SCOPE OF THE WORK

Although the effect of substituents on the spectral frequencies in acetophenones, benzoyl chlorides and several chalcones such as substituted styryl 1-naphthyl, 2-naphthyl, 1-pyrene, phenothiazine, 5-methyl-2-furyl, 2,5-dimethyl-3-furyl, 2,5-dimethyl-3-thienyl, 1-methyl-2-pyrrolyl, biphenyl and 9H-fluorenlyl ketones\(^1\), 26, 28, 39, 40-43, 46-48, 52, 58 has been investigated in the literature, no attention has been paid so far in this direction for 4-ethoxyphenyl, 2-benzimidazole, 1-(substituted phenyl)-3-(1-pyrenyl), 1-(substituted phenyl)-3-(1-naphthyl), 1-(substituted phenyl)-3-(2-naphthyl) chalcones. Further the IR, \(^1\)H and \(^{13}\)C NMR spectra of these chalcones have been recorded and interpreted. But similar study in the 4-ethoxyphenyl, 2-benzimidazole, 1-(substituted phenyl)-3-(1-pyrenyl), 1-(substituted phenyl)-3-(1-naphthyl), 1-(substituted phenyl)-3-(2-naphthyl) chalcones were not known. It has been synthesized above chalcones and recorded their infrared, \(^1\)H and \(^{13}\)C NMR spectra with a view to seek structure-parameter correlation. For the above chalcones, the effects of different substituents on antimicrobial activity related with the structure have been analyzed.
2. EXPERIMENTAL

The following compounds namely, 4-ethoxyacetophenone, benzimidazole-2-methyl ketone, various substituted acetophenones, substituted benaldehydes, 1-pyrenaldehyde, 1-napthaldehyde and 2-napthaldehydes have been procured from Sigma Aldrich Grade.

2.1. Preparation of 4-ethoxy phenyl chalcones (Series A)

This series of chalcones were prepared by SiO$_2$-H$_3$PO$_4$ catalyzed solvent-free Aldol condensation method.

2.1.1. Styryl 4-ethoxyphenyl ketone$^{53}$

Equimolar quantities of 4-ethoxy acetophenone (0.648 g, 2 mmol), benzaldehyde (0.212 g, 2 mmol) and the catalyst SiO$_2$-H$_3$PO$_4$ (0.4 g) were taken in a 50 mL corning glass tube and tightly capped. The reaction mixture was subjected to microwave irradiation for 8-10 minutes in a microwave oven (Scheme 1) (LG Grill, Intellowave, Microwave Oven, 160-800W) and then cooled to room temperature. Added 10 ml of dichloromethane, the organic layer has been separated which on evaporation yields the solid product. The solid, on recrystallization with benzene-hexane mixture gives glittering pale yellow solid. Then this was recrystallized using ethanol to obtain pale yellow glittering solid melting at 71-72°C. M.F. C$_{17}$H$_{16}$O$_2$; M.W. 252; Mass (m/z): 252[M$^+$].

2.1.2. 3-Bromostyryl 4-ethoxyphenyl ketone$^{53}$

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 3-bromobenzaldehyde (0.740 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 88-89°C. M.F. C$_{17}$H$_{15}$BrO$_2$; M.W. 331; Mass (m/z): 331[M$^+$].

2.1.3. 4-Bromostyryl 4-ethoxyphenyl ketone$^{53}$

This compound was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-bromobenzaldehyde (0.740 g, 2 mmol) and SiO$_2$-
H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 132-133°C. M.F. C$_{17}$H$_{15}$BrO$_2$; M.W. 331; Mass (m/z): 331[M$^+$].

2.1.4. 3-Chlorostyryl 4-ethoxylphenyl ketone$^{53}$

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 3-chlorobenzaldehyde (0.280 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 89-90°C. M.F. C$_{17}$H$_{15}$BrO$_2$; M.W. 286; Mass (m/z): 286[M$^+$], 288[M$^{2+}$].

2.1.5. 4-Chlorostyryl 4-ethoxylphenyl ketone$^{53}$

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-chlorobenzaldehyde (0.280 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 126-127°C. M.F. C$_{17}$H$_{15}$BrO$_2$; M.W. 286; Mass (m/z): 286[M$^+$], 288[M$^{2+}$].

2.1.6. 4-Fluorostyryl 4-ethoxylphenyl ketone$^{53}$

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-fluorobenzaldehyde (0.248g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 101-102°C. M.F. C$_{17}$H$_{15}$FO$_2$; M.W. 270; Mass (m/z): 270[M$^+$], 272[M$^{2+}$].

2.1.7. 2-Methoxystyryl 4-ethoxylphenyl ketone$^{53}$

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 2-methoxybenzaldehyde (0.272 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 98-99°C. M.F. C$_{19}$H$_{20}$O$_2$; M.W. 282; Mass (m/z): 282[M$^+$].
2.1.8. 4-Methoxystyryl 4-ethoxylphenyl ketone

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-methoxybenzaldehyde (0.272 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 91-92°C. M.F. C$_{19}$H$_{20}$O$_2$; M.W. 282; Mass (m/z): 282[M$^+$].

2.1.9. 4-Methylstyril 4-ethoxylphenyl ketone

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-methylbenzaldehyde (0.240 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 97-98°C. M.F. C$_{19}$H$_{20}$O; M.W. 266; Mass (m/z): 266[M$^+$].

2.1.10. 3-Nitrostyryl 4-ethoxylphenyl ketone

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-methylbenzaldehyde (0.302 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 91-92°C. M.F. C$_{17}$H$_{18}$NO$_3$; M.W. 297; Mass (m/z): 297[M$^+$].

2.1.11. 4-Nitrostyryl 4-ethoxylphenyl ketone

This chalcone was prepared by following the above procedure, 4-ethoxy acetophenone (0.648 g, 2 mmol), 4-methylbenzaldehyde (0.302 g, 2 mmol) and SiO$_2$-H$_3$PO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 118-119°C. M.F. C$_{17}$H$_{18}$NO$_3$; M.W. 297; Mass (m/z): 297[M$^+$].

2.2. Preparation of 2-Benzimidazole chalcones (Series B)

This series of chalcones were prepared by fly-ash:H$_2$SO$_4$ catalyzed solvent-free Aldol condensation method.
2.2.1. Styryl 2-benzimidazole ketone

An appropriate equimolar quantities of Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), benzaldehyde (0.212 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g) have been taken in borosil tube and tightly capped. The mixture has been exposed to microwave for 8-10 minutes in a microwave oven (LG Grill, Intellowave, Microwave Oven, 160-800W) and then cooled to room temperature. The organic layer has been separated with dichloromethane which on evaporation yields the solid product. The solid, on recrystallization with benzene-hexane mixture gives glittering solid. The insoluble catalyst has been recycled by washing the solid reagent remained on the filter by ethyl acetate (8 mL) followed by drying in an oven at 100°C for 1h. Then the crude chalcone was recrystallized using ethanol to obtain pale yellow glittering solid melting at 216-217 (215-217 °C)$_{136}$; M.F. C$_{16}$H$_{12}$N$_2$O; M.W. 248; Mass (m/z): 248[M$^+$]

2.2.2. 2-Bromostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 2-bromobenzaldehyde (0.740 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 258-261(257-260 °C)$_{136}$; M.F. C$_{16}$H$_{11}$Br N$_2$O$_2$; M.W. 327; Mass (m/z): 327[M$^+$], 329 [M$^{2+}$]

2.2.3. 3-Bromostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 3-bromobenzaldehyde (0.740 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 249-251(248-250 °C)$_{136}$; M.F. C$_{16}$H$_{11}$Br N$_2$O$_2$; M.W. 327; Mass (m/z): 327[M$^+$], 329 [M$^{2+}$].
2.2.4. 4-Bromostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 4-bromobenzaldehyde (0.740 g, 2 mmol) and fly-
ash:H₂SO₄ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 224-226(223-226 °C)¹³⁶; M.F. C₁₆H₁₁Br N₂O₂; M.W. 327;
Mass (m/z): 327[M⁺], 329 [M²⁺].

2.2.5. 2-Chlorostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 2-chlorobenzaldehyde (0.280 g, 2 mmol) and fly-
ash:H₂SO₄ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 190-192(189-190 °C)¹³⁶; M.F. C₁₆H₁₁ClN₂O₂; M.W. 287;
Mass (m/z): 287[M⁺], 289 [M²⁺].

2.2.6. 3-Chlorostyryl 2-benzimidazole ketone

It was prepared by following the above procedure, Benzimidazole-2-methyl ketone
(0.332 g, 2 mmol), 3-chlorobenzaldehyde (0.280 g, 2 mmol) and fly-ash:H₂SO₄ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid
melting at 238-239(237-238 °C)¹³⁶; M.F. C₁₆H₁₁ClN₂O₂; M.W. 287; Mass (m/z): 287[M⁺],
289 [M²⁺].

2.2.7. 4-Chlorostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 4-chlorobenzaldehyde (0.280 g, 2 mmol) and fly-
ash:H₂SO₄ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 225-226(224-225 °C)¹³⁶; M.F. C₁₆H₁₁ClN₂O₂; M.W. 287;
Mass (m/z): 287[M⁺], 289 [M²⁺].
2.2.8. 4-Dimethylaminostyryl 2-benzimidazole ketone

This compound was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 4-dimethylaminobenzaldehyde (0.298 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 261-262(260-261\(^\circ\)C)\textsuperscript{136}; M.F. C$_{18}$H$_{17}$N$_3$O; M.W. 291; Mass (m/z): 291[M$^+$].

2.2.9. 4-Fluorostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 4-fluorobenzaldehyde (0.248 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 177-178(178-177\(^\circ\)C)\textsuperscript{136}; M.F. C$_{16}$H$_{11}$FN$_2$O$_2$; M.W. 266; Mass (m/z): 266[M$^+$], 268[M$^{2+}$].

2.2.10. 2-Hydroxystyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 2-hydroxybenzaldehyde (0.244 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 139-140(138-139\(^\circ\)C)\textsuperscript{136}; M.F. C$_{16}$H$_{12}$N$_2$O$_2$; M.W. 274; Mass (m/z): 274[M$^+$].

2.2.11. 3-Hydroxy styryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 3-hydroxybenzaldehyde (0.244 g, 2 mmol) and fly-ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 248-249(247-248\(^\circ\)C)\textsuperscript{136}; M.F. C$_{16}$H$_{12}$N$_2$O$_2$; M.W. 274; Mass (m/z): 274[M$^+$].
2.2.12. 4-Hydroxystyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 4-hydroxybenzaldehyde (0.244 g, 2 mmol) and fly-
ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 114-115(113-114°C)$^{136}$; M.F. C$_{16}$H$_{12}$N$_2$O$_2$; M.W. 274;
Mass (m/z): 274[M$^+$].

2.2.13. 2-Methoxystyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 2-methoxybenzaldehyde (0.272 g, 2 mmol) and fly-
ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 197-198(196-197°C)$^{136}$; M.F. C$_{17}$H$_{14}$N$_2$O$_2$; M.W. 291;
Mass (m/z): 291[M$^+$].

2.2.14. 3-Methoxystyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 3-methoxybenzaldehyde (0.272 g, 2 mmol) and fly-
ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 165-166(164-165°C)$^{136}$; M.F. C$_{17}$H$_{14}$N$_2$O$_2$; M.W. 291;
Mass (m/z): 291[M$^+$].

2.2.15. 4-Methoxystyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-
methyl ketone (0.332 g, 2 mmol), 3-methoxybenzaldehyde (0.272 g, 2 mmol) and fly-
ash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 191-192(190-191°C)$^{136}$; M.F. C$_{17}$H$_{14}$N$_2$O$_2$; M.W. 291;
Mass (m/z): 291[M$^+$].
2.2.16. 2-Methylstyril 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 2-methylbenzaldehyde (0.240 g, 2 mmol) and flyash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 198-199$^\circ$C; M.F. C$_{17}$H$_{14}$N$_2$O; M.W. 262; Mass (m/z): 262[M$^+$].

2.2.17. 3-Methylstyril 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 3-methylbenzaldehyde (0.240 g, 2 mmol) and flyash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 205-206(204-205$^\circ$C); M.F. C$_{17}$H$_{14}$N$_2$O; M.W. 262; Mass (m/z): 262[M$^+$].

2.2.18. 4-Methylstyril 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 4-methylbenzaldehyde (0.240 g, 2 mmol) and flyash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 211-212(210-211$^\circ$C); M.F. C$_{17}$H$_{14}$N$_2$O; M.W. 262; Mass (m/z): 262[M$^+$].

2.2.19. 2-Nitrostyril 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 2-nitrobenzaldehyde (0.302 g, 2 mmol) and flyash:H$_2$SO$_4$ (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 220-221(219-220$^\circ$C); M.F. C$_{16}$H$_{11}$N$_3$O$_3$; M.W. 293; Mass (m/z): 262[M$^+$].
2.2.20. 3-Nitrostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 3-nitrobenzaldehyde (0.302 g, 2 mmol) and fly-ash:H\textsubscript{2}SO\textsubscript{4} (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 262-263(261-262°C)\textsuperscript{136}, M.F. C\textsubscript{16}H\textsubscript{11}N\textsubscript{3}O\textsubscript{3}; M.W. 293; Mass (m/z): 262[M\textsuperscript{+}].

2.2.21. 4-Nitrostyryl 2-benzimidazole ketone

This chalcone was prepared by following the above procedure, Benzimidazole-2-methyl ketone (0.332 g, 2 mmol), 4-nitrobenzaldehyde (0.302 g, 2 mmol) and fly-ash:H\textsubscript{2}SO\textsubscript{4} (0.4 g). Finally the product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 232-233(231-232°C)\textsuperscript{136}, M.F. C\textsubscript{16}H\textsubscript{11}N\textsubscript{3}O\textsubscript{3}; M.W. 293; Mass (m/z): 262[M\textsuperscript{+}].

2.3. Preparation of 3-(1-pyrenyl)-1-(substituted phenyl)-2-propen-1-ones (Series C)

This series of chalcones were prepared by green chemistry NaOH: H\textsubscript{2}O ground method.

2.3.1. 3-(1-Pyrenyl)-1-phenyl-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46g, 2 mmol), acetophenone (0.240 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured in to ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 177-178(176-177°C)\textsuperscript{137}; M.F. C\textsubscript{25}H\textsubscript{16}O; M.W. 332; Mass (m/z): 332[M\textsuperscript{+}].

2.3.2. 3-(1-Pyrenyl)-1-(2-chlorophenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 2-chloroacetophenone (0.310 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were
ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 190-191°C; M.F. C_{25}H_{15}ClO; M.W. 366; Mass (m/z): 366[M⁺].

2.3.3. 3-(1-pyrenyl)-1-(4-chlorophenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 4-chloroacetophenone (0.310 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 141-143°C; M.F. C_{25}H_{15}ClO; M.W. 366; Mass (m/z): 366[M⁺].

2.3.4. 3-(1-Pyrenyl)-1-(4-ethoxyphenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 4-ethoxyacetophenone (0.328 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 165-166°C; M.F. C_{27}H_{20}O_{2}; M.W. 376; Mass (m/z): 376[M⁺].

2.3.5. 3-(1-Pyrenyl)-1-(2-hydroxyphenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 2-hydroxyacetophenone (0.272 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 168-169°C; M.F. C_{25}H_{16}O_{2}; M.W. 348; Mass (m/z): 348[M⁺].
2.3.6. 3-(1-Pyrenyl)-1-(3-methoxyphenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 3-methoxyacetophenone (0.300 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 145-146°C; M. F. C\textsubscript{26}H\textsubscript{18}O\textsubscript{2}; M.W. 362; Mass (m/z): 362[M\textsuperscript{+}].

2.3.7. 3-(1-Pyrenyl)-1-(4-methoxyphenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 4-methoxyacetophenone (0.300 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 163-164°C; M. F. C\textsubscript{26}H\textsubscript{18}O\textsubscript{2}; M.W. 362; Mass (m/z): 362[M\textsuperscript{+}].

2.3.8. 3-(1-Pyrenyl)-1-(3-nitrophenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46 g, 2 mmol), 3-nitroacetophenone (0.300 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured into ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 174-175°C; M. F. C\textsubscript{25}H\textsubscript{15}NO\textsubscript{3}; M.W. 377; Mass (m/z): 377[M\textsuperscript{+}].

2.3.9. 3-(1-Pyrenyl)-1-(4-nitrophenyl)-2-propen-1-one

An appropriate equimolar quantities of pyrene-1-aldehyde (0.46g, 2 mmol), 4-nitroacetophenone (0.300 g, 2 mmol), NaOH (0.5 g) and 10 mL distilled water were ground with Pestle and Mortar for 10 minutes. The ground reaction mixture was poured in
to ice-cold water, the obtained solid was filtered and recrystallized with ethanol afforded a pale yellow glittering solid melting at 137-138°C; M. F. C_{25}H_{15}NO_{3}; M.W. 377; Mass (m/z): 377[M^+].

2.4. Preparation of 3-(1-naphthyl)-1-(substituted phenyl)-2-propen-1-ones (Series D)

This series of chalcones were prepared by hydroxyapatite catalyzed solvent-free Aldol condensation method.

2.4.1. 3-(1-naphthyl)-1-phenyl-2-propen-1-one

Appropriate equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), acetophenone (0.240 g, 2 mmol) and hydroxyapatite (0.4 g) were taken in a 50 mL corning glass tube and tightly capped. The reaction mixture was subjected to microwave irradiation for 4-6 minutes in a microwave oven (LG Grill, Intellowave, Microwave Oven, 160-800W) and then cooled to room temperature. Added 10 ml of dichloromethane, the organic layer has been separated which on evaporation yields the solid product. The solid was recrystallized using ethanol to obtain pale yellow glittering solid melting at 96-97°C. M.F. C_{19}H_{14}O; M.W. 258; Mass (m/z): 258[M^+].

2.4.2. 3-(1-Naphthyl)-1-(4-bromo phenyl)-2-propen-1-one

This chalcones were synthesized by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 4-bromoacetophenone (0.398 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 118-119°C. M.F. C_{19}H_{13}BrO; M.W. 337; Mass (m/z): 337[M^+].

2.4.3. 3-(1-Naphthyl)-1-(3-chloro phenyl)-2-propen-1-one

This chalcone was synthesized by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 3-chloroacetophenone (0.310 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 104-104°C. M.F. C\textsubscript{19}H\textsubscript{13}ClO; M.W. 219; Mass (m/z): 219\textsuperscript{[M]+}.

\textbf{2.4.4. 3-(1-Naphthyl)-1-(4-chloro phenyl)-2-propen-1-one}

This chalcone was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 4-chlorobromoacetophenone (0.310 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 112-113\textdegree C\textsuperscript{139}. M.F. C\textsubscript{19}H\textsubscript{13}ClO; M.W. 219; Mass (m/z): 219\textsuperscript{[M]+}.

\textbf{2.4.5. 3-(1-Naphthyl)-1-(2-hydroxy phenyl)-2-propen-1-one}

This chalcone was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 2-hydroxyacetophenone (0.272 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 97-98\textdegree C. M.F. C\textsubscript{19}H\textsubscript{14}O\textsubscript{2}; M.W. 274; Mass (m/z): 274\textsuperscript{[M]+}.

\textbf{2.4.6. 3-(1-Naphthyl)-1-(3-hydroxy phenyl)-2-propen-1-one}

This chalcone was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 3-hydroxyacetophenone (0.272 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 118-119\textdegree C. M.F. C\textsubscript{19}H\textsubscript{14}O\textsubscript{2}; M.W. 274; Mass (m/z): 274\textsuperscript{[M]+}.

\textbf{2.4.7. 3-(1-Naphthyl)-1-(4-hydroxy phenyl)-2-propen-1-one}

This chalcone was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 4-hydroxyacetophenone (0.272 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale
yellow glittering solid melting at 125-126°C. M.F. C_{19}H_{14}O_2; M.W. 274; Mass (m/z): 274[M^+].

2.4.8. 3-(1-Naphthyl)-1-(4-methyl phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 4-methylacetophenone (0.272 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 132-133°C (132-133°C). M.F. C_{20}H_{16}O_2; M.W. 272; Mass (m/z): 272[M^+].

2.4.9. 3-(1-Naphthyl)-1-(3-nitro phenyl)-2-propen-1-one

This chalcone was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 3-nitroacetophenone (0.330 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 121-122°C. M.F. C_{19}H_{13}NO_3; M.W. 303; Mass (m/z): 303[M^+].

2.4.10. 3-(1-Naphthyl)-1-(4-nitro phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 1-naphthaldehyde (0.46 g, 2 mmol), 4-nitroacetophenone (0.330 g, 2 mmol) and hydroxyapatite (0.4 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 117-118°C (117-119°C). M.F. C_{19}H_{13}NO_3; M.W. 303; Mass (m/z): 303[M^+].

2.5. Preparation of 3-(2-naphthyl)-1-(substituted phenyl)-2-propen-1-ones (Series E)

This series of chalcones were prepared by sulphated titania TiO_2SO_4^2- catalyzed solvent-free Aldol condensation method.
2.5.1. 3-(2-Naphthyl)-1-phenyl-2-propen-1-one

Appropriate equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), acetophenone (0.240 g, 2 mmol) and sulphated titania (0.2 g) were taken in a 50 mL corning glass tube and tightly capped. The reaction mixture was subjected to microwave irradiation for 4-6 minutes in a microwave oven (LG Grill, Intellowave, Microwave Oven, 160-800W) and then cooled to room temperature. Added 10 ml of dichloromethane, the organic layer has been separated which on evaporation yields the solid product. The insoluble catalyst was washed with 10 mL of ethyl acetate and dried at 110°C for 1 h then it is recycled for further reaction runs. The obtained solid was recrystallized using ethanol to obtain pale yellow glittering solid melting at 111-112°C. M.F. C_{19}H_{14}O; M.W. 258; Mass (m/z): 258[M+].

2.5.2. 3-(2-Naphthyl)-1-(4-bromo phenyl)-2-propen-1-one

This chalcone was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-bromoacetophenone (0.398 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 98-99°C. M.F. C_{19}H_{13}BrO; M.W. 337; Mass (m/z): 337[M+].

2.5.3. 3-(2-Naphthyl)-1-(4-chloro phenyl)-2-propen-1-one

This chalcone was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-bromoacetophenone (0.310 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 116-117°C. M.F. C_{19}H_{13}ClO; M.W. 219; Mass (m/z): 219[M+].
2.5.4. 3-(2-Naphthyl)-1-(4-fluoro phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-fluoroacetophenone (0.274 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 105-106(105-106°C)\(^\text{139}\). M.F. C\(_{19}\)H\(_{13}\)FO; M.W. 276; Mass (m/z): 276[M\(^+\)].

2.5.5. 3-(2-Naphthyl)-1-(4-hydroxy phenyl)-2-propen-1-one

This chalcones were synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46g, 2 mmol), 4-hydroxyacetophenone (0.272 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 88-89°C. M.F. C\(_{19}\)H\(_{14}\)O\(_2\); M.W. 274; Mass (m/z): 274[M\(^+\)].

2.5.6. 3-(2-Naphthyl)-1-(4-iodo phenyl)-2-propen-1-one

This chalcones were synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-hydroxyacetophenone (0.464 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 118-119(115-116°C)\(^\text{140}\). M.F. C\(_{19}\)H\(_{13}\)IO; M.W. 384; Mass (m/z): 384[M\(^+\)].

2.5.7. 3-(2-Naphthyl)-1-(4-methoxy phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-methoxyacetophenone (0.300 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 122-123°C. M.F. C\(_{20}\)H\(_{16}\)O\(_2\); M.W. 288; Mass (m/z): 288[M\(^+\)].
2.5.8. 3-(2-Naphthyl)-1-(4-methly phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-methylacetophenone (0.288 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 112-113°C. M.F. C_{20}H_{16}O; M.W. 272; Mass (m/z): 272[M^+].

2.5.9. 3-(2-Naphthyl)-1-(2-nitro phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 2-nitroacetophenone (0.330 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 132-133°C. M.F. C_{19}H_{13}NO_3; M.W. 303; Mass (m/z): 303[M^+].

2.5.10. 3-(2-Naphthyl)-1-(3-nitro phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 3-nitroacetophenone (0.330 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 127-128°C. M.F. C_{19}H_{13}NO_3; M.W. 303; Mass (m/z): 303[M^+].

2.5.11. 3-(2-Naphthyl)-1-(4-nitro phenyl)-2-propen-1-one

This chalcones was synthesised by using above procedure with equimolar quantities of 2-naphthaldehyde (0.46 g, 2 mmol), 4-nitroacetophenone (0.330 g, 2 mmol) and sulphated titania (0.2 g). The crude product was recrystallized using ethanol to obtain pale yellow glittering solid melting at 119-110(118-120°C)^{138}. M.F. C_{19}H_{13}NO_3; M.W. 303; Mass (m/z): 303[M^+].
2.6. Spectra

2.6.1 IR Spectra

The IR spectra of all compounds were recorded in Avatar-300, Thermo Nikolet Fourier Transform infrared spectrophotometer using KBr discs with the frequency range of 4000-400cm\(^{-1}\).

2.6.2. NMR Spectra

The NMR spectra of compounds were recorded in (i) INSTRUM AV300 type spectrometer, operating at 500MHz was used for \(^1\)H spectra and 125.46MHz for \(^{13}\)C spectra in DMSO solvent using TMS as internal standard. (ii) The Bruker AV400 type NMR spectrometer operating at 400MHz was used for \(^1\)H spectra and 100MHz for \(^{13}\)C spectra in DMSO solvent using CDCl\(_3\) as internal standard.

2.6.3. Mass spectra

Mass spectra of all compounds were recorded on a SIMADZU GC-MS2010 Spectrometer and VARIAN-SATURN 2200 GC-MS spectrometer using Electron Impact (EI) techniques.

2.7. Antimicrobial activity measurements

2.7.1. Materials and methods

The chemicals namely nutrient broth, Mueller Hinton agar, potato dextrose agar, Tween-80 solution and other materials required have been procured from Himedia, Mumbai.

2.7.2. Collection of Microorganisms

_Bacillus subtilis, Escherichia coli, Klebsila pneumonia, Micrococcus luteus, Pseudomonas aerogenosa, Staphylococcus areus, Aspergillus niger, Mucor species and Trichoderma viride_ were procured from the Research Department of Microbiology,
Sengunthar Arts and Science College, Thiruchengode, Namakkal District, Tamilnadu. The stock cultures have been stored in the refrigerator for further studies.

2.7.3. **Innoculum preparation**

The nutrient broth was procured from Himedia, Mumbai. The nutrient broth was prepared by weighing 1.3 g of the broth and dissolved it in 100 ml of sterile distilled water. The flask was swirled gently while adding the nutrient broth and the pH of the medium was adjusted to 7.0. The Erleumayer flask was plugged with non-adsorbent cotton and sterilized in an autoclave at 121°C and 15 lbs/inc² pressure for 15 minutes. After cooling inside a laminar flow, a loopful of fresh bacterial sample was inoculated and incubated in an orbital shaker at 37°C for 24 h. Then the cultures were diluted 1:50 with sterile physiological saline and 0.5 ml of the inoculum was used for the preparation of the spread plate. The same procedure has been adopted for all test bacterial samples.

2.7.4. **Preparation of agar slants**

Nutrients agar medium was prepared and sterilized in an autoclave at 121°C and 15 lbs/inc² pressure for 15 minutes. After sterilization the medium was dispensed into the test tubes. The test tubes were kept in the slanting position on a support. After complete solidification of the medium, streaking of the microorganism was done in the slant area using sterile inoculation loop. After the streaking the test tubes were incubated at 37°C for 24 h. After good growth, the slants have been stored in a deep freezer (2°C) for further studies.

2.7.5. **Preparation of Mueller Hinton agar plates**

The Mueller Hinton agar of weight 38 g was dissolved in 1000 ml of sterile distilled water. The pH of the medium was adjusted to 7.0. The flask was plugged with cotton and sterilized at 121°C and 15 lbs/inc² pressure for 15 min. After sterilization, the
medium was cooled to 45-47° C, poured 15 ml of it in each sterile Petri-plates and allowed to solidify.

2.7.6. Preparation of test compound

The newly synthesized Chalcone compounds of weight 15 mg of each was dissolved in 1 ml of DMSO solvent. Using 100 μml solution, the discs were impregnated and placed on the Mueller Hinton solidified Agar medium to find out the antimicrobial activity of the compounds on each organism.

By adapting the above procedure, the antimicrobial activity of the five series of chalcones has been studied on six microorganisms and the results have been discussed.

2.7.7. Antibacterial sensitivity assay

Antibacterial sensitivity assay was performed using Kirby-Bauer\textsuperscript{141} disc diffusion technique. In each Petri plate about 0.5 ml of the test bacterial sample was spread uniformly over the solidified Mueller Hinton agar using sterile glass spreader. Then the discs with 5mm diameter made up of WhatmanNo.1 filter paper, impregnated with the solution of the compound were placed on the medium using sterile forceps. The plates were incubated for 24 hours at 37° C by keeping the plates upside down to prevent the collection of water droplets over the medium. After 24 h, the plates were visually examined and the diameter values of the zone of inhibition were measured. Triplicate results were recorded by repeating the same procedure.

2.7.8 Preparation of the Potato dextrose agar medium

PDA agar medium was prepared in a conical flask by dissolving 3.9 g of the agar in 100 ml distilled water. It was sterilized in the autoclave for 15min. at 121° C and 15 lbs/inch\textsuperscript{2} pressure. Then the medium was allowed for solidification for an hour. After
that the fungal species was inoculated in the medium and kept for 5 to 7 days at room
temperature.

2.7.9. Preparation of the fungal inoculam

About 20 to 25 ml of sterile water (after cooling) is mixed with the medium. The
water over the medium is swirled and decanted with the fungal species. Tween-80(1 to 2
ml) may be added with this solution for uniform growth.

2.7.10 Antifungal sensitivity assay

Antifungal sensitivity assay was performed using Kirby-Bauer\textsuperscript{141} disc diffusion
technique. PDA medium was prepared and sterilized as above. It was poured (ear bearing
heating condition) in the Petri-plate which was already filled with 1 mL of the fungal
species. The plate was rotated clockwise and counter clock-wise for uniform spreading of
the species. The discs were impregnated with the test solution. The test solution was
prepared by dissolving 15 mg of the chalcone in 1mL of DMSO solvent. The medium was
allowed to solidify and kept for 24 h. Then the plates were visually examined and the
diameter values of zone of inhibition were measured. Triplicate results were recorded by
repeating the same procedure.