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# $ZrOCl_2 \cdot 8H_2O$: an efficient Lewis acid catalyst for the one-pot multicomponent synthesis of β-acetamido ketones

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**Abstract**—Aromatic aldehydes were reacted in one-pot with enolisable ketones, acetonitrile and acetyl chloride at ambient temperature in the presence of $ZrOCl_2 \cdot 8H_2O$ to furnish the corresponding β-acetamido ketones in very good to excellent yields. X-ray crystallographic analysis of one *anti*-β-acetamido ketone exhibited a two-dimensional supramolecular framework by a combination of N–H···O, C–H···O and C–H···π (arene) hydrogen bonds.
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## 1. Introduction

Due to several advantages over conventional multi-step synthesis and also because of their promising applications in pharmaceutical chemistry for the generation of structural scaffolds and combinatorial libraries for drug development,[1] one-pot multicomponent reactions have recently drawn the attraction of organic chemists. β-Acetamido or -amino ketones are potential intermediates for the generation of β-amino alcohols[2]—structural units common in natural nucleoside antibiotics such as nikkomycins or neopolyoxins.[3]

The reported one-pot syntheses of the title compounds from aldehydes, enolisable ketones, acetyl chloride and acetonitrile are based on $CoCl_2$,[4a,b] Montmorillonite K-10 Clay,[4c] $SiO_2$–$H_2SO_4$[4d] or $BiCl_3$ generated in situ from BiOCl and acetyl chloride.[5] Recently, because of the easy availability in earth crust[6] and low toxicity,[7] Zr(IV) compounds, especially $ZrCl_4$ have received considerable attention in various organic reactions,[8] but reported zirconium oxychloride based reactions are only a few.[9] In one of our earlier reports we have established for the first time that $ZrOCl_2 \cdot 8H_2O$ acts as an efficient Lewis acid catalyst for acylation of alcohols, phenols, amines, thiol and thiophenols.[9a] In further continuation to our ongoing research on metal oxysalt-based organic reactions[5,9a,10] we were prompted to explore the efficacy of $ZrOCl_2 \cdot 8H_2O$ as an activator for the one-pot synthesis of β-acetamido ketones from aromatic aldehydes, enolisable ketones, acetyl chloride and acetonitrile.

## 2. Results and discussions

### 2.1. Synthesis

Under the optimized reaction conditions (Table 1, Scheme 1), benzaldehyde (**1**, ~1 equiv) reacted at room temperature with acetophenone (**11**, ~1 equiv), acetyl chloride (~2 equiv) and acetonitrile (reagent as well as solvent) in the presence of $ZrOCl_2 \cdot 8H_2O$ (~20 mol%) to furnish the corresponding β-acetamido ketone (**15**) in excellent yield (90%, entry 2, Table 1 and entry 1, Table 2). Other benzaldehyde derivatives containing electron-withdrawing (entries 5–7, Table 2) and -donating groups (entries 8–10 and 12, Table 2) in the aromatic ring also reacted with acetophenone (**11**), acetyl chloride and acetonitrile affording the corresponding β-acetamido ketones (**16–21** and **23**) in very high to excellent yields with concomitant acetylation of

**Table 1**. Standardisation of reaction condition

| Entry | Metal salts (mol%) | Time (h) | Yield of product (%) |
|---|---|---|---|
| 1 | $ZrOCl_2 \cdot 8H_2O$ (15) | 12 | 83 |
| 2 | $ZrOCl_2 \cdot 8H_2O$ (20) | 5 | 90 |
| 3 | $ZrCl_4$ (20) | 5 | 87 |

*Keywords*: One-pot multicomponent synthesis; $ZrOCl_2 \cdot 8H_2O$; Lewis acid catalyzed; β-Acetamido ketone.
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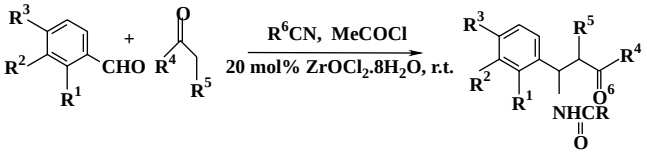
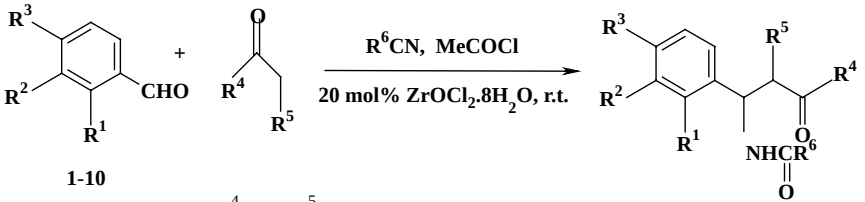



**Scheme 1.**

the *m*-hydroxy benzaldehyde derived product (entry 10, Table 2). *p*-Dimethylaminobenzaldehyde, however, was inert to the present reaction conditions. Other enolisable ketones (α-unsubstituted and -substituted, **12**–**14**) also served as good substrates for the present one-pot synthesis. Thus, the reaction of benzaldehyde (**1**) with *p*-methoxyacetophenone (**12**), acetyl chloride and acetonitrile in the presence of $ZrOCl_2 \cdot 8H_2O$ proceeded efficiently resulting in the desired product (**22**) in 92% yield (entry 11, Table 2). α-Substituted enolisable ketones such as ethyl methyl ketone (**13**) also reacted with benzaldehyde (**1**) or *p*-tolualdehyde (**8**) resulting in high yields of the title compounds (**26** and **27**), although, with moderate or poor diastereoselectivity in favour of the *anti*-isomers (entries 16 and 17, Table 2). Similarly, propiophenone (**14**) was equally a good substrate to react separately with benzaldehyde (**1**), *p*-chloro-(**5**), *o*-nitro-benzaldehyde (**2**), *p*-tolualdehyde (**8**) or vanilin (**9**) in the presence of other components affording the corresponding β-acetamido ketones (**28**–**32**, entries 18–22, Table 2) with concomitant acetylation of the phenolic –OH groups of the products in relevant system (entry 22, Table 2), but the reactions were either not diastereoselective (entries 18–20) or proceeded with poor diastereoselectivity (entries 21 and 22). The reaction of salicylaldehyde (**10**) with propiophenone (**14**) and other components was, however, tricky; only 31% of the desired acetylated product (**33**) could be isolated (entry 23). The preparative efficacy of this one-pot synthesis was further checked by scaling-up (∼10 folds) of the reaction of benzaldehyde (**1**) with acetophenone (**11**) and other ingredients in solvent as well as in solvent-free conditions (entries 2 and 3, Table 2), which proceeded in 92 and 93% respective yields. The reaction also proceeded with equal efficacy (entry 4, Table 2) with the recovered and isolated $ZrOCl_2 \cdot 8H_2O$. It may be mentioned here that a mixture of benzaldehyde (**1**),

**Table 2**. $ZrOCl_2 \cdot 8H_2O$ catalyzed one-pot synthesis of β-acetamido ketone




**1-10**

**11,** $R^4$ = Ph, $R^5$ = H
**12,** $R^4$ = *p*-OMe-$C_6H_4$-, $R^5$ = H
**13,** $R^4$ = Me, $R^5$ = Me
**14,** $R^4$ = Ph, $R^5$ = Me

**15-30**

| Entry | Aromatic aldehyde | β-Acetamido ketone | Time (h) | Yield (%)[a] | *syn*/*anti* |
|---|---|---|---|---|---|
| 1 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**15**) | 5 | 90 | — |
| 2 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**15**) | 5 | 92[b] | — |
| 3 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**15**) | 5 | 93[c] | — |
| 4 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**15**) | 5 | 90[d] | — |
| 5 | $R^1$=$NO_2$, $R^2$=$R^3$=H (**2**) | $R^1$=$NO_2$, $R^2$=$R^3$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**16**) | 12 | 85 | — |
| 6 | $R^2$=$NO_2$, $R^1$=$R^3$=H (**3**) | $R^2$=$NO_2$, $R^1$=$R^3$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**17**) | 6 | 92 | — |
| 7 | $R^3$=$NO_2$, $R^1$=$R^2$=H (**4**) | $R^3$=$NO_2$, $R^1$=$R^2$=$R^5$=H, $R^4$=Ph, $R^6$=Me (**18**) | 7 | 69 | — |
| 8 | $R^1$=$R^2$=H, $R^3$=Cl (**5**) | $R^1$=$R^2$=$R^5$=H, $R^3$=Cl, $R^4$=Ph, $R^6$=Me (**19**) | 8 | 91 | — |
| 9 | $R^1$=$R^2$=H, $R^3$=OMe (**6**) | $R^1$=$R^2$=$R^5$=H, $R^3$=OMe, $R^4$=Ph, $R^6$=Me (**20**) | 7 | 92 | — |
| 10 | $R^1$=$R^3$=H, $R^2$=OH (**7**) | $R^1$=$R^3$=$R^5$=H, $R^2$=OAc, $R^4$=Ph, $R^6$=Me (**21**) | 4.5 | 92 | — |
| 11 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=$R^5$=H, $R^4$=*p*-MeO–$C_6H_4$, $R^6$=Me (**22**) | 4 | 92 | — |
| 12 | $R^3$=Me, $R^1$=$R^2$=H (**8**) | $R^1$=$R^2$=$R^5$=H, $R^4$=Ph, $R^3$=$R^6$=Me (**23**) | 4 | 94 | — |
| 13 | $R^2$=$NO_2$, $R^1$=$R^3$=H (**3**) | $R^1$=$R^3$=$R^5$=H, $R^2$=$NO_2$, $R^4$=*p*-OMe–$C_6H_4$, $R^6$=Me (**24**) | 48 | 61 | — |
| 14 | $R^2$=$NO_2$, $R^1$=$R^3$=H (**3**) | $R^2$=$NO_2$, $R^1$=$R^3$=$R^5$=H, $R^4$=$R^6$=Ph (**25**) | 36 | 86[e] | — |
| 15 | $R^2$=$NO_2$, $R^1$=$R^3$=H (**3**) | $R^2$=$NO_2$, $R^1$=$R^3$=$R^5$=H, $R^4$=$R^6$=Ph (**25**) | 36 | 89[f] | — |
| 16 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=H, $R^4$=$R^5$=$R^6$=Me (**26**) | 2 | 86 | 1:4.5[g] |
| 17 | $R^1$=$R^2$=H, $R^3$= Me (**8**) | $R^1$=$R^2$=H, $R^3$=$R^4$=$R^5$=$R^6$=Me (**27**) | 3.5 | 81 | 1:1.6[g] |
| 18 | $R^1$=$R^2$=$R^3$=H (**1**) | $R^1$=$R^2$=$R^3$=H, $R^4$=Ph, $R^5$=$R^6$=Me (**28**) | 6 | 94 | 1.1:1[h] |
| 19 | $R^1$=$R^2$=H, $R^3$=Cl (**5**) | $R^1$=$R^2$=H, $R^3$=Cl, $R^4$=Ph, $R^5$=$R^6$=Me (**29**) | 2 | 83 | 1:1[h] |
| 20 | $R^1$=$NO_2$, $R^2$=$R^3$=H (**2**) | $R^1$=$NO_2$, $R^2$=$R^3$=H, $R^4$=Ph, $R^5$=$R^6$=Me (**30**) | 10 | 80 | 1.1:1[h] |
| 21 | $R^1$=$R^2$=H, $R^3$=Me (**8**) | $R^1$=$R^2$=H, $R^3$=$R^5$=$R^6$=Me, $R^4$=Ph (**31**) | 4 | 94 | 2.2:1[g] |
| 22 | $R^1$=H, $R^2$=OMe, $R^3$=OH (**9**) | $R^1$=H, $R^2$=OMe, $R^3$=OAc, $R^4$=Ph, $R^5$=$R^6$=Me (**32**) | 12 | 74 | 1.5:1[h] |
| 23 | $R^1$=OH, $R^2$=$R^3$=H (**10**) | $R^1$=OAc, $R^2$=$R^3$=H, $R^4$=Ph, $R^5$=$R^6$=Me (**33**) | 11 | 31 | 1:1[h] |

[a] Chromatographed yield.
[b] Scale-up experiment (∼10 fold).
[c] Under neat condition (∼10 fold).
[d] With recovered $ZrOCl^2 \cdot 8H_2O$.[9]
[e] Using PhCN (∼2 equiv) in $CH^2Cl_2$.
[f] Using PhCN (∼3 equiv) in neat condition.
[g] Ratio of methine protons of *syn* and *anti* isomers (by $^1$H NMR).
[h] Ratio of isolated yields (by preparative TLC) of *syn* and *anti* isomers.

acetophenone (**11**), acetyl chloride and aceonitrile in the presence of $ZrCl_4$ (~20 mol%) also produced the corresponding β-acetamido ketone (**15**) in excellent yield (entry 3, Table 1), but $ZrOCl_2 \cdot 8H_2O$ was the catalyst of choice because of its moisture stability, recoverability and reusability without any loss of the catalytic activity.

Unlike our earlier report on BiOCl based one-pot synthesis of β-acetamido ketones[5] a mixture of *m*-nitrobenzaldehyde (**3**), acetophenone (**11**), benzoyl chloride in the presence of acetonitrile and $ZrOCl_2 \cdot 8H_2O$ failed to generate the corresponding β-acetamido ketone even after 7 days. But, the use of a different nitrile such as benzonitrile in solvent ($CH_2Cl_2$) or solvent-free conditions (entries 14 and 15) in a mixture of benzaldehyde, acetophenone and acetyl chloride in the presence of $ZrOCl_2 \cdot 8H_2O$ (~20 mol%) could lead to the expected product (**25**) in similar yields as with the acetonitrile counterpart (entry 1, Table 2) although these reactions proceeded with a much slower rate.

A mixture of chalcone (~1 equiv), acetyl chloride (~2 equiv) and $ZrOCl_2 \cdot 8H_2O$ (20 mol%) in acetonitrile failed to produce any β-acetamido ketone (**15**). Thus, this reaction also follows a similar mechanistic pathway as described in our earlier report of the one-pot synthesis based on BiOCl.[5] It is also noteworthy to mention that neither a mixture of benzaldehyde, acetophenone, acetic anhydride and $ZrOCl_2 \cdot 8H_2O$ in acetonitrile nor a mixture of benzaldehyde acylal, acetophenone and $ZrOCl_2 \cdot 8H_2O$ in acetonitrile could generate any of the corresponding β-acetamido ketone (**15**) even after stirring each mixture for a prolonged time.

### 2.2. X-ray crystallographic study of *anti*-31

The structures of the *anti*-isomers were confirmed by the X-ray crystallographic analysis of one model *anti*-isomer (*anti*-**31**). Single crystals of *anti*-**31** were grown by slow crystallization from a solution of EtOAc–hexane. The molecular view of compound *anti*-**31** is shown in Figure 1. The torsion angles C1–C7–C8–C10 −158.7(1)° and C7–C8–C10–C13 −179.0(1)° indicate that the propyl chain connecting the two nearby planar phenyl rings, C1–C6 and C13–C18, is almost straight; the dihedral angles between the two phenyl rings is 44.98(3)°. The crystal packing of *anti*-**31** reveals that the molecules are linked into two-dimensional supramolecular framework by a combination of N–H···O, C–H···O and C–H···π (arene) hydrogen bonds.

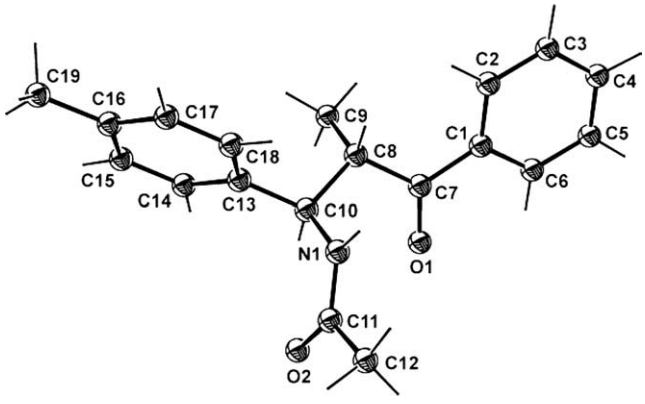



**Figure 1.** ORTEP diagram of single crystal of *anti*-**31**.

## 3. Conclusion

The efficacy of $ZrOCl_2 \cdot 8H_2O$ for the one-pot generation of β-acetamido ketones from aromatic aldehyde, enolisable ketone, acetyl chloride and acetonitrile in solvent as well as in solvent-free conditions has been established. The special feature of the present procedure lies in the easy availability, low toxicity, moisture compatibility, recoverability and reusability of the catalyst. Although, it exhibits very low to moderate diastereoselectivity but very high to excellent yields of the products make this procedure a suitable competitor with the other existing methodologies particularly for the synthesis of β-acetamido ketones based on α-unsubstituted ketones. X-ray structure analysis of one model *anti*-isomer exhibits two-dimensional supramolecular assembly by a combination of N–H···O, C–H···O and C–H···π (arene) intermolecular interactions.

## 4. Experimental

### 4.1. General

All melting points are uncorrected. IR spectra were recorded on Perkin Elmer 297 spectrophotometer. NMR spectra were recorded on Bruker DPX-300/Mercury 400/Unity 500 spectrometer using $CDCl_3$ as solvent and TMS as the internal standard. Elemental analyses were performed on a Perkin Elmer auto analyzer 2400 II.

### 4.2. General experimental procedure

(*a*) *In solution*. To a solution of aldehydes (~1 mmol) and acetophenone (~1 mmol) in dry acetonitrile (4 mL) was added $ZrOCl_2 \cdot 8H_2O$ (~20 mol%). To the resulting suspension was finally added acetyl chloride (~2 mmol) and the reaction mixture was stirred at ambient temperature. After the completion of the reaction (checked by TLC with EtOAc-pet.ether (60-80 °C) the mixture was diluted with $CH_2Cl_2$ (20 mL), washed with brine solution (1×20 mL) and the aqueous layer was then extracted with $CH_2Cl_2$ (3× 15 mL). The combined organic layer was washed with $NaHCO_3$ solution (1×15 mL) followed by water (1× 20 mL). The organic layer was dried over $Na_2SO_4$, concentrated to dryness and the crude residue was purified on silica gel column after elution with 3:2 EtOAc–pet. ether (60-80 °C). Mixture of diastereomers were separated by preparative TLC on silica plates using 1:2 EtOAc–pet. ether (60-80 °C).

(*b*) *In solvent-free conditions*. To a mixture of aromatic aldehydes (~1 mmol), acetophenone (~1 mmol) and dry acetonitrile (~3 mmol) was added $ZrOCl_2 \cdot 8H_2O$ (~20 mmol). Finally, acetyl chloride (~2 mmol) was added and the reaction mixture was stirred at ambient temperature. After completion of the reaction, it was diluted with $CH_2Cl_2$ (15 mL) and then worked-up as described in (a).

**4.2.1. *N*-(3-Oxo-1,3-diphenyl-propyl)-acetamide (15).**[4c] (Yield 240 mg, 90%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 104–105 °C, (lit.[4c] mp 102–104 °C). $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 2.03 (s, 3H), 3.40–3.48 (dd, $J$=6.0, 16.9 Hz, 1H), 3.72–3.79 (dd, $J$=5.1, 16.9 Hz, 1H), 5.53–5.60 (m, 1H), 6.68–6.70 (br d, $J$=7.5 Hz, 1H), 7.22–7.35 (m, 5H), 7.42–7.47 (m, 2H), 7.54–7.58 (m, 1H), 7.88–7.91 (m, 2H). $^{13}$C NMR (50 MHz, $CDCl_3$): $\delta$ 23.5, 43.3, 50.0, 126.6, 127.5, 128.2, 128.7, 128.8, 133.6, 136.7, 141.6, 169.6, 199.3.

**4.2.2. *N*-[1-(2-Nitro-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (16).**[4c] (Yield 265 mg, 85%). Off white crystals, (EtOAc-pet.ether, 60-80 °C) mp 191–192 °C, (lit.[4c] mp 186–188 °C). $^1$H NMR (200 MHz, $CDCl_3$): $\delta$ 2.02 (s, 3H), 3.60–3.69 (m, 2H), 5.96–5.99 (m, 1H), 7.09 (s, 1H), 7.37–7.50 (m, 3H), 7.56–7.59 (m, 2H), 7.71–7.74 (m, 1H), 7.93–7.96 (m, 3H). $^{13}$C NMR (50 MHz, $CDCl_3$): $\delta$ 23.4, 42.3, 47.5, 125.2, 128.4, 128.5, 128.9, 129.9, 133.6, 133.9, 136.3, 136.8, 148.5, 169.5, 198.7.

**4.2.3. *N*-[1-(3-Nitro-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (17).**[4c] (Yield 287 mg, 92%). White crystals, (EtOAc/pet. ether, 60:80) mp 139–140 °C, (lit.[5] mp 139–140 °C). $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 2.08 (s, 3H), 3.47–3.55 (dd, $J$=5.4, 17.5 Hz, 1H), 3.77–3.84 (dd, $J$=4.9, 17.5 Hz, 1H), 5.63–5.69 (m, 1H), 6.95–6.98 (d, $J$=7.5 Hz, 1H), 7.44–7.51 (m, 3H), 7.57–7.62 (t, $J$=7.3 Hz, 1H), 7.70–7.73 (d, $J$=7.5 Hz, 1H), 7.88–7.90 (br d, $J$=7.3 Hz, 2H), 8.07–8.10 (d, $J$=8.1 Hz, 1H), 8.22 (br s, 1H). $^{13}$C NMR (50 MHz, $CDCl_3$): $\delta$ 23.5, 43.0, 49.2, 121.4, 122.4, 128.2, 128.9, 129.6, 133.2, 134.1, 136.3, 143.7, 148.4, 169.9, 198.0.

**4.2.4. *N*-[1-(4-Nitro-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (18).**[4c] (Yield 215 mg, 69%). White amorphous solid, (EtOAc-pet.ether, 60-80 °C) mp 154 °C, (lit.[4c] mp 148–149 °C). $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 2.08 (s, 3H), 3.46–3.54 (dd, $J$=5.4, 17.6 Hz, 1H), 3.77–3.85 (dd, $J$=4.8, 17.6 Hz, 1H), 5.6–5.7 (m, 1H), 6.96–6.98 (d, $J$=7.7 Hz, 1H), 7.44–7.62 (m, 5H), 7.87–7.90 (d, $J$=7.3 Hz, 2H), 8.15–8.18 (d, $J$=8.6 Hz, 2H). $^{13}$C NMR (50 MHz, $CDCl_3$): $\delta$ 23.5, 42.7, 49.3, 123.9, 127.5, 128.2, 128.9, 134.1, 136.2, 147.1, 148.8, 174.2, 198.1.

**4.2.5. *N*-[1-(4-Chlorophenyl)-3-oxo-3-phenyl-propyl]-acetamide (19).**[4a] (Yield 274 mg, 91%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 149–150 °C, (lit.[4a] mp 150 °C). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 2.00 (s, 3H), 3.37–3.43 (dd, $J$=5.6, 17.2 Hz, 1H), 3.69–3.74 (dd, $J$=4.8, 17.1 Hz, 1H), 5.49–5.54 (m, 1H), 6.81–6.83 (d, $J$=7.6 Hz, 1H), 7.27 (s, 4H), 7.42–7.46 (t, $J$=7.8 Hz, 2H), 7.54–7.58 (t, $J$=7.2 Hz, 1H), 7.87–7.89 (d, $J$=8.4 Hz, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 23.6, 43.1, 49.5, 128.1, 128.3, 128.9, 129.0, 133.4, 133.9, 136.7, 139.8, 169.7, 198.6.

**4.2.6. *N*-[1-(4-Methoxy-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (20).**[4d] (Yield 273 mg, 92%). White crystals, (EtOAc-pet.ether 60-80 °C) mp 110–112 °C. IR (KBr): 3310, 1690, 1650, 1550, 1510, 1240, 1030, 760, 690 cm$^{-1}$. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 1.98 (s, 3H), 3.36–3.42 (dd, $J$=6.4, 17.2 Hz, 1H), 3.69–3.75 (dd, $J$=5.6, 17.2 Hz, 1H), 3.74 (s, 3H), 5.47–5.51 (dd, $J$=6, 13.2 Hz, 1H), 6.64–6.66 (d, $J$=7.2 Hz, 1H), 6.80–6.84 (m, 2H), 7.23–7.25 (d, $J$=8.8 Hz, 2H), 7.41–7.45 (t, $J$=7.8 Hz, 2H), 7.53–7.57 (t, $J$=7.6 Hz, 1H), 7.88–7.91 (d, $J$=8 Hz, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 23.6, 43.6, 49.8, 55.5, 114.2, 127.9, 128.3, 128.9, 133.3, 133.6, 136.9, 159.1, 169.6, 198.8.

**4.2.7. *N*-[1-(3-Acetoxy-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (21).**[4c] (Yield 299 mg, 92%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 114–115 °C, (lit.[4c] mp 114–115 °C). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 1.99 (s, 3H), 2.26 (s, 3H), 3.39–3.45 (dd, $J$=5.8, 17.0 Hz, 1H), 3.68–3.74 (dd, $J$=5.6, 17.2 Hz, 1H), 5.53–5.58 (m, 1H), 6.73–6.75 (br d, $J$=8.4 Hz, 1H), 6.95–6.98 (dd, $J$=2.2, 7.8 Hz, 1H), 7.08–7.09 (t, $J$=2.0 Hz, 1H), 7.17–7.19 (d, $J$=8.0 Hz, 1H), 7.27–7.31 (t, $J$=7.8 Hz, 1H), 7.42–7.46 (m, 2H), 7.53–7.57 (m, 1H), 7.88–7.89 (d, $J$=7.6 Hz, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 21.3, 23.6, 43.2, 49.7, 120.1, 120.9, 124.2, 128.3, 128.9, 129.8, 133.79, 136.8. 143.0, 151.1, 169.5, 169.7, 198.5.

**4.2.8. *N*-[3-(4-Methoxy-phenyl)-3-oxo-1-phenyl-propyl]-acetamide (22).** (Yield 273 mg, 92%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 130 °C. IR (KBr): 3260, 1680, 1640, 1600, 1570, 1255, 1170, 990, 700 cm$^{-1}$. $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 2.00 (s, 3H), 3.31–3.39 (dd, $J$=5.9, 16.6 Hz, 1H), 3.64–3.71 (dd, $J$=5.3, 16.6 Hz, 1H), 3.84 (s, 3H), 5.50–5.57 (m, 1H), 6.86 (br s, 1H), 6.89 (br d, $J$=8.8 Hz, 2H), 7.18–7.34 (m, 5H), 7.88 (br d, $J$=8.8 Hz, 2H). $^{13}$C NMR (75 MHz, $CDCl_3$): $\delta$ 23.4, 42.8, 50.0, 55.5, 113.8, 126.4, 127.3, 128.5, 129.7, 130.4, 141.1, 163.8, 169.5, 197.1. Anal. Calcd for $C_{18}H_{19}O_3N$: C, 72.71; H, 6.44; N, 4.71; Found: C, 72.65; H, 6.82; N, 4.67.

**4.2.9. *N*-[1-(4-Methyl-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (23).** (Yield 264 mg, 94%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 112 °C. IR (KBr): 3295, 1680, 1650, 1550, 1350, 1200, 810, 755, 690 cm$^{-1}$. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 2.02 (s, 3H), 2.30 (s, 3H), 3.39–3.47 (dd, $J$=6.2, 16.7 Hz, 1H), 3.71–3.78 (dd, $J$=5.1, 16.7 Hz, 1H), 5.49–5.56 (m, 1H), 6.61–6.63 (d, $J$=7.2 Hz, 1H), 7.10–7.23 (m, 4H), 7.42–7.59 (m, 3H), 7.90–7.93 (d, $J$=7.5 Hz, 2H). $^{13}$C NMR (75 MHz, $CDCl_3$): $\delta$ 20.8, 23.2, 43.1, 49.6, 126.2, 127.9, 128.5, 129.2, 133.3, 136.5, 136.9, 137.7, 169.4, 198.4. Anal. Calcd for $C_{18}H_{19}O_2N$: C, 76.84; H, 6.80; N, 4.97; Found: C, 76.94; H, 7.18; N, 4.90.

**4.2.10. *N*-[3-(4-Methoxy-phenyl)-1-(3-nitro-phenyl)-3-oxo-propyl]-acetamide (24).** (Yield 209 mg, 61%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 132 °C. IR (KBr): 3300, 3065, 2840, 1665, 1645, 1600, 1520, 1350, 1255, 1240, 1170, 1020, 990, 820, 805, 735, 660 cm$^{-1}$. $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 2.08 (s, 3H), 3.39–3.47 (dd, $J$=5.3, 17.2 Hz, 1H), 3.70–3.78 (dd, $J$=5.0, 17.2 Hz, 1H), 3.86 (s, 3H), 5.60–5.66 (m, 1H), 6.90–6.94 (d, $J$=8.7 Hz, 2H), 7.05–7.07 (d, $J$=7.9 Hz, 1H), 7.45–7.50 (t, $J$=7.9 Hz, 1H), 7.69–7.71 (d, $J$=7.6 Hz, 1H), 7.86–7.88 (d, $J$=8.6 Hz, 2H), 8.07–8.09 (d, $J$=8.1 Hz, 1H), 8.20 (s, 1H). $^{13}$C NMR (75 MHz, $CDCl_3$): $\delta$ 23.3, 42.1, 49.1, 55.4, 113.8, 121.1, 122.1, 129.1, 129.3, 130.3, 132.7, 143.4, 164.0, 169.6, 192.9, 196.4. Anal. Calcd for $C_{18}H_{18}O_5N_2$: C, 63.15; H, 5.29; N, 8.18; Found: C; 63.27, H; 5.03, N, 7.98.

**4.2.11. *N*-[1-(3-Nitro-phenyl)-oxo-3-phenyl-propyl]-benzamide (25).** (Yield 322 mg, 86%). White crystals, (EtOAc-pet.ether, 60-80 °C) mp 194–195 °C. IR (KBr): 3290, 2930, 1685, 1635, 1525, 1355, 1225, 760, 730 $cm^{-1}$. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 3.57–3.64 (dd, $J$=5.4, 17.4 Hz, 1H), 3.87–3.95 (dd, $J$=4.9, 17.4 Hz, 1H), 5.83–5.88 (m, 1H), 7.44–7.62 (m, 7H), 7.78 (br d, $J$=7.8 Hz, 1H), 7.84–7.93 (m, 5H), 8.10 (br d, $J$=8.1 Hz, 1H), 8.28 (s, 1H). $^{13}C$ NMR (75 MHz, $CDCl_3$): $\delta$ 42.5, 49.6, 121.39, 122.5, 127.1, 128.2, 128.7, 128.9, 129.6, 131.9, 132.9, 133.7, 134.1, 136.2, 143.5, 166.9, 198.6. Anal. Calcd for $C_{22}H_{18}N_2O_4$: C, 70.57; H, 4.84; N, 7.48; Found: C, 70.16; H, 4.96; N, 7.23.

**4.2.12. *N*-1-[2-Methyl-3-oxo-1-phenyl-butyl]-acetamide (26).**[4c] (Yield 188 mg, 86%).

*anti*-Isomer: (liquid). $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.08–1.10 (d, $J$=7.1 Hz, 3H), 1.98 (s, 3H), 2.07 (s, 3H), 3.00–3.07 (m, 1H), 5.34–5.39 (t, $J$=6.9 Hz, 1H), 6.29–6.32 (d, $J$=7.9 Hz, 1H), 7.20–7.31 (m, 5H).

*syn*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 126 °C, (lit.[4c] mp 126–127 °C). $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.19–1.22 (d, $J$=7.2 Hz, 3H), 1.96 (s, 3H), 2.05 (s, 3H), 3.08–3.17 (m, 1H), 5.13–5.18 (dd, $J$=8.9, 5.3 Hz, 1H), 6.91–6.94 (d, $J$=7.7 Hz, 1H), 7.2–7.3 (m, 5H).

**4.2.13. *N*-[2-Methyl-1-(4-methyl-phenyl)-3-oxo-butyl]-acetamide (27).**[4c] (Yield 189 mg, 81%).

*anti*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 162 °C. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.19–1.22 (d, $J$=7.2 Hz, 3H), 1.97 (s, 3H), 2.05 (s, 3H), 2.59 (s, 3H), 3.11–3.16 (m, 1H), 5.12–5.17 (m, 1H), 6.18–6.25 (d, $J$=12.6 Hz, 1H), 7.21–7.97 (m, 4H).

*syn*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 134 °C, (lit.[4c] mp 134 °C). $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.18–1.20 (d, $J$=7.1 Hz, 3H), 2.03 (s, 3H), 2.08 (s, 3H), 2.31 (s, 3H), 3.08–3.12 (m, 1H), 5.10–5.15 (m, 1H), 6.83–6.86 (d, $J$=7.9 Hz, 1H), 7.12 (s, 4H).

**4.2.14. *N*-(2-Methyl-3-oxo-1,3-diphenyl-propyl)-acetamide (28).**[4c] (Yield 264 mg, 94%).

*anti*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 174 °C, (lit.[4c] mp 140 °C). $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.38–1.40 (d, $J$=7.1 Hz, 3H), 2.13 (s, 3H), 4.07–4.16 (m, 1H), 5.37–5.41 (dd, $J$=3.8, 8.9 Hz, 1H), 7.16–7.54 (m, 9H), 7.75–7.77 (d, $J$=7.4 Hz, 2H).

*syn*-Isomer: white crystal. (EtOAc-pet.ether, 60-80 °C) mp 120 °C. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.21–1.24 (d, $J$=6.9 Hz, 3H), 2.00 (br s, 3H), 4.03–4.13 (m, 1H), 5.45–5.50 (t, $J$=7.7 Hz, 1H), 5.99–6.02 (d, $J$=5.9 Hz, 1H), 7.23–7.60 (m, 8H), 7.90–7.93 (d, $J$=7.3 Hz, 2H).

**4.2.15. *N*-[1-(4-Chloro-phenyl)-2-methyl-3-oxo-3-phenyl-propyl]-acetamide (29).**[4c] (Yield 262 mg, 83%).

*anti*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 162 °C. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.38–1.40 (d, $J$=7.0 Hz, 3H), 2.12 (s, 3H), 4.05–4.08 (m, 1H), 5.31–5.35 (m, 1H), 7.19 (s, 4H), 7.39–7.44 (t, $J$=7.6 Hz, 2H), 7.53–7.58 (t, $J$=7.2 Hz, 1H), 7.55 (s, 1H, exchangeable, –NH), 7.74–7.77 (d, $J$=7.5 Hz, 2H). $^1H$ NMR (300 MHz, $CDCl_3$, $D_2O$-exchange): $\delta$ 1.37–1.39 (d, $J$=7.2 Hz, 3H), 2.12 (s, 3H), 4.04–4.08 (dd, $J$=3.9, 7.1 Hz, 1H), 5.32–5.33 (d, $J$=3.7 Hz, 1H), 7.18(s, 4H), 7.38–7.40 (m, 2H), 7.52–7.57 (t, $J$=7.3 Hz, 1H), 7.74–7.76 (d, $J$=7.3 Hz, 2H).

*syn*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 164 °C. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.21 1.23 (d, $J$=6.8 Hz, 3H), 2.01 (s, 3H), 4.00–4.07 (m, 1H), 5.38–5.43 (t, $J$=7.3 Hz, 1H), 6.03–6.05 (d, $J$=6.0 Hz, 1H, exchangeable, –NH), 7.28 (s, 4H), 7.42–7.62 (m, 3H), 7.89–7.92 (d, $J$=7.6 Hz, 2H).

$^1H$ NMR (300 MHz, $CDCl_3$, $D_2O$-exchange): $\delta$ 1.19–1.21 (d, $J$=6.9 Hz, 3H), 1.98 (s, 3H), 3.99–4.04 (t, $J$=7 Hz, 1H), 5.37–5.39 (d, $J$=6.9 Hz, 1H), 7.25 (s, 4H), 7.44–7.49 (m, 2H), 7.55–7.60 (m, 1H), 7.87–7.89 (d, $J$=7.3 Hz, 2H).

**4.2.16. *N*-[2-Methyl-1-(2-nitro-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (30).**[4c] (Yield 261 mg, 80%).

*anti*-Isomer: off white crystals, (EtOAc-pet.ether, 60-80 °C) mp 142 °C, (lit.[4c] mp 122–124 °C). $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.43–1.46 (d, $J$=7.1 Hz, 3H), 2.09 (s, 3H), 4.13–4.40 (m, 1H), 5.78–5.82 (dd, $J$=3.6, 8.0 Hz, 1H), 7.28–7.95 (m, 10H).

*syn*-Isomer: oil, $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.25–1.27 (d, $J$=7.0 Hz, 3H), 2.02 (s, 3H), 4.42–4.47 (m, 1H), 5.71–5.76 (t, $J$=7.9 Hz, 1H), 6.64–6.66 (d, $J$=6.7 Hz, 1H), 7.35–7.93 (m, 9H).

**4.2.17. *N*-[2-Methyl-1-(4-methyl-phenyl)-3-oxo-3-phenyl-propyl]-acetamide (31).** (Yield 278 mg, 94%).

*anti*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 170 °C. IR (KBr): 3313, 2987, 1685, 1647, 1543, 1367, 1142, 970, 702 $cm^{-1}$. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.36–1.38 (d, $J$=7.16 Hz, 3H), 2.10 (s, 3H), 2.24 (s, 3H), 4.05–4.14 (m, 1H), 5.33–5.37 (m, 1H), 7.01–7.04 (d, $J$=7.93 Hz, 2H), 7.11–7.14 (d, $J$=8.01 Hz, 2H), 7.37–7.56 (m, 4H), 7.76–7.79 (d, $J$=7.68 Hz, 2H). $^{13}C$ NMR (75 MHz, $CDCl_3$): $\delta$ 16.5, 20.9, 23.3, 44.4, 55.5, 126.2, 128.2, 128.6, 129.1, 133.4, 136.4, 136.7, 137.8, 169.9, 204.9. Anal. Calcd for $C_{19}H_{21}O_2N$: C, 77.25; H, 7.16; N, 4.74; Found: C, 77.23; H, 7.14; N, 4.48.

*syn*-Isomer: white crystals, (EtOAc-pet.ether, 60-80 °C) mp 119 °C. IR (KBr): 3240, 3055, 1678, 1639, 1553, 1371, 1290, 972, 710 $cm^{-1}$. $^1H$ NMR (300 MHz, $CDCl_3$): $\delta$ 1.20–1.22 (d, $J$=6.96 Hz, 3H), 1.98 (s, 3H), 2.29 (s, 3H), 4.00–4.14 (m, 1H), 5.41–5.46 (t, $J$=7.64 Hz, 1H), 5.99–6.02 (d, $J$=7.68 Hz, 1H), 7.08–7.11 (d, $J$=7.83 Hz, 2H), 7.20–7.28 (d, $J$=8.0 Hz, 2H), 7.43–7.59 (m, 3H), 7.89–7.92 (d, $J$=7.38 Hz, 2H). $^{13}C$ NMR (75 MHz, $CDCl_3$): $\delta$ 14.0, 20.9, 23.1, 45.6, 54.8, 126.8, 128.1, 128.6, 129.1, 133.0, 136.3, 136.9, 137.5, 169.7, 201.9. Anal. Calcd for $C_{19}H_{21}O_2N$: C, 77.25; H, 7.16; N, 4.74; Found: C, 77.23; H, 6.94; N, 4.35.

**4.2.18. Acetic acid 5-(1-acetylamino-2-methyl-3-oxo-3-phenyl-propyl)-2-methoxy-phenyl ester (32).**[11] (Yield 273 mg, 74%).

*anti*-Isomer: white crystals, (EtOAc/pet. ether, 60:80 °C) mp 177 °C.[11] $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 1.37–1.40 (d, $J$=7.0 Hz, 3H), 2.12 (s, 3H), 2.26 (s, 3H), 3.72 (s, 3H), 4.08–4.17 (m, 1H), 5.31–5.37 (m, 1H), 6.81–6.91 (m, 3H), 7.40–7.62 (m, 4H), 7.76–7.79 (d, $J$=7.5 Hz, 2H).

*syn*-Isomer: oil, $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 1.27–1.29 (d, $J$=6.6 Hz, 3H), 2.02 (s, 3H), 2.28 (s, 3H), 3.77 (s, 3H), 4.04–4.10 (m, 1H), 5.43–5.49 (t, $J$=7.6 Hz, 1H), 6.17–6.19 (d, $J$=6.8 Hz, 1H), 6.80–6.95 (m, 3H), 7.40–7.60 (m, 3H), 7.87–7.90 (d, $J$=7.4 Hz, 2H).

**4.2.19. Acetic acid 3-(1-acetylamino-2-methyl-3-oxo-3-phenyl-propyl)-phenyl ester (33).**[4c] (Yield 105 mg, 31%).

*anti*-Isomer: white crystals, (EtOAc/pet. ether, 60:80) mp 162 °C, (lit.[4c] mp 142 °C). $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 1.32–1.35 (d, $J$=7.1 Hz, 3H), 2.08, (s, 3H), 2.41 (s, 3H), 3.96–4.04 (m, 1H), 5.50–5.54 (dd, $J$=3.6, 8.4 Hz, 1H), 7.03–7.08 (m, 2H), 7.17–7.22 (t, $J$=7.1 Hz, 2H), 7.37–7.55 (m, 4H), 7.75–7.78 (d, $J$=7.4 Hz, 2H).

*syn*-Isomer: oil, $^1$H NMR (300 MHz, $CDCl_3$): $\delta$ 1.29–1.31 (d, $J$=6.1 Hz, 3H), 1.97 (s, 3H), 2.39 (s, 3H), 4.08 (br s, 1H), 5.74–5.76 (m, 1H), 5.87 (s, 1H), 7.06–7.60 (m, 7H), 7.90–7.92 (d, $J$=7.5 Hz, 2H).

Crystallographic data of the compound *anti*-**31** has been deposited with the Cambridge Crystallographic Data Centre as supplementary publication number CCDC 285299. Copies of the data can be obtained free of charge, on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK [fax: +44 1223 336033 or e-mail: deposit@ccdc.cam.ac.uk].

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