 3.10 (t, 2H, J=7.1 Hz), 3.65 (t, 2H, J=6.1 Hz), 3.85 (s, 3H), 6.90–6.93 (d, 2H, J=6.8 Hz), 7.92–7.95 (d, 2H, J= 7.1 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 26.9, 34.9, 44.8, 55.4, 113.7, 129.9, 130.2, 163.5, 197.5; IR (cm^{-1}): 1675; MS (FAB): 213 (M^++1).

4.2.58. Cyclohexylphenylmethanone[20e] (4.13a). ^{1}H NMR (200 MHz, $CDCl_3$): δ 1.42–1.86 (m, 10H), 3.25–3.26 (m, 1H), 7.45–7.49 (m, 4H), 7.92–7.95 (d, 1H, J=6.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 25.8, 25.9, 29.4, 45.6, 128.2, 128.5, 132.7, 136.3, 203.9; IR (cm^{-1}): 1680; MS (FAB): 189 (M^++1).

4.2.59. Cyclohexyl(4-methylphenyl)methanone[17d] (4.14a). Mp 61–63 °C (lit. 61–63 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ 1.17–1.81 (m, 10H), 2.32 (s, 3H), 3.12–3.19 (m, 1H), 7.16–7.18 (d, 2H, J=8.6 Hz), 7.76–7.78 (d, 2H, J=8.0 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6, 25.9, 25.9, 29.4, 45.5, 128.3, 129.2, 133.8, 143.4, 203.5; IR (cm^{-1}): 1676; MS (FAB): 203 (M^++1).

4.2.60. Cyclohexyl(4-methoxyphenyl)methanone[20e] (4.15a). Mp 61–63 °C; ^{1}H NMR (200 MHz, $CDCl_3$): δ 1.25–1.83 (m, 10H), 3.16–3.26 (m, 1H), 3.86 (s, 3H), 6.91–6.95 (d, 2H, J=8.8 Hz), 7.91–7.95 (d, 2H, J=8.8 Hz); ^{13}C NMR (100 MHz, $CDCl_3$): δ 25.9, 25.9, 29.5, 45.3, 55.4, 113.7, 129.2, 130.5, 163.2, 202.4; IR (cm^{-1}): 1670; MS (FAB): 219 (M^++1).

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