**Palladium Catalyzed Acylations: One-pot Synthesis of Indenones**

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ABSTRACT:

An efficient one-pot synthesis of substituted indenones was accomplished starting from simple *ortho*-iodoketones and aldehydes. [Pd]-catalyzed direct acylation of *ortho*-iodoketones with aldehydes was employed as the key step. Subsequent intramolecular aldol condensation afforded the indenones. Notably, a variety of indenones were achieved. Significantly, the natural product neo-lignan was accomplished in one-pot.



Introduction:

Indenones are ubiquitous structural units, which exhibit broad biological spectrum and also served as useful intermediates for the synthesis of natural products (Figure 1).1 Till now, various notable approaches have been reported on their synthesis.2-8 For example, transition-metal catalyzed annulations of alkynes was accomplished using carbon monoxide (CO) as carbonylating agent.3 While, annulations of internal alkynes with 2-halobenzaldehydes or 2-(methoxycarbonyl)phenylboronic acids or 2-bromophenylboronic acids or 2-iodobenzonitriles have also been established using transition-metal catalysis (Scheme 1).4-6 On the other hand, radical mediated cyclizations were also reported (Scheme 1).7-8



**Figure 1**: Compounds containing indenone core.

Nevertheless, there are certain drawbacks with most of these methods. For example, regioselectivity is a common problem in transition-metal mediated annulations, particularly, when unsymmetrical alkynes were used.9 Therefore, the development of new synthetic methods with high regioselectivity is beneficial. In continuation of our ongoing research interest on transition-metal catalysis,10 very recently, we have disclosed an environmentally benign acylations,10h for the synthesis of various ketones. In this transformation, an iodoarene was coupled with an aldehyde in the presence of [Pd]-catalyst and oxidant. Significantly, unlike earlier reports, the reaction was feasible without activating the aldehyde functionality.9f On the other hand, we have developed acid promoted domino one-pot synthesis of indanones and indenones from simple cinnamic acid esters.11 Herein, we describe an efficient one-pot domino process, for the synthesis of a wide variety of indenones starting from readily available *ortho*-iodoketones and aldehydes. Significantly, this strategy was applied to the one-pot synthesis of naturally occurring neo-lignan **1**.1e



Inspired by our recent results on [Pd]-catalyzed direct acylations,10h we envisioned that the indenones **8** could be obtained from 1,2-diketobenzenes **7** by employing intramolecular aldol condensation. The required 1,2-diketobenzenes **7** would be achieved from *ortho*-iodoketones **6** using [Pd]-catalyzed acylations (retrosynthetic analysis A; Scheme 2). Alternatively, the indenones **9** could also be accomplished from 1,2-diketones **10**. These 1,2-diketones **7**, which in turn would be synthesized from *ortho*-iodoketones **11** (retrosynthetic analysis B; Scheme 2).



**Scheme 2**: Retrosynthetic analysis.

Results and Discussion:

Thus, we initially examined the reaction with *ortho*-iodopropiophenone **6a** and benzaldehyde **5a**. Particularly, the acylation reaction was carried out under established conditions (i.e. reported conditions with iodo-arenes and benzaldehydes). We were delighted to find that the reaction was amenable to this ketone **6a** as well. Then, the isolated 1,2-diketobenzene **7aa** was subjected to an acid mediated intramolecular aldol condensation. As expected, furnished the desired indenone **8aa** (Scheme 3). With these encouraging results, next, we performed acylation and subsequent intramolecular aldol condensation12 in one-pot manner. To our delight, as anticipated, gave the indenone **8aa**, in 63% overall yield (Scheme 3).



**Scheme 3**: Step-wise and one-pot synthesis of indenone **8aa**.

With these conditions, next, we investigated the scope of this protocol by exploring the reaction between various *ortho*-iodopropiophenones **6a-d** and benzaldehydes **5a-m**. Gratifyingly, the reaction was found amenable and furnished the products **8aa-df**,in fair to good yields (Table 1). Delightfully, the reaction proceeded smoothly with electron deactivating (F, Cl & Br) as well as electron donating (alkyl, OMe etc.) substituents on the aromatic ring of benzaldehydes **5e-5p**. Significantly, the reaction was also successful with 2-furaldehyde **5m** (Table 1, **8am**). The reaction was also compatible with the protecting group free *para*-hydroxybenzaldehyde **5j** (Table 1, **8aj**). It is worth mentioning that in case of electron rich aromatic systems (iodoarenes or benzaldehydes), the direct formation of indenones was observed, albeit in minor amounts, before the addition of H2SO4 (Table 1, **8aj-df**). Presumably the aldol condensation would be induced either by the silver ions (Ag+) from Ag2O or the carboxylic acid that might be generated from the corresponding benzaldehyde. As in our previous report, the acylation reaction was unsuccessful with benzaldehydes bearing strong electron withdrawing nitro group and heteroaromatic aldehydes. These observations appear to be common under such radical mediated conditions. May be due to destalizing nature of electron withdrawing groups.13

**Table 1**: Scope and generality of formation of indenones **8aa-df**.

Since, it was noticed that in case of the electron rich aromatic ring (benzaldehydes or iodoarenes), minor amounts of indenones **8** was formed before the addition of H2SO4. We supposed that the reaction could be driven to the target products; if both or at-least one of the aromatic rings is/are sufficiently electron rich. In such situations, even a mild acid(s) such as Ag+ of Ag2O or in-situ formed carboxylic acids could be able to promote the subsequent intramolecular aldol condensation. Therefore, the reaction was performed with the iodoarenes **6a-d** and benzaldehydes **5k-p**, under standard conditions, without the subsequent addition of H2SO4 (i.e. ***Conditions B***). To our delight, the reaction proceeded smoothly and furnished the indenones **8an-dk** (Table 2).

**Table 2**: Scope and generality of formation of indenones **8an-dk**.



Furthermore, the structure of **8ap** was also confirmed by the single crystal x-ray diffraction analysis (Figure 2).



**Figure 2**: X-ray crystal structure of indenone **8ap**. Thermal ellipsoids is drawn at 50% probability level.

Furthermore, to check the scope and applicability of the present protocol, the reaction was also performed with *ortho*-iodobenzophenones **11a-d** (Table 3). In contrast to the above, in this case, aliphatic aldehydes **5q-s** were employed for the [Pd]-catalyzed acylations. Gratifyingly, the protocol was also quite successful and gave the indenones **9ar-dr** (Table 3; ***Conditions A*** *or* ***Conditions B***). Notably, the reaction was compatible with heteroaromatic *ortho*-iodobenzophenone **11c** (Table 3).

**Table 3**: Scope and generality of formation of indenones **9aa-dr**.



To further demonstrate the applicability of the method, we investigated the coupling between the ketone **11e** and aliphatic aldehydes **5q** and **5r**. Notably, in case of diketone intermediates derived from the *ortho*-iodoacetophenone **11e**, as anticipated, proceeded through the formation of relatively more stable enolate under acidic conditions and gave the indenones **9eq** and **9er** (Scheme 4; ***Conditions A***).



**Scheme 4**: Synthesis of indenones **9eq** and **9er**.

On the other hand, the reaction between the ketone **6a** and cyclohexanaldehyde **5s**, furnished as the inseparable mixture of indenone **8as** and spirocyclic ketone **9as** (Scheme 5). This may be due to the competitive enolization of both ketones and an independent subsequent intramolecular aldol condensation. When the reaction was conducted separately on the isolated mixture of **8as** and **9as** with H2SO4, no change was noticed in the ratio of **8as** and **9as** (i.e. ***Conditions A***). Thus reveals that the reaction might proceeds through the irreversible independent path without equilibration for aldol condensation reaction.



**Scheme 5**: Formation of indenones mixture **8as** and **9as**.

In 1984, Otto R. Gottlieb et al. isolated neo-lignan from the fruits of virola sebifera.1f Later, in 1998 the research group of Davi C. Harrowven revised its structure as neo-lignan **1** with the help of their synthetic, physical and spectroscopic studies, (see; Figure 1). To further demonstrate the synthetic utility of the present strategy, it was investigated for one-pot synthesis of neo-lignan **1**. Thus, the *ortho*-iodoketone **6c** was subjected to the reaction with piperonaldehyde **5n**, under standard conditions without H2SO4 (i.e. ***Conditions B***). Significantly, the natural product neo-lignan **1** was obtained in one-pot (Scheme 6).



**Scheme 6**: One-pot synthesis of neo-lignan **1**.

In summary, we have developed an efficient one-pot protocol for synthesis of substituted indenones starting from simple *ortho*-iodoketones and aldehydes. The [Pd]-catalyzed direct acylation was employed as the key step. Subsequent intramolecular aldol condensation afforded the indenones in one-pot. Significantly, a variety of indenones were achieved. Delightfully, the strategy was applied to the one-pot synthesis of naturally occurring neo-lignan.

**Experimental:**

IR spectra were recorded on a FTIR spectrophotometer. 1H NMR spectra were recorded on 400 MHz spectrometer at 295 K in CDCl3; chemical shifts (δ ppm) and coupling constants (Hz) are reported in standard fashion with reference to either internal standard tetramethylsilane (TMS) (δH =0.00 ppm) or CHCl3 (δH = 7.25 ppm). 13C NMR spectra were recorded on 100 MHz spectrometer at RT in CDCl3; chemical shifts (δ ppm) are reported relative to CHCl3 [δC = 77.00 ppm (central line of triplet)]. In the 13C NMR, the nature of carbons (C, CH, CH2 and CH3) was determined by recording the DEPT-135 spectra. In the 1H-NMR, the following abbreviations were used throughout: s = singlet, d = doublet, t = triplet, q = quartet, qui =quintet, sept = septet, dd = doublet of doublet, m = multiplet and br. s = broad singlet. High-resolution mass spectra (HR-MS) were recorded on Q-TOF electron spray ionization (ESI) mode and atmospheric pressure chemical ionization (APCI) modes. Melting points were determined on an electrothermal melting point apparatus and are uncorrected.

 All small scale reactions were carried out using Schlenk tube. Reactions were monitored by TLC on silica gel using a combination of hexane and ethyl acetate as eluents. Reactions were generally run under argon or a nitrogen atmosphere. Solvents were distilled prior to use; petroleum ether with a boiling range of 60 to 80 °C was used. Acme’s silica-gel (60–120 mesh) was used for column chromatography (approximately 20 g per one gram of crude material).

The aldehydes **5a-s** which have been used are commercially available.

The *ortho*-iodo ketones **6a-d**, **11a-e** were synthesized from the corresponding *ortho*-iodo aldehydes by Grignard reaction followed by PCC Oxidation which is reported in literature.10a, 14

**GP-1 [General procedure for preparation of 8**, **9 (conditions A)]:**

GP-1 was carried out with *ortho*-iodoketone **6a-d**, **11a-e** (104-152 mg, 0.40 mmol**)** andaldehyde **5a-s (**166.4-366.5 mg, 1.6 mmol) in the presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12-20 h. Progress of the reaction was monitored by TLC till the *ortho*-iodoketone **6a-d**, **11a-e** was completed. Then the reaction mixture was removed from oil bath and allow to reach room temperature and then added conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature. Progress of the products **8aa-df**, **9ar-er** formation was monitored by TLC till the reaction was completed. The reaction mixture was quenched by the addition of aqueous NaHCO3 solution and then extracted with ethyl acetate (3 × 15 mL). The organic layers were washed with saturated NaCl solution, dried (Na2SO4) and filtered. Evaporation of the solvent under reduced pressure and purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate) furnished the products **8**, **9** (34.0-112.0 mg, 46-79 %) as viscous liquid/solid. The products **8am**, **8as**, **8dk**, **9er**, **9eq**, **9ar** are reported in literature.15

**GP-2 [General procedure for preparation of 8**, **9 (condition B)]:**

GP-1 was carried out with *ortho*-iodoketone **6a-d**, **11d** (104-152 mg, 0.40 mmol**)** andaldehyde **5 (**166.4-366.5 mg, 1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 20-30 h. Progress of the products **8an-dk**, **9dq-dr** formation was monitored by TLC till the reaction was completed. Then reaction mixture was removed from oil bath and allow to reach room temperature and the reaction mixture was quenched by the addition of aqueous NaHCO3 solution and then extracted with ethyl acetate (3 × 15 mL). The organic layer was washed with saturated NaCl solution, dried over Na2SO4 and filtered. Evaporation of the solvent under reduced pressure and purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate) furnished the products **8**, **9** (68.7-104.0 mg, 65-75 %) as viscous liquid/solid.

**2-methyl-3-phenyl-1*H*-inden-1-one (8aa**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5a (**166.4 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 35 minute for the product **8aa** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8aa** (55.4 mg, 63%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 96:04), *Rf*(**6a**)=0.50, *Rf*(**8aa**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1870, 1680, 1591, 1470, 1280, 1189, 954, 718 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.53-7.43 (m, 6H), 7.31-7.26 (m, 1H), 7.20-7.16 (m, 1H), 7.05 (d, 1H, *J*=7.3 Hz), 1.92 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.3 (Cq), 154.7 (Cq), 145.7 (Cq), 133.1 (CH), 132.7 (Cq), 131.1 (s, C=C), 131.0 (Cq), 129.2 (CH), 128.7 ( 2×CH), 128.1 (CH), 128.0 (2×CH), 122.5 (CH), 120.4 (CH), 8.6 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H13O]+=[M+H]+: 221.0961; found 221.0961.

**2-methyl-3-(p-tolyl)-1*H*-inden-1-one (8ab**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5b (**192.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 25 minute for the product **8ab** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ab** (60.8 mg, 65%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 96:04), *Rf*(**6a**)=0.50, *Rf*(**8ab**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1812, 1686, 1597, 1456, 1287, 1189, 939, 718 cm-11H NMR (CDCl3, 400 MHz): *δ*=7.46 (d, 1H, *J*=6.8 Hz), 7.39-7.37 (m, 2H), 7.33-7.26 (m, 3H), 7.18 (t, 1H, *J*=7.0 Hz), 7.07 (d, 1H, *J*=7.3 Hz), 2.43 (s, 3H) 1.92 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.4 (Cq), 154.8 (Cq), 145.7 (Cq), 139.3 (Cq), 133.0 (CH), 131.3 (Cq), 130.6 (Cq), 129.8 (Cq), 129.4 (2×CH), 128.0 (3×CH),122.4 (CH), 120.4(CH), 21.5 (CH3), 8.6 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C17H15O]+=[M+H]+: 235.1117; found 235.1107.

**3-(4-ethylphenyl)-2-methyl-1*H*-inden-1-one (8ac**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5c (**214.6 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 13 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 30 minute for the product **8ac** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ac** (58.5mg, 59%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 96:04), *Rf*(**6a**)=0.50, *Rf*(**8ac**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2938, 1819, 1686, 1599, 1457, 1287, 1088, 930, 718 cm-1 1H NMR (CDCl3, 400 MHz): *δ*=7.46 (d, 1H, *J*=7.3 Hz), 7.42-7.40 (m, 2H), 7.34 (d, 2H, *J*=8.3 Hz), 7.28 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.17 (t, 1H, *J*=7.3 Hz), 7.07 (d, 1H, *J*=7.3 Hz), 2.73 (q, 2H, *J*=7.3 Hz), 1.93 (s, 3H), 1.30 (t, 3H, *J*=7.3 Hz), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.4 (Cq), 154.8 (Cq), 145.7 (Cq), 145.6 (Cq), 133.0 (CH), 131.3 (Cq), 130.6 (Cq), 130.0 (Cq), 128.2 (2×CH), 128.1 (2×CH),128.0 (CH), 122.4(CH), 120.4(CH), 28.8 (CH2), 15.4 (CH2), 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C18H17O]+=[M+H]+: 249.1274; found 249.1274.

**2-methyl-3-(naphthalen-1-yl)-1*H*-inden-1-one (8ad**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5d (**249.6 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 15 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for30 minute for the product **8ad** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ad** (62.7 mg, 58%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 96:04), *Rf*(**6a**)=0.50, *Rf*(**8ad**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 2829, 1689, 1590, 1450, 1287, 1187, 938, 719 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.95 (dd, 2H, *J*=8.3 and *J*=4.4 Hz), 7.76 (d, 1H, *J*=8.3 Hz), 7.60-7.54 (m, 1H), 7.53-7.51 (m, 2H), 7.47-7.44 (m, 2H), 7.20-7.18 (m, 2H), 6.65-6.63 (m, 1H), 1.76 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.2 (Cq), 154.9 (Cq), 146.8 (Cq), 133.8 (Cq), 133.4 (CH), 133.3 (Cq), 130.6 (Cq), 130.6 (Cq), 130.4 (Cq), 129.3 (CH), 128.6 (CH), 128.1 (CH), 126.3(CH), 126.3(CH), 125.8(CH), 125.7(CH), 125.4(CH), 122.3(CH), 120.8(CH), 8.8 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C20H15O]+=[M+H]+: 271.1117; found 271.1121.

**3-(4-fluorophenyl)-2-methyl-1*H*-inden-1-one (8ae**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5e (**198.4 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 30 minute for the product **8ae** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ae** (64.7 mg, 68%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 96:04), *Rf*(**6aa**)=0.50, *Rf*(**8ae**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1891, 1687, 1597, 1460, 1287, 1089, 938, 728 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.48-7.44 (m, 3H), 7.30 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.23-7.17 (m, 3H), 7.02 (d, 1H, *J*=6.8 Hz), 1.90 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.0 (Cq), 164.2 (Cq, 1*J*C-F=254 Hz), 161.7 (Cq), 153.7 (Cq), 145.5 (Cq), 133.2 (CH), 131.1 (Cq), 131.0 (Cq), 130.0 (CH), 129.9 (CH), 128.7 (Cq), 128.2 (CH), 122.6 (CH), 120.2 (CH), 116.0 (CH), 115.8 (CH), 8.6 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H12FO]+=[M+H]+: 239.0867; found 239.0870.

**3-(4-chlorophenyl)-2-methyl-1*H*-inden-1-one (8af**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5f (**224.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 30 minute for the product **8af** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8af** (69.2 mg, 68%)as a yellow solid, m.p=75-77°C. [TLC control (petroleum ether/ethyl acetate 96:04), *Rf*(**6a**)=0.50, *Rf*(**8af**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2939, 1810, 1683, 1590, 1453, 1287, 1080, 938, 719 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.50-7.47 (m, 3H), 7.43-7.40 (m, 2H), 7.30 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.19 (t, 1H, *J*=7.8 Hz), 7.01 (d, 1H, *J*=7.3 Hz), 1.90 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.9 (Cq), 153.4 (Cq), 145.3 (Cq), 135.1 (Cq), 133.2 (CH), 131.4 (Cq), 131.1 (Cq), 130.9 (Cq), 129.4 (d, 2×CH), 129.1 (2×CH), 128.3 (CH), 122.7 (CH), 122.2 (CH), 8.6 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H12ClO]+=[M+H]+: 255.0571; found 255.0567.

**3-(4-bromophenyl)-2-methyl-1*H*-inden-1-one (8ag**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5g (**296.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 15 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 30 minute for the product **8ag** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ag** (73.8 mg, 62%)as a yellow solid, m.p=71-74°C. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.50, *Rf*(**8ag**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 2821, 1686, 1599, 1450, 1287, 1087, 945, 710 cm-1.1H NMR (CDCl3, 400 MHz): *δ*=7.64 (d, 2H, *J*=8.3 Hz), 7.47 (d, 1H, *J*=7.3 Hz), 7.36-7.33 (m, 2H), 7.29 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.20-7.17 (m, 1H), 7.0 (d, 1H, *J*=7.3 Hz), 1.89 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.9 (Cq), 153.4 (Cq), 145.2 (Cq), 133.2 (CH), 132.0 (2×CH), 131.5 (Cq), 131.4 (Cq), 130.9 (Cq), 129.6 (2C×CH), 128.2 (CH), 123.3 (Cq), 122.7(CH), 120.1 (CH), 8.6 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H12BrO]+=[M+H]+: 299.0066; found 299.0079.

**3-(2-bromophenyl)-2-methyl-1*H*-inden-1-one (8ah**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5h (**296.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 13 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 30 minute for the product **8ah** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ah** (69.3 mg, 58%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.50, *Rf*(**8ah**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2933, 2829, 1689, 1598, 1450, 1287, 1087, 945, 710 cm-1.1H NMR (CDCl3, 400 MHz): *δ*=7.72 (dd, 1H, *J*=8.0 and *J*=1.2 Hz), 7.47-7.41 (m, 2H), 7.32-7.23 (m, 3H), 7.18-7.15 (m, 1H), 6.70 (d, 1H, *J*=7.3 Hz), 1.77 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.0 (Cq), 154.7 (Cq), 145.7 (Cq), 134.2 (Cq), 133.4 (CH), 133.3 (CH), 133.0 (Cq), 130.3 (Cq), 130.2 (CH), 129.6 (CH), 128.0 (CH), 127.5 (CH), 122.5 (CH), 122.2 (Cq), 120.5 (CH), 8.8 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H12BrO]+=[M+H]+: 299.0066; found 299.0060

**3-(2,4-dichlorophenyl)-2-methyl-1*H*-inden-1-one (8ai**)GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5i (**280.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 30 minute for the product **8ai** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 97:03 to 95:05) furnished the product **8ai** (65.9 mg, 57%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.50, *Rf*(**8ai**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2939, 1810, 1683, 1590, 1453, 1287, 1080, 938, 719 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.57 (d, 1H, *J*=1.9 Hz), 7.47 (d, 1H, *J*=6.8 Hz), 7.37 (dd, 1H, *J*=8.0 and *J*=2.2 Hz),7.29-7.23 (m, 2H), 7.18 (t, 1H, *J*=7.3 Hz), 6.71 (d, 1H, *J*=7.3 Hz), 1.77 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.6 (Cq), 151.9 (Cq), 145.3 Cq), 135.4 (Cq), 133.8 (Cq), 133.5 (CH), 130.5 (Cq), 130.5 (CH), 130.2 (2×Cq), 130.2 (CH), 128.2 (CH), 127.4 (CH), 122.7 (CH), 120.3 (CH), 8.8 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H11Cl2O]+=[M+H]+: 289.0181; found 289.0188.

**3-(4-hydroxyphenyl)-2-methyl-1*H*-inden-1-one (8aj**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5j (**195.2 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 15 minute for the product **8aj** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **8aj** (61.3 mg, 65%)as a yellow solid, m.p=168-171°C [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6a**)=0.50, *Rf*(**8aj**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2878, 1710, 1686, 1589, 1451, 1297, 1187, 947, 711 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.46 (d, 1H, *J*=6.3 Hz), 7.42-7.38 (m, 2H), 7.29 (td, 2H, *J*=7.4 and *J*=1.2 Hz), 7.20-7.16 (m, 1H), 7.08 (d, 1H, *J*=7.3 Hz), 7.00-6.97 (m, 2H), 5.43 (brd. s , 1H), 1.92 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.5 (Cq), 156.5 (Cq), 154.6 (Cq), 145.7 (Cq), 133.0 (CH), 131.4 (Cq), 130.1 (Cq), 129.8 (2×CH), 128.1 (CH), 125.2 (Cq), 122.4 (CH), 120.4 (CH), 115.6 (2×CH), 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C16H13O2]+=[M+H]+: 237.0910; found 237.0899.

**3-(4-methoxyphenyl)-2-methyl-1*H*-inden-1-one (8ak**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5k (**217.6 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 20 minute for the product **8ak** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **8ak** (62.1mg, 62%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.50, *Rf*(**8ak**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2878, 1710, 1686, 1589, 1451, 1297, 1187, 947, 711 cm-1.1H NMR (CDCl3, 400 MHz): *δ*=7.48-7.44 (m, 3H), 7.29 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.20-7.16 (m, 1H), 7.09 (d, 1H, *J*=7.3 Hz), 7.02 (d, 2H, *J*=8.8 Hz), 3.88 (s, 3H), 1.92 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.3 (Cq), 160.3 (Cq), 154.5 (Cq), 145.7 (Cq), 133.0 (CH), 131.4 (Cq), 130.1 (Cq), 129.6 (2×CH), 128.0 (CH), 125.1 (Cq), 122.3 (CH), 120.4 (CH), 114.1 (2×CH), 55.4 (CH3), 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C17H15O2]+=[M+H]+: 251.1067; found 251.1063.

**2-methyl-3-(4-propoxyphenyl)-1*H*-inden-1-one (8al**): GP-1 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**)** andaldehyde **5l (**262.7 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 15 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 20 minute for the product **8al** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **8al** (62.3 mg, 56%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 94:06), *Rf*(**6a**)=0.50, *Rf*(**8al**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 2821, 1686, 1599, 1450, 1287, 1087, 945, 710 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.45-7.42 (m, 3H), 7.27 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.16 (t, 1H, *J*=7.3 Hz), 7.09 (d, 1H, *J*=6.8 Hz), 7.04-7.00 (m, 2H), 3.98 (t, 2H, *J*=6.6 Hz), 1.92 (s, 3H), 1.89-1.80 (m, 2H), 1.06 (t, 3H, *J*=7.5 Hz), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.3 (Cq), 159.9 (Cq), 154.5 (Cq), 145.7 (Cq), 132.9 (CH), 131.4 (Cq), 129.9 (Cq), 129.6 (2×CH), 127.9 (CH), 124.7 (Cq), 122.2 (CH), 120.4 (CH), 114.6 (2×CH), 69.6 (CH2), 22.5 (CH2), 10.5 (CH3) 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C19H19O2]+=[M+H]+: 279.1380; found 279.1382.

**3-(benzo[d][1,3]dioxol-5-yl)-2-methyl-1*H*-inden-1-one (8an**): GP-2 was carried out *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**),** aldehyde **5n (**256.0 mg,1.6 mmol) , Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 28 h for the product **8an** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **8an** (69.7 mg, 66%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.80, *Rf*(**8an**)=0.30, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2878, 1710, 1689, 1596, 1481, 1287, 1187, 937, 721 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.45 (d, 1H, *J*=6.3 Hz), 7.28 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.17 (t, 1H, *J*=7.3 Hz), 7.06 (d, 1H, *J*=6.8 Hz), 7.00-6.92 (m, 3H), 6.03 (s, 2H), 1.91 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.1 (Cq), 154.4 (Cq), 148.3 (Cq), 147.9 (Cq), 145.5 (Cq), 133.0 (CH), 131.2 (Cq), 130.4 (Cq), 128.0 (CH), 126.4 (Cq), 122.3 (CH), 122.3 (CH), 120.3 (CH), 108.6 (CH), 108.4 (CH), 101.4 (CH2), 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C17H13O3]+=[M+H]+: 265.0859; found 265.0848.

**3-(3,4-dimethoxyphenyl)-2-methyl-1*H*-inden-1-one (8ao**): GP-2 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**),** aldehyde **5o (**265.6 mg,1.6 mmol) , Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 28 h for the product **8ao** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **8ao** (80.7 mg, 72%)as yellow solid, m.p=102-104°C. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.70, *Rf*(**8ao**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2934, 1711, 1689, 1590, 1451, 1285, 1134, 1079, 930, 716 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.45 (d, 1H, *J*=6.3 Hz), 7.28 (td, 1H, *J*=7.4 and *J*=1.2 Hz), 7.19-7.15 (m, 1H), 7.11-7.07 (m, 2H), 6.99 (dd, 2H, *J*=5.1 and *J*=3.1 Hz), 3.94 (s, 3H), 3.91 (s, 3H), 1.93 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.2 (Cq), 154.6 (Cq), 149.8 (Cq), 148.9 (Cq), 145.6 (Cq), 133.0 (CH), 131.3 (Cq), 130.2 (Cq), 128.0 (CH), 125.3 (Cq), 122.3 (CH), 121.2 (CH), 120.3 (CH), 111.1 (CH), 111.0 (CH), 56.0 (CH3), 55.9 (CH3), 8.7 ( CH3) ppm. HR-MS (ESI+) m/z calculated for [C18H17O3]+=[M+H]+: 281.1172; found 281.1173.

**2-methyl-3-(3,4,5-trimethoxyphenyl)-1*H*-inden-1-one (8ap**): GP-2 was carried out with *ortho*-iodoketone **6a (**104.0mg, 0.40mmol**),** aldehyde **5p (**313.9 mg,1.6 mmol) , Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 28 h for the product **8ap** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **8ap** (93 mg, 75%)as a yellow solid, m.p=109-112°C. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**6a**)=0.80, *Rf*(**8ap**)=0.50, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2878, 1710, 1686, 1589, 1451, 1297, 1187, 947, 711 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.45 (d, 1H, *J*=6.8 Hz), 7.31-7.27 (m, 1H), 7.17 (t, 1H, *J*=7.0 Hz), 7.09 (d, 1H, *J*=7.3 Hz), 6.67 (s, 2H), 3.91 (s, 3H), 3.88 (s, 6H), 1.93 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.1 (Cq), 154.7 (Cq), 153.4 (3× Cq), 145.6 (Cq), 138.7 (Cq), 133.1 (CH), 131.1 (Cq), 130.7 (Cq), 128.1 (CH), 122.5 (CH), 120.3 (CH), 105.2 (2×CH), 60.9 (CH3), 56.2 (2×CH3), 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C19H18O4]+=[M]+: 310.1205; found 310.1202.

**6-methyl-7-(p-tolyl)-5*H*-indeno[5,6-d][1,3]dioxol-5-one (8bb**): GP-1 was carried out with *ortho*-iodoketone **6b (**121.6 mg, 0.40mmol**)** andaldehyde **5b (**192.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 14 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 15 minute for the product **8bb** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **8bb** (84.6 mg, 76%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6b**)=0.30, *Rf*(**8bb**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1757, 1690, 1590, 1450, 1286, 1089, 930, 719 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.34-7.29 (m, 4H), 6.98 (s, 1H), 6.59 (s, 1H), 5.97 (s, 2H), 2.42 (s, 3H), 1.86 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=196.9 Cq), 152.9 (Cq), 151.2 (Cq), 146.9 (Cq), 142.4 (Cq), 139.2 (Cq), 129.7 (Cq), 129.6 (Cq), 129.4 (2×CH), 127.9 (2×CH), 125.1 (Cq), 105.0 (CH), 103.5 (CH), 101.9 (CH2), 21.4 (CH3) 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C18H15O3]+=[M+H]+: 279.1016; found 279.1011.

**5,6-dimethoxy-2-methyl-3-(p-tolyl)-1*H*-inden-1-one (8cb**): GP-1 was carried out with *ortho*-iodoketone **6c (**128.0 mg, 0.40mmol**)** andaldehyde **5b (**192.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 13 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 10 minute for the product **8cb** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **8cb** (80.0 mg, 68%)as a yellow solid, m.p=158-160°C. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6c**)=0.30, *Rf*(**8cb**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1808, 1680, 1595, 1450, 1287, 1189, 936, 710 cm-1 1H NMR (CDCl3, 400 MHz): *δ*=7.34 (dd, 4H, *J*=7.3 and , *J*=1.2 Hz), 7.10 (s, 1H), 6.63 (s, 1H), 3.88 (s, 3H), 3.85 (s, 3H), 2.43 (s, 3H), 1.86 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.9 (Cq), 153.1 (Cq), 152.3 (Cq), 148.2 (Cq), 140.2 (Cq), 139.2 (Cq), 129.9 (Cq), 129.4 (Cq), 129.4 (2×CH), 127.9 (2×CH), 123.4 (Cq), 107.4 (CH), 105.4 (CH), 56.3 (CH3), 56.3 (CH3), 21.4 (CH3) 8.6 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C19H19O3]+=[M+H]+: 295.1329; found 295.1324.

**5,6-dimethoxy-3-(4-methoxyphenyl)-2-methyl-1*H*-inden-1-one (8ck**): GP-2 was carried out with *ortho*-iodoketone **6c (**128.0mg, 0.40mmol**),** aldehyde **5k (**217.6 mg,1.6 mmol) , Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 28 h for the product **8ck** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **8ck** (91.8 mg, 74%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6c**)=0.50, *Rf*(**8ck**)=0.70, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2931, 1729, 1689, 1591, 1456, 1287, 1088, 934, 722 cm-1.1H NMR (CDCl3, 400 MHz): *δ*=7.42 (d, 2H, *J*=8.8 Hz), 7.10 (s, 1H), 7.02 (d, 2H, *J*=8.8 Hz), 6.65 (s, 1H), 5.28 (DCM), 3.88 (s, 3H), 3.88 (s, 3H), 3.86 (s, 3H), 1.87 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.8 (Cq), 160.2 (Cq), 152.9 (Cq), 152.2 (Cq), 148.2 (Cq), 140.2 (Cq), 129.5 (2×CH), 129.0 (Cq), 125.2 (Cq), 123.6 (Cq), 114.2 (2×CH), 107.4 (CH), 105.5 (CH), 56.4 (*C*H3), 56.3 (*C*H3), 55.3 (*C*H3), 8.7 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C19H19O4]+=[M+H]+: 311.1278; found 311.1274.

**3-(benzo[d][1,3]dioxol-5-yl)-5,6-dimethoxy-2-methyl-1*H*-inden-1-one (8cn**)1e: GP-2 was carried out with *ortho*-iodoketone **6c (**128.0mg, 0.40mmol**),** aldehyde **5n (**240.0 mg,1.6 mmol) , Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 28 h for the product **8cn** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 85:15) furnished the product **8cn** (68.7 mg, 53%)as yellow solid, m.p=214-216°C. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6c**)=0.50, *Rf*(**8cn**)=0.70, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2931, 1719, 1685, 1590, 1456, 1287, 1088, 935, 728 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.11 (s, 1H), 6.96-6.93 (m, 3H), 6.64 (s, 1H), 6.05 (s, 2H), 3.89 (s, 3H), 3.87 (s, 3H), 1.87 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.3 (Cq), 152.9 (Cq), 152.3 (Cq), 148.3 (Cq), 148.2 (Cq), 148.0 (Cq), 140.1 (Cq), 129.3 (Cq), 126.6 (Cq), 123.4 (Cq), 122.1 (CH), 108.7 (CH), 108.3 (CH), 107.5 (CH), 105.4 (CH), 101.4 (CH2), 56.4 (2×CH3), 8.8 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C19H17O5]+=[M+H]+: 325.1071; found 325.1063.

**3-(3,4-dimethoxyphenyl)-5,6-dimethoxy-2-methyl-1*H*-inden-1-one (8co**): GP-2 was carried out with *ortho*-iodoketone **6c (**128.0mg, 0.40mmol**),** aldehyde **5o (**265.6 mg,1.6 mmol) , Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 28 h for the product **8co** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 90:10 to 85:15) furnished the product **8co** (88.4 mg, 65%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6c**)=0.50, *Rf*(**8co**)=0.70, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2934, 1719, 1689, 1587, 1451, 1387, 1243, 1084, 905, 708 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.10 (s, 1H), 7.06 (dd, 1H, *J*=7.3 and *J*=1.9 Hz), 7.01-6.97 (m, 2H), 6.67 (s, 1H), 3.95 (s, 3H), 3.91 (s, 3H), 3.88 (s, 3H), 3.85 (s, 3H), 1.89 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.7 (Cq), 153.0 (Cq), 152.2 (Cq), 149.7 (Cq), 149.0 (Cq), 148.3 (Cq), 140.2 (Cq), 129.0 (Cq), 125.4 (Cq), 123.5 (Cq), 121.0 (CH), 111.3 (CH), 111.0 (CH), 107.4 (CH), 105.5 (CH), 56.4 (CH3), 56.3 (CH3), 56.0 (CH3), 55.9 (CH3), 8.8 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C20H21O5]+=[M+H]+: 341.1384; found 341.1383.

**4,5,6-trimethoxy-2-methyl-3-(p-tolyl)-1*H*-inden-1-one** (**8db**): GP-1 was carried out with *ortho*-iodoketone **6d (**140.0 mg, 0.40mmol**)** andaldehyde **5b (**192.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 10 minute for the product **8db** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **8db** (101.9 mg, 79%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6d**)=0.30, *Rf*(**8db**)=0.70, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2898, 1719, 1686, 1588, 1451, 1345, 1297, 1187, 947, 711 cm-1.NMR (CDCl3, 400 MHz): *δ*=7.32 (d, 2H, *J*=7.8 Hz), 7.24 (d, 2H, *J*=7.8 Hz), 6.99 (s, 1H), 3.88 (s, 3H), 3.85 (s, 3H), 3.33 (s, 3H), 2.41 (s, 3H), 1.77 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.3 (Cq), 155.6 (Cq), 153.4 (Cq), 148.4 (Cq), 147.2 (Cq), 138.4 (Cq), 131.1 (Cq), 130.4 (Cq), 129.3 (Cq), 128.4 (2×CH), 128.0 (2×CH), 127.2 (Cq), 104.6 (CH), 61.2 (CH3), 61.0 (CH3), 56.4 (CH3), 21.4 (CH3), 8.4 ( CH3) ppm. HR-MS (ESI+) m/z calculated for [C20H21O4]+=[M+H]+: 325.1434; found 325.1438.

**3-(4-chlorophenyl)-4,5,6-trimethoxy-2-methyl-1*H*-inden-1-one (8df**): GP-1 was carried out with *ortho*-iodoketone **6d (**140.0 mg, 0.40mmol**)** andaldehyde **5f (**224.0 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 14 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 15 minute for the product **8df** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **8df** (102.9 mg, 75%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**6d**)=0.30, *Rf*(**8df**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1910, 1689, 1592, 1453, 1288, 1089, 930, 738 cm-11H NMR (CDCl3, 400 MHz): *δ*=7.41 (d, 2H, *J*=8.8 Hz), 7.36 (d, 2H, *J*=8.8 Hz), 6.99 (s, 1H,), 3.87 (s, 3H), 3.85 (s, 3H), 3.33 (s, 3H), 1.75 (s, 3H), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=196.9 (Cq), 154.0 (Cq), 153.6 (Cq), 148.3 (Cq), 147.3 (Cq), 134.3 (Cq), 132.6 (Cq), 131.1 (Cq), 129.6 (2×CH), 128.9 (Cq), 128.0 ( 2×CH), 126.9 (Cq), 104.9 (CH), 61.1 (CH3), 61.0 (CH3), 56.5 (CH3), 8.4 (CH3) ppm. HR-MS (ESI+) m/z calculated for [C19H18ClO4]+=[M+H]+: 345.0888; found 345.0881.

**3-(4-methoxyphenyl)-2-propyl-1*H*-inden-1-one (9br**): GP-1 was carried out with *ortho*-iodoketone **11b (**135.2 mg, 0.40mmol**)** andaldehyde **5r (**137.6 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 12 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 20 minute for the product **9br** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **9br** (64.5 mg, 58%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**11b**)=0.30, *Rf*(**9br**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2978, 1700, 1689, 1590, 1452, 1297, 1187, 945, 721 cm-1.. 1H NMR (CDCl3, 400 MHz): *δ*=7.45 (d, 1H, *J*=6.8 Hz), 7.40 (d, 2H, *J*=8.8 Hz), 7.28 (td, 1H, *J*=7.5 and *J*=0.9 Hz), 7.20-7.16 (m, 1H), 7.05-7.02 (m, 3H), 3.88 (s, 3H), 2.33-2.29 (m, 2H), 1.58-1.47 (m, 2H), 0.89 (t, 3H, *J*=7.3 Hz), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=198.4 (Cq), 160.2 (Cq), 154.9 (Cq), 145.9 (Cq), 134.6 (Cq), 133.0 (CH), 131.2 (Cq), 129.3 (2×CH), 128.1 (CH), 125.1 (Cq), 122.2 (CH), 120.4 (CH), 114.1 (2×CH), 55.3 (CH3), 25.4 (CH2), 22.6 (CH2), 14.3 (CH3), ppm. HR-MS (ESI+) m/z calculated for [C19H19O2]+=[M+H]+: 279.1380; found 279.1385.

**2-propyl-3-(thiophen-2-yl)-1*H*-inden-1-one (9cr**): GP-1 was carried out with *ortho*-iodoketone **11c (**125.6 mg, 0.40mmol**)** andaldehyde **5r (**137.6 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 17 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 20 minute for the product **9cr** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 93:07) furnished the product **9cr** (59.0 mg, 58%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 95:05), *Rf*(**11c**)=0.50, *Rf*(**9cr**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2930, 1790, 1686, 1590, 1455, 1277, 1099, 984, 716 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.49 (dd, 1H, *J*=5.1 and *J*=1.2 Hz), 7.41-7.40 (m, 2H), 7.34 (d, 1H, *J*=7.3 Hz), 7.29-7.25 (m, 2H), 7.17-7.12 (m, 2H), 2.44-2.40 (m, 2H), 1.56-1.46 (m, 2H), 0.90 (t, 3H, *J*=7.3 Hz), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.4 (Cq), 147.2 (Cq), 144.9 (Cq), 135.0 (Cq), 134.3 (Cq), 133.0 (CH), 131.1 (Cq), 128.6 (CH), 128.3 (CH), 128.1 (CH), 127.8 (CH), 122.4 (CH), 120.8 (CH), 25.7 (CH2), 22.5 (CH2), 14.3 (CH3), ppm. HR-MS (ESI+) m/z calculated for [C16H15OS]+=[M+H]+: 255.0838; found 255.0841.

**6-ethyl-7-(4-methoxyphenyl)-5*H*-indeno[5,6-d][1,3]dioxol-5-one (9dr**): GP-1 was carried out with *ortho*-iodoketone **11d (**152.8 mg, 0.40mmol**)** andaldehyde **5q (**115.3 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 18 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 5 minute for the product **9dr** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **9dr** (75.1mg, 61%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**11d**)=0.30, *Rf*(**9dr**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2878, 1714, 1696, 1589, 1461, 1297, 1187, 947, 741 cm-1.1H NMR (CDCl3, 400 MHz): *δ*=7.36 (d, 2H, *J*=8.8 Hz), 7.01 (d, 2H, *J*=8.8 Hz), 6.97 (s, 1H), 6.56 (s, 1H), 5.97 (s, 2H), 3.87 (s, 3H), 2.29 (q, 2H, *J*=7.3 Hz),1.08 (t, 3H, *J*=7.3 Hz), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=196.9 (Cq), 160.2 (Cq), 152.6 (Cq), 151.1 (Cq), 147.0 (Cq), 142.4 (Cq), 134.9 (Cq), 129.2 (2×CH), 125.1 (Cq), 125.1 (Cq), 114.2 (2×CH), 104.8 (CH), 103.6 (CH), 101.9 (CH2), 55.3 (CH3), 16.8 (CH2), 14.1 (CH3), ppm. HR-MS (ESI+) m/z calculated for [C19H17O4]+=[M+H]+: 309.1121; found 309.1126.

**7-(4-methoxyphenyl)-6-propyl-5*H*-indeno[5,6-d][1,3]dioxol-5-one (9dr**): GP-1 was carried out with *ortho*-iodoketone **11d (**152.8 mg, 0.40mmol**)** andaldehyde **5r (**137.6 mg,1.6 mmol) in presence of Pd(OAc)2 (5.0 mg, 5 mol %), Ag2O (111.3 mg, 0.48 mmol) and TBHP (257.1 mg, 2.0 mmol) and allowed the reaction mixture to stir at 120 °C for 20 h then Then reaction mixture was removed from oil bath and allow to reach room temperature and then added Conc. H2SO4 (0.1 ml, 2.0 mmol) and allowed the reaction mixture stirred at room temperature for 10 minute for the product **9dr** formation**.** Purification of the crude material by silica gel column chromatography (petroleum ether/ethyl acetate, 95:05 to 90:10) furnished the product **9dr** (72.1 mg, 56%)as a yellow viscous liquid. [TLC control (petroleum ether/ethyl acetate 90:10), *Rf*(**11d**)=0.30, *Rf*(**9dr**)=0.80, UV detection]. IR (MIR-ATR, 4000–600 cm-1): *νmax*=2931, 1871, 1687, 1590, 1459, 1281, 1189, 933, 728 cm-1. 1H NMR (CDCl3, 400 MHz): *δ*=7.35 (d, 2H, *J*=8.8 Hz), 7.01 (d, 2H, *J*=8.8 Hz), 6.97 (s, 1H), 6.54 (s, 1H), 5.97 (s, 2H), 3.87 (s, 3H), 2.26-2.22 (m, 2H), 1.51-1.45 (m, 2H), 0.87 (t, 3H, *J*=7.3 Hz), ppm. 13C NMR (CDCl3, 100 MHz): *δ*=197.0 (Cq), 160.1 (Cq), 153.1 (Cq), 151.1 (Cq), 146.9 (Cq), 142.4 (Cq), 133.6 (Cq), 129.2 (2×CH), 125.1 (Cq), 125.0 (Cq), 114.2 (2×CH), 104.8 (CH), 103.5 (CH), 101.8 (CH2), 55.3 (CH3), 25.4 (CH2), 22.7 (CH2), 14.2 (CH3), ppm. HR-MS (ESI+) m/z calculated for [C20H19O4]+=[M+H]+: 323.1278; found 323.1280.

ASSOCIATED CONTENT

Supporting Information

Copies of 1H-NMR and 13C-NMR spectra of all compounds and CIF file for **8ap** are provided. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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