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Fly-ash:H_2O Assisted Aldol Condensation: Effect of Solvents on the Synthesis of Some Aryl (*E*) 2-propen-1-ones

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Some series of aryl (*E*) 2-propen-1-ones have been synthesized by Fly-ash:H_2O catalyzed aldol condensation of aryl methyl ketones and substituted benzaldehydes at reflux condition in atmospheric pressure. The yields of chalcones are higher than 60%. These chalcones were characterized by their physical constants and spectral data, for known compounds their purities were checked with the physical constants and spectral data earlier published in the literature. The effects of solvents on the product have been studied.

Keywords: Aryl (*E*) 2-propen-1-ones, Fly-ash:H_2O, IR and NMR spectra, Solvent effects

1. INTRODUCTION

Homogeneous and heterogeneous catalytic Aldol and Crossed-Aldol condensation is an important powerful tool for the formation of carbon-carbon bond in synthesis of many kinds of carbonyl compounds [1, 2]. Homogeneous thermal aldol reaction is found to be very slow and took longer time with low yields due to the complication of separation of catalyst and product.

However, the heterogeneous thermal reactions are good because the rate of the reaction is fast, the reaction time is short and yield is high with easy isolation of products from the catalyst. Various catalysts have been used for synthesizing chalcones such as silica-sulphuric acid [3, 4], anhydrous zinc chloride [5], Clay [6], ground chemistry catalysts-grinding the reactants with sodium hydroxide [7], aqueous alkali in lower temperature[8], solid sulphonic acid from aqueous alkali in lower temperature [9], solid sulphonic acid from bamboo [9], barium hydroxide [10] anhydrous sodium bicarbonate [11], microwave assisted synthesis [12], and sulfated titania [13].

Chalcones possess various multipronged activities such as antimicrobial [14], antidepressants [15], antiplosmodial [16], anti-aids [17] and insect antifeedant activities [18-20]. In the present investigation, the author wishes to report a new catalyst Fly-ash:H_2O for synthesizing chalcones by Crossed-Aldol condensation reaction. The yields of chalcones are more than 80%.

The synthesized chalcones were characterized by their physical constants, Mass, IR and NMR spectral data as they were unknown compounds so far. The purities of the known synthesized chalcones have been checked by their physical constants and their spectral data earlier published in literature. The effects of solvents on these reactions have been studied.

2. EXPERIMENTAL

Material and methods

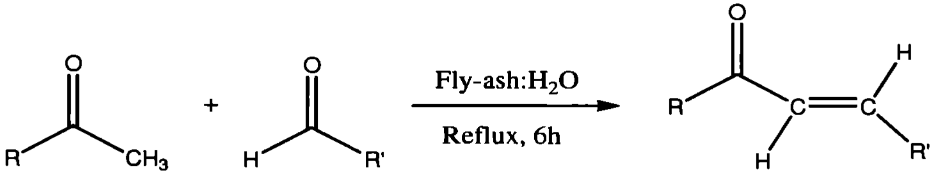
All chemicals used were purchased from Sigma-Aldrich Corporation (St. Louis, MO, USA) and E-Merck chemical company (Merck Limited, Mumbai, India). Melting points of all pyrazolines have been determined in open glass capillaries on Mettler FP51 melting point apparatus (Mettler-Toledo India Private Limited, Mumbai, India) and are uncorrected.

Infrared spectra (KBr, 4000-400 cm^{-1}) have been recorded by AVATAR-300 Fourier transform spectrophotometer (Thermo Nicolet, USA). The NMR spectra of all imines were recorded by Bruker AV400 spectrometer (BRUKER AXS GMBH, Karlsruhe, Germany), operated 400 MHz frequency for recording ^1H and 100 MHz for ^{13}C NMR spectra in $CDCl_3$ solvent using TMS as internal standard.

Electron impact (70 eV) and chemical ionization mode FAB+ mass spectra have been recorded by VARIAN-SATURN 2200 GC-MS spectrometer (Varian 92 Medical Systems, Palo Alto, CA, USA).

Synthesis of chalcones

An equi-molar quantities of aryl methyl ketones (2 mmol) and substituted benzaldehydes (2 mmol), Fly-ash (0.5 g) with 20 cm^3 of water [20, 21] (Scheme 1) were refluxed for 4 h. and the completion of the reaction was monitored by thin layer chromatography. After completion of the reaction, the solid product was separated by filtration. Afterwards, the product was purified by column chromatography using dichloromethane and ethyl acetate mixture as eluent.



R = C_6H_5, 4-NO_2-C_6H_4, 4-X-1-naphthyl, 6-X-2-naphthyl, 9H-Fluorene
5-X-2-thienyl, 1-Pyrene
R' = Substituted benzaldehydes

Scheme 1. Synthesis of aryl chalcones by fly-ash:H_2O catalyzed aldol condensation.

3. RESULTS AND DISCUSSION

The waste and harmful air pollutant fly-ash containing many chemical species such as [12, 21-24] SiO_2, Fe_2O_3, Al_2O_3, CaO, MgO and insoluble residues. This pollutant is used as useful catalyst for organic synthesis. Due to the presence of the above said species, they assisted the aldol condensation in aqueous phase reaction between aryl methyl ketone and aldehydes.

Therefore, the author has attempted to synthesize aryl chalcones by Crossed-Aldol condensation between aryl methyl ketones and benzaldehydes in the presence of fly-ash catalyst under refluxed condition in room temperature and pressure (Scheme 1). Quantitatively, 2 mmol of aryl ketones such as 4-X-phenyl methyl, 4-X-1-naphthyl methyl, 6-X-2-naphthyl methyl, 9*H*- Fluorene-2-methyl and 5-X-2-thienyl (X = electron donating or electron withdrawing substituents), 2 mmol of substituted benzaldehydes, 0.5 g of fly-ash and 20 cm^3 of water were refluxed for 4h and produced the respective chalcones.

The proposed general mechanism of this reaction is shown in Fig. 1. It follows the removal of water and proton giving the product. The yields of chalcones are more than 60%. Analytical, physical constants, yield and mass fragments (m/z) are presented in Table 1.

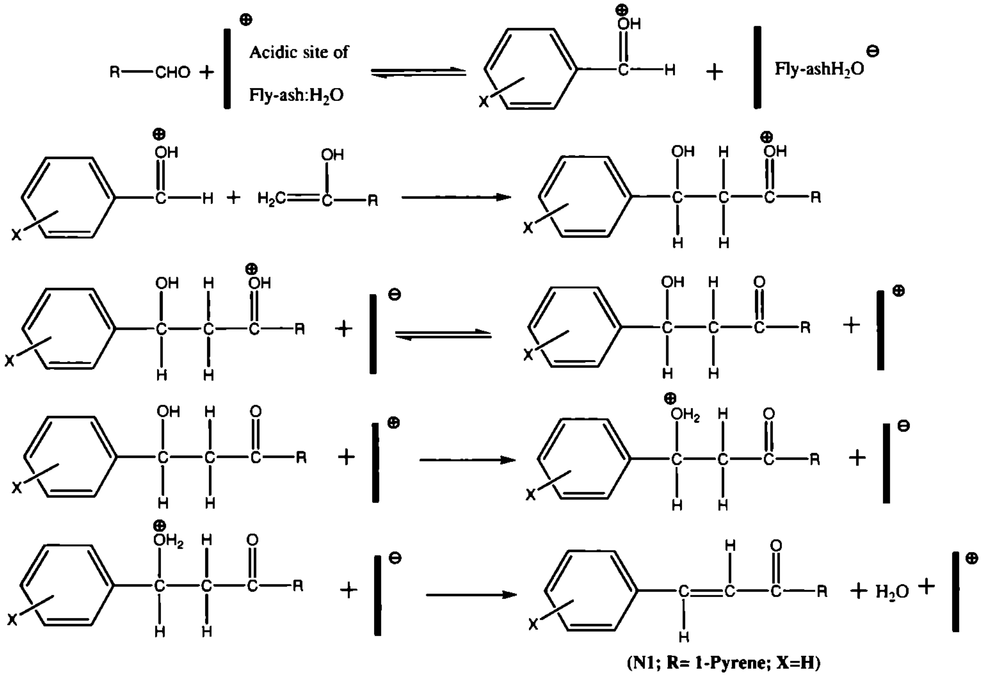


Fig. 1. The proposed mechanism for the synthesis of chalcones by fly-ash:H_2O catalyzed aldol condensation.

Table 1. Analytical and mass spectral data of chalcones synthesized by Fly-ash- H_2SO_4 catalyzed aryl methyl ketones and substituted benzaldehydes reaction of the type $R{-}CO{-}CH_3 + R'{-}CHO \rightarrow R{-}CO{-}CH{=}CH{-}R'$ under microwave irradiation.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
A1	C_6H_5	C_6H_5	$C_6H_5COCH{=}CHC_6H_5$	208	68	55-56 (55-58)[20]	208[M$^+$]
A2	C_6H_5	4-NH_2-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-NH_2-4	224	60	117-118 (117-117)[20]	244[M$^+$]
A3	C_6H_5	4-Br-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-Br-4	286	67	101-102 (101-102)[20]	286[M$^+$] 288[M^{2+}]
A4	C_6H_5	4-Cl-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-Cl-4	252	67	116-117 (116-117)[20]	252[M$^+$] 254[M^{2+}] 254[M^{2+}]
A5	C_6H_5	4-$N(CH_3)_2C_6H_4$	$C_6H_5COCH{=}CHC_6H_4$-$N(CH_3)_2$-4	234	62	115-116 (115-116)[20]	234[M$^+$]
A6	C_6H_5	4-F-C_6H_4	$C_6H_5COCH{=}CHC_6H_5$-F-4	225	62	85-86 (85-86)[20]	225[M$^+$] 227[M^{2+}] 227[M^{2+}]
A7	C_6H_5	4-OH-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-OH-4	224	63	187-188 (187-118)[20]	224[M$^+$]
A8	C_6H_5	4-I-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-I-4	333	61	87-88 (87-88)[20]	333[M$^+$] 335[M^{2+}]
A9	C_6H_5	4-$OCH_3C_6H_4$	$C_6H_5COCH{=}CHC_6H_4$-OCH_3-4	276	65	127-128 (127-128)[20]	276[M$^+$]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
A10	C_6H_5	4-CH_3-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-CH_3-4	219	66	42-47 (42-47)[20]	219[M^+]
A11	C_6H_5	4-NO_2-C_6H_4	$C_6H_5COCH{=}CHC_6H_4$-NO_2-4	254	68	158-159 (158-159) [20]	254[M^+]
B1	4-NO_2-C_6H_4	C_6H_5	4-$FC_6H_4COCH{=}CHC_6H_5$	253	68	119-120 (118-119)[25]	253[M^+]
B2	4-NO_2-C_6H_4	3-Br-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-Br-3	332	65	162-163 (162-163)[25]	332[M^+] 334[M^{2+}]
B3	4-NO_2-C_6H_4	4-Br-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-Br-4	332	65	168-169 (168-169)[25]	332[M^+] 334[M^{2+}]
B4	4-NO_2-C_6H_4	3-Cl-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-Cl-3	287	64	142-143 (142-143)[25]	287[M^+] 289[M^{2+}]
B5	4-NO_2-C_6H_4	4-Cl-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-Cl-4	287	65	244-245 (244-245)[25]	287[M^+] 289[M^{2+}]
B6	4-NO_2-C_6H_4	4-F-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-F-4	271	63	228-229 (228-229)[25]	271[M^+] 273[M^{2+}]
B7	4-NO_2-C_6H_4	2-OCH_3-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-OCH_3-2	283	67	156-157 (156-157)[25]	283[M^+]
B8	4-NO_2-C_6H_4	4-OCH_3-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-OCH_3-4	283	68	188-189 (188-189)[25]	283[M^+]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
B9	4-NO_2-C_6H_4	4-CH_3-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$-CH_3-4	267	68	176-177 (176-177)[25]	267[M^+]
B10	4-NO_2-C_6H_4	2-NO_2-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$- NO_2-2	298	63	184-185 (184-185)[25]	298[M^+]
B11	4-NO_2-C_6H_4	3-NO_2-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$- NO_2-3	298	65	181-182 (180-181)[25]	298[M^+]
B12	4-NO_2-C_6H_4	4-NO_2-C_6H_4	4-NO_2-$C_6H_4COCH{=}CHC_6H_4$- NO_2-4	298	66	217-218 (216-217)[25]	298[M^+]
C1	$C_{10}H_7$(1-Naph)	C_6H_5	$C_{10}H_7COCH{=}CHC_6H_5$	258	61	100-102 (100-102)[26]	258[M^+]
C2	$C_{10}H_7$(1-Naph)	3-NH_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NH_2-3	273	60	97-98 (97-98)[26]	273[M^+]
C3	$C_{10}H_7$(1-Naph)	4-NH_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NH_2-4	273	61	115-116 (115-116)[26]	273[M^+]
C4	$C_{10}H_7$(1-Naph)	3-Br-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-Br-3	336	66	79-80 (79)[26]	336[M^+] 338[M^{2+}]
C5	$C_{10}H_7$(1-Naph)	3-Cl-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-Cl-3	292	65	88-89 (87)[26]	292[M^+] 294[M^{2+}]
C6	$C_{10}H_7$(1-Naph)	4-Cl-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-Cl-4	292	63	108-109 (108)[26]	292[M^+] 294[M^{2+}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
C7	$C_{10}H_7$(1-Naph)	4-$N(CH_3)_2C_6H_4$	$C_{10}H_7COCH{=}CHC_6H_4$- $N(CH_3)_2$-4	301	62	85-56 (85)[26]	301[M^+]
C8	$C_{10}H_7$(1-Naph)	4-OH-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-OH-4	274	60	104-105 (104)[26]	274[M^+]
C9	$C_{10}H_7$(1-Naph)	4-OCH_3-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-OCH_3-4	288	67	93-94 (93)[26]	288[M^+]
C10	$C_{10}H_7$(1-Naph)	4-CH_3-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-CH_3-4	272	64	96-97 (96)[26]	272[M^+]
C11	$C_{10}H_7$(1-Naph)	2-NO_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NO_2-2	303	60	84-85 (83)[26]	303[M^+]
C12	$C_{10}H_7$(1-Naph)	3-NO_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NO_2-3	303	64	110-111 (110)[26]	303[M^+]
C13	$C_{10}H_7$(1-Naph)	4-NO_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NO_2-4	303	62	114-115 (114-115)[26]	303[M^+]
D1	4-$BrC_{10}H_6$ (1-Naph)	C_6H_5	4-$BrC_{10}H_6COCH{=}CHC_6H_5$	337	60	103-104 (103-104)[4]	337[M^+] 339[M^{2+}]
D2	4-$BrC_{10}H_6$ (1-Naph)	3-NH_2-C_6H_4	4-$BrC_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-3	352	61	72-23 (72-73)[4]	352[M^+] 354[M^{2+}]
D3	4-$BrC_{10}H_6$ (1-Naph)	4-NH_2-C_6H_4	4-$BrC_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-4	352	62	81-82 (81-82)[4]	352[M^+] 354[M^{2+}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
D4	4-$BrC_{10}H_6$ (1-Naph)	3-Br-C_6H_4	4-$BrC_{10}H_6COCH{=}CHC_6H_4$-Br-3	416	64	(107-108) (107-108)[4]	416[M^+] 418[M^{2+}] 420[M^{4+}]
D5	4-$BrC_{10}H_6$ (1-Naph)	3-Cl-C_6H_4	4-$BrC_{10}H_6COCH{=}CHC_6H_4$-Cl-3	372	65	110-111 (110-111)[4]	372[M^+] 374[M^{2+}] 420[M^{4+}]
D6	4-$BrC_{10}H_6$ (1-Naph)	4-Cl-C_6H_4	4-$BrC_{10}H_6COCH{=}CHC_6H_4$-Cl-4	372	65	100-101 (100-101)[4]	372[M^+] 374[M^{2+}] 420[M^{4+}]
D7	4-$BrC_{10}H_6$ (1-Naph)	4-$N(CH_3)_2C_6H_4$	4-$BrC_{10}H_6COCH{=}CHC_6H_4$- $N(CH_3)_2$-4	380	64	118-119 (118-119)[4]	380[M^+] 382[M^{2+}]
D8	4-$BrC_{10}H_6$ (1-Naph)	4-OH-C_6H_4	4-Br-$C_{10}H_6COCH{=}CHC_6H_4$-OH-4	353	62	97-98 (97-98)[4]	353[M^+] 355[M^{2+}]
D9	4-$BrC_{10}H_6$ (1-Naph)	4-OCH_3-C_6H_4	4-Br-$C_{10}H_6COCH{=}CHC_6H_4$-OCH_3-4	367	66	123-124 (100-102)[4]	367[M^+] 368[M^{2+}]
D10	4-$BrC_{10}H_6$ (1-Naph)	4-CH_3-C_6H_4	4-Br-$C_{10}H_6COCH{=}CHC_6H_4$-CH_3-4	351	67	95-96 (95-96)[4]	351[M^+] 353[M^{2+}]
D11	4-$BrC_{10}H_6$ (1-Naph)	2-NO_2-C_6H_4	4-Br-$C_{10}H_6COCH{=}CHC_6H_4$-NO_2-2	382	61	76-77 (100-102)[4]	382[M^+] 384[M^{2+}]
D12	4-$BrC_{10}H_6$ (1-Naph)	3-NO_2-C_6H_4	4-Br-$C_{10}H_6COCH{=}CHC_6H_4$-NO_2-3	382	63	88-89 (88-89)[4]	382[M^+] 384[M^{2+}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
D13	4-BrC$_{10}$H$_6$ (1-Naph)	4-NO$_2$-C$_6$H$_4$	4-Br-C$_{10}$H$_6$COCH=CHC$_6$H$_4$-NO$_2$-4	382	66	116-117 (116-117)[4]	382[M$^+$] 384[M^{2+}]
E1	4-ClC$_{10}$H$_6$ (1-Naph)	C$_6$H$_5$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_5$	292	94	122-123 (122-123)[5]	292[M$^+$] 294[M^{2+}]
E2	4-ClC$_{10}$H$_6$ (1-Naph)	3-NH$_2$-C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$-NH$_2$-3	307	61	97-98 (97-98)[5]	307[M^{2+}] 309[M^{2+}]
E3	4-ClC$_{10}$H$_6$ (1-Naph)	4-NH$_2$-C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$-NH$_2$-4	335	62	123-124 (123-124)[5]	335[M$^+$] 357[M^{2+}]
E4	4-ClC$_{10}$H$_6$ (1-Naph)	3-Br-C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$-Br-3	371	64	118-119 (118-119)[5]	371[M$^+$] 373[M^{2+}] 385[M^{4+}]
E5	4-ClC$_{10}$H$_6$ (1-Naph)	3-Cl-C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$-Cl-3	327	65	67-68 (67-68)[5]	327[M$^+$] 329[M^{2+}] 331[M^{4+}]
E6	4-ClC$_{10}$H$_6$ (1-Naph)	4-Cl-C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$-Cl-4	327	65	115-116 (115-116)[5]	327[M$^+$] 329[M^{2+}] 331[M^{4+}]
E7	4-ClC$_{10}$H$_6$ (1-Naph)	4-N(CH$_3$)$_2$C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$- N(CH$_3$)$_2$-4	335	64	123-124 (123-124)[5]	335[M$^+$] 337[M^{2+}]
E8	4-ClC$_{10}$H$_6$ (1-Naph)	4-OH-C$_6$H$_4$	4-ClC$_{10}$H$_6$COCH=CHC$_6$H$_4$-OH-4	308	62	82-83 (82-83)[5]	308[M$^+$] 310[M^{2+}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
E9	4-$ClC_{10}H_6$ (1-Naph)	4-OCH_3-C_6H_4	4-$ClC_{10}H_6COCH{=}CHC_6H_4$-$OCH_3$-4	322	66	140-141 (140-141)[5]	322[M^+] 324[M^{2+}]
E10	4-$ClC_{10}H_6$ (1-Naph)	4-CH_3-C_6H_4	4-$ClC_{10}H_6COCH{=}CHC_6H_4$-$CH_3$-4	306	67	137-138 (137-138)[5]	306[M^+] 308[M^{2+}]
E11	4-$ClC_{10}H_6$ (1-Naph)	2-NO_2-C_6H_4	4-$ClC_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-2	337	61	111-112 (111-112)[5]	337[M^+] 339[M^{2+}]
E12	4-$ClC_{10}H_6$ (1-Naph)	3-NO_2-C_6H_4	4-$ClC_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-3	337	63	100-102 (100-102)[5]	337[M^+] 339[M^{2+}]
E13	4-$ClC_{10}H_6$ (1-Naph)	4-NO_2-C_6H_4	4-$ClC_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-4	337	66	100-102 (100-102)[5]	337[M^+] 339[M^{2+}]
F1	4-$OCH_3C_1H_6$ (1-Naph)	C_6H_5	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_5$	310	65	113-114 (113-114)[27]	310[M^+]
F2	4-$OCH_3C_{10}H_6$ (1-Naph)	3-NH_2-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-3	303	61	99-100 (99-100)[27]	303[M^+]
F3	4-$OCH_3C_{10}H_6$ (1-Naph)	4-NH_2-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-4	303	61	82-83 (82-83)[27]	303[M^+]
F4	4-$OCH_3C_{10}H_6$ (1-Naph)	3-Br-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-Br-3	367	65	78-79 (78-79)[27]	367[M^+] 369[M^{2+}]
F5	4-$OCH_3C_{10}H_6$ (1-Naph)	3-Cl-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-Cl-3	322	66	121-122 (120-121)[27]	322[M^+] 324[M^{2+}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
F6	4-$OCH_3C_{10}H_6$ (1-Naph)	4-Cl-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-Cl-4	322	68	137-138 (137-138)[27]	322[M^+] 324[M^{2+}]
F7	4-$OCH_3C_{10}H_6$ (1-Naph)	4-$N(CH_3)_2C_6H_4$	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$N(CH_3)_2$-4	331	60	126-127 (126-127)[27]	331[M^+]
F8	4-$OCH_3C_{10}H_6$ (1-Naph)	4-OH-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-OH-4	304	60	113-114 (113-114)[27]	304[M^+]
F9	4-$OCH_3C_{10}H_6$ (1-Naph)	4-OCH_3-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$OCH_3$-4	318	67	88-89 (88-89)[27]	318[M^+]
F10	4-$OCH_3C_{10}H_6$ (1-Naph)	4-CH_3-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$CH_3$-4	302	66	114-115 (114-115)[27]	302[M^+]
F11	4-$OCH_3C_{10}H_6$ (1-Naph)	2-NO_2-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-2	333	62	106-107 (106-107)[27]	333[M^+]
F12	4-$OCH_3C_{10}H_6$ (1-Naph)	3-NO_2-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-3	333	65	113-114 (113-114)[27]	333[M^+]
F13	4-$OCH_3C_{10}H_6$ (1-Naph)	4-NO_2-C_6H_4	4-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-4	333	68	141-142 (141-142)[27]	333[M^+]
G1	4-$CH_3C_{10}H_6$ (1-Naph)	C_6H_5	4-$CH_3C_{10}H_6COCH{=}CHC_6H_5$	272	68	98-99 (98-99)[28]	272[M^+]
G2	4-$CH_3C_{10}H_6$ (1-Naph)	3-NH_2-C_6H_4	4-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-3	287	62	70-71 (70-71)[28]	287[M^+]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
G3	4-$CH_3C_{10}H_6$ (1-Naph)	4-NH_2-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-$NH_2$-4	287	61	122-123 (122-123)[28]	287[M^+]
G4	4-$CH_3C_{10}H_6$ (1-Naph)	3-Br-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-Br-3	350	66	126-127 (126-127)[28]	350[M^+] 352[M^{2+}]
G5	4-$CH_3C_{10}H_6$ (1-Naph)	3-Cl-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-Cl-3	306	65	97-98 (97-98)[28]	306[M^+] 308[M^{2+}]
G6	4-$CH_3C_{10}H_6$ (1-Naph)	4-Cl-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-Cl-4	306	66	100-102 (100-102)[28]	306[M^+] 308[M^{2+}]
G7	4-$CH_3C_{10}H_6$ (1-Naph)	4-$N(CH_3)_2C_6H_4$	4-$CH_3C_{10}H_6COCH=CHC_6H_4$- $N(CH_3)_2$-4	315	61	135-136 (135-136)[28]	315[M^+]
G8	4-$CH_3C_{10}H_6$ (1-Naph)	4-OH-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-OH-4	288	60	110-111 (110-111)[28]	288[M^+]
G9	4-$CH_3C_{10}H_6$ (1-Naph)	4-OCH_3-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-$OCH_3$-4	302	68	106-107 (106-107)[28]	302[M^+]
G10	4-$CH_3C_{10}H_6$ (1-Naph)	4-CH_3-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-$CH_3$-4	286	67	122-123 (122-23)[28]	286[M^+]
G11	4-$CH_3C_{10}H_6$ (1-Naph)	2-NO_2-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-$NO_2$-2	317	60	87-88 (87-88)[28]	317[M^+]
G12	4-$CH_3C_{10}H_6$ (1-Naph)	3-NO_2-C_6H_4	4-$CH_3C_{10}H_6COCH=CHC_6H_4$-$NO_2$-3	317	65	100-102 (100-102)[28]	317[M^+]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
G13	4-$CH_3C_{10}H_6$ (1-Naph)	4-NO_2-C_6H_4	4-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-4	317	68	100-102 (100-102) 28]	317[M^+]
H1	$C_{10}H_7$(2-Naph)	C_6H_5	$C_{10}H_7COCH{=}CHC_6H_5$	258	67	104-105 (104-105)[29]	258[M^+]
H2	$C_{10}H_7$(2-Naph)	3-NH_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NH_2-3	273	60	101-102 (101)[29]	273[M^+]
H3	$C_{10}H_7$(2-Naph)	4-NH_2-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-NH_2-4	258	60	124-125 (100-102)[29]	258[M^+]
H4	$C_{10}H_7$(2-Naph)	3-Br-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-Br-3	337	64	86-88 (100-102)[29]	337[M^+] 339[M^{2+}]
H5	$C_{10}H_7$(2-Naph)	3-Cl-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-Cl-3	293	65	114-115 (114-115)[29]	293[M^+] 295[M^{2+}]
H6	$C_{10}H_7$(2-Naph)	4-Cl-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-Cl-4	293	66	147-148 (147-148[29]	293[M^+] 295[M^{2+}]
H7	$C_{10}H_7$(2-Naph)	4-$N(CH_3)_2C_6H_4$	$C_{10}H_7COCH{=}CHC_6H_4$- $N(CH_3)_2$-4	301	61	112-113 (112-113)[29]	301[M^+]
H8	$C_{10}H_7$(2-Naph)	4-OH-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-OH-4	274	60	141-142 (141-142)[29]	274[M^+]
H9	$C_{10}H_7$(2-Naph)	4-OCH_3-C_6H_4	$C_{10}H_7COCH{=}CHC_6H_4$-OCH_3-4	288	67	83-84 (83-84)[29]	288[M^+]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
H10	$C_{10}H_7$(2-Naph)	4-CH_3-C_6H_4	$C_{10}H_7COCH=CHC_6H_4$-CH_3-4	272	66	118-119 (118-119)[29]	272[M^+]
H11	$C_{10}H_7$(2-Naph)	2-NO_2-C_6H_4	$C_{10}H_7COCH=CHC_6H_4$-NO_2-2	303	60	138-139 (138-139[29]	303[M^+]
H12	$C_{10}H_7$(2-Naph)	3-NO_2-C_6H_4	$C_{10}H_7COCH=CHC_6H_4$-NO_2-3	303	64	139-140 (138-139)[29]	303[M^+]
H13	$C_{10}H_7$(2-Naph)	4-NO_2-C_6H_4	$C_{10}H_7COCH=CHC_6H_4$-NO_2-4	303	66	148-149 (147-148)[29]	303[M^+]
I1	6-$OCH_3C_{10}H_6$ (2-Naph)	C_6H_5	6-$OCH_3C_{10}H_6COCH=CHC_6H_5$	288	68	67-68 (67-68)[30]	288[M^+]
I2	6-$OCH_3C_{10}H_6$ (2-Naph)	3-NH_2-C_6H_4	6-$OCH_3C_{10}H_6COCH=CHC_6H_4$-$NH_2$-3	303	63	97-98 (97-98)[30]	303[M^+]
I3	6-$OCH_3C_{10}H_6$ (2-Naph)	4-NH_2-C_6H_4	6-$OCH_3C_{10}H_6COCH=CHC_6H_4$-$NH_2$-4	303	64	86-87 (86-87)[30]	303[M^+]
I4	6-$OCH_3C_{10}H_6$ (2-Naph)	3-Br-C_6H_4	6-$OCH_3C_{10}H_6COCH=CHC_6H_4$-Br-3	367	63	114-115 (114-115)[30]	367[M^+] 369[M^{2+}]
I5	6-$OCH_3C_{10}H_6$ (2-Naph)	3-Cl-C_6H_4	6-$OCH_3C_{10}H_6COCH=CHC_6H_4$-Cl-3	324	61	101-102 (101-102)[30]	324[M^+] 326[M^{2+}]
I6	6-$OCH_3C_{10}H_6$ (2-Naph)	4-Cl-C_6H_4	6-$OCH_3C_{10}H_6COCH=CHC_6H_4$-Cl-4	324	64	122-123 (122-123)[30]	324[M^+] 326[M^{2+}]

cont. Table I.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
I7	6-$OCH_3C_{10}H_6$ (2-Naph)	4-$N(CH_3)_2C_6H_4$	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$N(CH_3)_2$-4	331	60	142-143 (142-143)[30]	331[M^+]
I8	6-$OCH_3C_{10}H_6$ (2-Naph)	4-OH-C_6H_4	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-OH-4	294	60	117-118 (117-118)[30]	294[M^+]
I9	6-$OCH_3C_{10}H_6$ (2-Naph)	4-OCH_3-C_6H_4	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$OCH_3$-4	317	67	107-108 (107-108)[30]	317[M^+]
I10	6-$OCH_3C_{10}H_6$ (2-Naph)	4-CH_3-C_6H_4	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$CH_3$-4	301	65	87-88 (87-88)[30]	301[M^+]
I11	6-$OCH_3C_{10}H_6$ (2-Naph)	2-NO_2-C_6H_4	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-2	333	61	92-93 (92-93)[30]	333[M^+]
I12	6-$OCH_3C_{10}H_6$ (2-Naph)	3-NO_2-C_6H_4	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-3	333	64	133-134 (133-134)[30]	333[M^+]
I13	6-$OCH_3C_{10}H_6$ (2-Naph)	4-NO_2-C_6H_4	6-$OCH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-4	333	65	148-149 (148-149)[30]	333[M^+]
J1	6-$CH_3C_{10}H_6$ (2-Naph)	C_6H_5	6-$CH_3C_{10}H_6COCH{=}CHC_6H_5$	272	67	123-124 (123-124)[31]	272[M^+]
J2	6-$CH_3C_{10}H_6$ (2-Naph)	3-NH_2-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-3	287	60	106-107 (106-107)[31]	287[M^+]
J3	6-$CH_3C_{10}H_6$(2-Naph)	4-NH_2-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NH_2$-4	287	60	95-96 (95-96)[31]	287[M^+]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
J4	6-$CH_3C_{10}H_6$ (2-Naph)	3-Br-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-Br-3	351	65	82-83 (82-83)[31]	351[M^+] 353[M^{2+}]
J5	6-$CH_3C_{10}H_6$ (2-Naph)	3-Cl-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-Cl-3	306	65	146-147 (146-147)[31]	306[M^+] 308[M^{2+}]
J6	6-$CH_3C_{10}H_6$ (2-Naph)	4-Cl-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-Cl-4	306	66	154-155 (154-155)[31]	306[M^+] 308[M^{2+}]
J7	6-$CH_3C_{10}H_6$ (2-Naph)	4-$N(CH_3)_2C_6H_4$	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$N(CH_3)_2$-4	315	60	73-74 (73-74)[31]	315[M^+]
J8	6-$CH_3C_{10}H_6$ (2-Naph)	4-OH-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-OH-4	228	60	80-81 (80-81)[31]	228[M^+]
J9	6-$CH_3C_{10}H_6$ (2-Naph)	4-OCH_3-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$OCH_3$-4	302	67	131-132 (131-133)[31]	302[M^+]
J10	6-$CH_3C_{10}H_6$ (2-Naph)	4-CH_3-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$CH_3$-4	286	68	109-110 (109-110)[31]	286[M^+]
J11	6-$CH_3C_{10}H_6$ (2-Naph)	2-NO_2-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-2	317	60	106-107 (106-107)[31]	317[M^+]
J12	6-$CH_3C_{10}H_6$ (2-Naph)	3-NO_2-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-3	317	62	134-135 (134-135)[31]	317[M^+]
J13	6-$CH_3C_{10}H_6$(2-Naph)	4-NO_2-C_6H_4	6-$CH_3C_{10}H_6COCH{=}CHC_6H_4$-$NO_2$-4	317	64	177-178 (177-178)[31]	317[M^+]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
K1	$C_{13}H_9$(2-Fluorene)	C_6H_5	$C_{13}H_9COCH{=}CHC_6H_5$	296	66	150-151 (149-150)[32]	296[M^+]
K2	$C_{13}H_9$(2-Fluorene)	3-NH_2-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$- NH_2-3	311	61	95-96 (95-96)[32]	311[M^+]
K3	$C_{13}H_9$(2-Fluorene)	3-Cl-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$-Cl-3	330	64	44-45 (43-44) [32]	330[M^+] 332[M^{2+}]
K4	$C_{13}H_9$(2-Fluorene)	4-Cl-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$-Cl-4	330	64	83-84 (83-84)[32]	330[M^+] 332[M^{2+}]
K5	$C_{13}H_9$(2-Fluorene)	4-$N(CH_3)_2C_6H_4$	$C_{13}H_9COCH{=}CHC_6H_4$- $N(CH_3)_2$-4	339	62	89-90 (89-90)[32]	339[M^+]
K6	$C_{13}H_9$(2-Fluorene)	4-OH-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$-OH-4	312	61	91-92 (91-92)[32]	312[M^+]
K7	$C_{13}H_9$(2-Fluorene)	4-OCH_3-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$-OCH_3-4	326	65	94-95 (94-95)[32]	326[M^{2+}]
K8	$C_{13}H_9$(2-Fluorene)	3-NO_2-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$-NO_2-3	341	64	77-78 (77-78)[32]	341[M^{2+}]
K9	$C_{13}H_9$(2-Fluorene)	4-NO_2-C_6H_4	$C_{13}H_9COCH{=}CHC_6H_4$-NO_2-4	341	68	85-86 (85-86)[32]	332[M^{2+}]
L1	5-BrC_4H_2S (2-Thienyl)	C_6H_5	5-$BrC_4H_2SCOCH{=}CHC_6H_5$	292	68	107-110 (110)[12]	292[M^+] 294[M^{2+}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
L2	5-BrC_4H_2S (2-Thienyl)	4-BrC_6H_4	5-$BrC_4H_2SCOCH{=}CHC_6H_4$-Br-4	71	65	173-174 (173-174)[12]	371[M^+] 373[M^{+2}] 375[M^{+4}]
L3	5-BrC_4H_2S (2-Thienyl)	2-ClC_6H_4	5-$BrC_4H_2SCOCH{=}CHC_6H_4Cl$-2	326	63	143-145 (143-145)[12]	326[M^+] 328[M^{+2}] 330[M^{+4}]
L4	5-BrC_4H_2S (2-Thienyl)	4-ClC_6H_4	5-$BrC_4H_2SCOCH{=}CHC_6H_4Cl$-4	326	67	155-157 (155-157)[12]	326[M+] 328[M+2] 330[M^{+4}]
L5	5-BrC_4H_2S (2-Thienyl)	4-$N(CH_3)_2C_6H_4$	5-$BrC_4H_2SCOCH{=}CHC_6H_4N(CH_3)_2$-4	335	60	142-143 (142-143)[12]	335[M^+] 337[M^{+2}]
L6	5-BrC_4H_2S (2-Thienyl)	4-FC_6H_4	5-$BrC_4H_2SCOCH{=}CHC_6H_4F$-4	309	61	164-165 (164-165)[12]	309[M^+] 311[M^{+2}] 313[M^{+4}]
L7	5-BrC_4H_2S (2-Thienyl)	4-IC_6H_4	5-$BrC_4H_2SCOCH{=}CHC_6H_4I$	416	64	184-185 (184-185)[12]	416[M^+] 418[M^{+2}] 420[M^{+4}]
L8	5-BrC_4H_2S (2-Thienyl)	4-$OCH_3C_6H_5$	5-$BrC_4H_2SCOCH{=}CHC_6H_4OCH_3$-4	322	68	148-149 (148-149) [12]	322[M^+] 324[M^{+2}]

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
L9	5-BrC_4H_2S (2-Thienyl)	4-$CH_3C_6H_5C_6H_5$	5-$BrC_4H_2SCOCH{=}CHC_6H_4CH_3$-4	306	68	151-152 (151-152)[12]	$306[M^{+}]$ $308[M^{+2}]$
M1	5-ClC_4H_2S (2-Thienyl)	C_6H_5	5-$ClC_4H_2SCOCH{=}CHC_6H_5$	248	68	161-162 (161-162)[33]	248[M+] $250[M^{+2}]$
M2	5-ClC_4H_2S (2-Thienyl)	3-BrC_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4$-Br-3	327	66	88-89 (88-89) [33]	$327[M^{+}]$ $329[M^{+2}]$ $331[M^{+4}]$
M3	5-ClC_4H_2S (2-Thienyl)	3-ClC_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4Cl$-3	327	65	125-126 (125-126)[33]	327[M+] 329[M+2] $331[M^{+4}]$
M4	5-ClC_4H_2S (2-Thienyl)	2-FC_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4F$-2	266	63	82-83 (82-83)[33]	$266[M^{+}]$ $268[M^{+2}]$ $270[M^{+4}]$
M5	5-ClC_4H_2S (2-Thienyl)	4-FC_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4F$-4	266	62	108-109	$266[M^{+}]$ $268[M^{+2}]$ $270[M^{+4}]$
M6	5-ClC_4H_2S (2-Thienyl)	4-OH-C_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4OH$-4	264	60	103-104 (103-104)[33]	$264[M^{+}]$ $266[M^{+2}]$
M7	5-ClC_4H_2S (2-Thienyl)	2-$OCH_3C_6H_4$	5-$ClC_4H_2SCOCH{=}CHC_6H_4OCH_3$-2	278	67	105-106 (105-106)[33]	$278[M^{+}]$ $280[M^{+2}]$

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
M8	5-ClC_4H_2S (2-Thienyl)	4-$OCH_3C_6H_4$	5-$ClC_4H_2SCOCH{=}CHC_6H_4OCH_3$-4	278	68	98-99 (98-99)[33]	278[M^+] 280[M^{+2}]
M9	5-ClC_4H_2S (2-Thienyl)	2-$CH_3C_6H_4$	5-$ClC_4H_2SCOCH{=}CHC_6H_4CH_3$-2	262	68	65-66 (65-66)[33]	262[M^+] 264[M^{+2}]
M10	5-ClC_4H_2S (2-Thienyl)	4-$CH_3C_6H_4$	5-$ClC_4H_2SCOCH{=}CHC_6H_4CH_3$-4	262	66	71-72 (71-72)[33]	262[M+] 264[M^{+2}]
M11	5-ClC_4H_2S (2-Thienyl)	4-NO_2-C_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4$-NO_2-4	293	66	75-76 (75-76)[33]	293[M+] 295[M^{+2}]
M12	5-ClC_4H_2S (2-Thienyl)	4-C_6H_5-O-C_6H_4	5-$ClC_4H_2SCOCH{=}CHC_6H_4$-O-C_6H_5-4	340	67	72-73 (72-73)[33]	340[M+] 342[M^{+2}]
N1	$C_{16}H_9$ (1-Pyrene)	C_6H_5	$C_{16}H_9COCH{=}CHC_6H_5$	332	67	122-123	332[M+], 255, 242, 229, 201, 176, 138, 103, 90, 77, 66, 64
N2	$C_{16}H_9$ (1-Pyrene)	3-Br-C_6H_4	$C_{16}H_9COCH{=}CHC_6H_4$-Br-3	411	64	119-120	411[M^+], 423[M^{2+}], 331, 272, 255, 242, 229, 208, 180, 168, 155, 138, 79

cont. Table 1.

Entry	R	R'	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
N3	$C_{16}H_9$ (1-Pyrene)	2-Cl-C_6H_4	$C_{16}H_9COCH{=}CHC_6H_4$-Cl-2	366	62	119-120	366[M^+], 368[M^{2+}], 331, 255, 242, 229, 201, 138, 131, 90, 77, 35
N4	$C_{16}H_9$ (1-Pyrene)	3-Cl-C_6H_4	$C_{16}H_9COCH{=}CHC_6H_4$-Cl-3	366	65	262-263	366[M^+], 368[M^{2+}], 331, 255, 242, 229, 138, 131, 90, 77, 76, 46, 35
N5	$C_{16}H_9$ (1-Pyrene)	4-Cl-C_6H_4	$C_{16}H_9COCH{=}CHC_6H_4$-Cl-4	366	66	96-97	366[M^+], 368[M^{2+}], 331, 255, 242, 201, 180, 168, 138, 131, 90, 77, 35
N6	$C_{16}H_9$ (1-Pyrene)	4-F-C_6H_4	$C_{16}H_9COCH{=}CHC_6H_4$-F-4	350	61	108-109	350[M^+], 331, 255, 249, 229, 212, 201, 175, 149, 138, 121, 101, 77, 19

cont. Table 1.

Entry	R	R′	Product	M.W.	Yield [%]	M.p. [°C]	Mass (m/z)
N7	$C_{16}H_9$ (1-Pyrene)	4-$CH_3C_6H_5$	$C_{16}H_9COCH{=}CHC_6H_4$-OCH_3-4	346	67	108-109	346[M^+], 331, 255, 242, 229, 208, 201, 176, 145, 138, 117, 91, 76, 66
N8	$C_{16}H_9$ (1-Pyrene)	3-NO_2-C_6H_5	$C_{16}H_9COCH{=}CHC_6H_4$-NO_2-3	377	65	128-129	377[M^+], 331, 276, 255, 239, 229, 201, 176, 138, 135, 131, 122, 90, 77, 46
N9	$C_{16}H_9$ (1-Pyrene)	4-NO_2-C_6H_5	$C_{16}H_9COCH{=}CHC_6H_4$-NO_2-4	377	67	136-137	377[M^+], 331, 276, 255, 242, 239, 229, 201, 176, 148, 138, 135, 131, 122

The spectroscopic data of 1-pyrenyl chalcones are summarized below.

E **1-(1-Pyrene)-3-phenyl-2-propen-1-one (N1):** IR, KBr (cm^{-1}): ν = 1632.88 CO$_{s\text{-}cis}$, 1596.71CO$_{s\text{-}trans}$, 1106.85 CH$_{ip}$, 763.28 CH$_{op}$, 1072.31 CH=CH$_{op}$, 556.41 C=C$_{op}$; ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = 8.470 (*d*, 1H, Hα, *J* = 17Hz), 8.145 (*d*, 1H, Hβ, *J* = 17Hz), 7.253-8.623 (*m*, 16H, ArH); ^{13}C NMR (100MHz, CDCl$_3$, ppm) δ = 196.12 (CO), 124.12 (Cα), 146.16 (Cβ), 124.47-146.14 (ArC).

E **1-(1- Pyrene)-3-(3-Brphenyl)-2-propen-1-one (N2):** IR, KBr (cm^{-1}): ν = 1667.71 CO$_{s\text{-}cis}$, 159.63 CO$_{s\text{-}trans}$, 1172.60 CH$_{ip}$, 861.38 CH$_{op}$, 1068.49 CH=CH$_{op}$, 583.43 C=C$_{op}$; ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = 8.725 (*d*, 1H, Hα, *J* = 15Hz), 9.175 (*d*, 1H, Hβ, *J* = 15Hz), 8.172-8.420 (*m*, 15H, ArH); ^{13}C NMR (100 MHz, CDCl$_3$, ppm) δ = 195.94 (CO), 121.14 (Cα), 146.56 (Cβ), 114.22-142.62 (Ar-C).

E **1-(1-Pyrene)-3-(2-chlorophenyl)-2-propen-1-one(N3):** IR, KBr (cm^{-1}): ν = 1633.17 CO$_{s\text{-}cis}$, 1590.90 CO$_{s\text{-}trans}$, 1119.54 CH$_{ip}$, 745.10 CH$_{op}$, 1079.45 CH=CH$_{op}$, 591.45 C=C$_{op}$; ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = 7.45 (*d*, 1H, Hα, *J* = 16Hz), 8.19 (*d*, 1H, Hβ, *J* = 16Hz), 7.257-8.690 (*m*, 15H, ArH); ^{13}C NMR (100MHz, CDCl$_3$, ppm) δ = 195.54 (CO), 124.05 (Cα), 141.63 (Cβ), 124.44-135-54 (Ar-C).

E **1-(1-Pyrene)-3-(3-Clphenyl)-2-propen-1-one (N4):** IR, KBr (cm^{-1}): ν = 1600.00 CO$_{s\text{-}cis}$, 1593.68 CO$_{s\text{-}trans}$, 1123.29 CH$_{ip}$, 761.64 CH$_{op}$, 1019.72 CH=CH$_{op}$, 569.86 C=C$_{op}$; ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = 7.477 (*d*, 1H, Hα, *J* = 18Hz), 8.235 (*d*, 1H, Hβ, *J* = 18Hz), 7.260-8.640 (*m*, 15H, ArH); ^{13}C NMR (100MHz, CDCl$_3$, ppm) δ = 195.41 (CO), 124.12 (Cα), 144.08 (Cβ), 124.43-136.60 (Ar-C).

E **1-(1-Pyrene)-3-(4-chlorophenyl)-2-propen-1-one** (**N5**): IR, KBr (cm^{-1}): ν = 1655.68 CO$_{s\text{-}cis}$, 1590.64 CO$_{s\text{-}trans}$, 1119.07 CH$_{ip}$, 780.23 CH$_{op}$, 1075.12 CH=CH$_{op}$, 602.64 C=C$_{op}$; ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = 7.034(*d*, 1H, Hα, *J* = 16Hz), 7.685 (*d*, 1H, Hβ, *J* = 16Hz), 7.099-8.634 (*m*, 15H, ArH); ^{13}C NMR (100MHz, CDCl$_3$, ppm) δ = 195.65 (CO), 124.04 (Cα), 144.44 (Cβ), 124.11-142.11 (Ar-C).

E **1-(1-Pyrene)-3-(4-Fphenyl)-2-propen-1-one** (**N6**): IR, KBr (cm^{-1}): ν = 1608.09 CO$_{s\text{-}cis}$, 1592.99 CO$_{s\text{-}trans}$, 1167.12 CH$_{ip}$, 782.09 CH$_{op}$, 1018.52 CH=CH$_{op}$, 613.05 C=C$_{op}$; ^{1}H NMR (400MHz, CDCl$_3$, ppm) δ = 6.97 (*d*, 1H, Hα, *J* = 16Hz), 7.707 (*d*, 1H, Hβ, *J* = 16Hz), 7.260-8.640

(*m*, 15H, ArH); ^{13}C NMR (100MHz, $CDCl_3$, ppm) δ = 188.51 (CO), 124.68 (Cα), 142.14 (Cβ), 125.11-131.03 (Ar-C).

E **1-(1-Pyrene)-3-(4-methylphenyl)-2-propen-1-one (N7)**: IR, KBr (cm^{-1}): ν = 1654.79 $CO_{s\text{-}cis}$, 1589.31 $CO_{s\text{-}trans}$, 1161.64 CH_{ip}, 66.94 CH_{op}, 1020.91 CH=CH_{op}, 606.86 C=C_{op}; ^{1}H NMR (400MHz, $CDCl_3$, ppm) δ = 7.418 (*d*, 1H, Hα, *J* = 16Hz), 7.606 (*d*, 1H, Hβ, *J* = 16Hz), 7.195-8.603 (*m*, 15H, ArH), 2.380 (*s*, 3H, CH_3); ^{13}C NMR (100MHz, $CDCl_3$, ppm) δ = 196.03 (CO), 124.12 (Cα), 146.34 (Cβ), 124.49-141.36 (Ar-C), 29.37 (CH_3).

E **1-(1- Pyrene)-3-(3-nitrophenyl)-2-propen-1-one (N8)**: IR, KBr (cm^{-1}): ν = 1662.98 $CO_{s\text{-}cis}$, 1529.01 $CO_{s\text{-}trans}$, 1122.05 CH_{ip}, 725.18 CH_{op}, 1074.51 CH=CH_{op}, 559.03 C=C_{op}; ^{1}H NMR (400MHz, $CDCl_3$, ppm) δ = 7.731 (*d*, 1H, Hα, *J* = 16Hz), 8.132 (*d*, 1H, Hβ, *J* = 16Hz), 7.605-8.715 (*m*, 15H, ArH); ^{13}C NMR (100MHz, $CDCl_3$, ppm) δ = 194.65 (CO), 122.68 (Cα), 142.38 (Cβ), 124.14-148.75 (Ar-C).

E **1-(1- Pyrene)-3-(4-nitrophenyl)-2-propen-1-one (N9):** IR, KBr(cm^{-1}): ν = 1658.30 $CO_{s\text{-}cis}$, 1594.55 $CO_{s\text{-}trans}$, 1117.93 CH_{ip}, 778.08 CH_{op}, 1072.71 CH=CH_{op}, 601.06 C=C_{op}; ^{1}H NMR (400MHz, $CDCl_3$, ppm) δ = 7.615 (*d*, 1H, Hα, *J* = 15Hz), 8.245 (*d*, 1H, Hβ, *J* = 15Hz), 7.701-8.710 (*m*, 15H, ArH); ^{13}C NMR (100MHz, $CDCl_3$, ppm) δ = 196.98 (CO), 124.13 (Cα), 142.19 (Cβ), 124.28-135.78 (Ar-C).

Influence of solvents

The effects of solvents on the percentage of the phenyl chalcones have been studied with ethanol, methanol, dichloromethane, toluene and tetrahydrofuran and presented in Table 2 with a statistical diagram as shown in Fig. 2. As can be observed in the Table 2 and the statistical diagram as in Fig. 2, (entries A1-A11) in the water medium H, halogens, 4-OCH_3, 4-CH_3 and 4-NO_2 substituted benzaldehydes gave higher yields than 4-NH_2, 4-F, 4-OH and 4-I substituted benzaldehydes. In ethanol solvent medium, H, 4-OCH_3, 4-CH_3 and 4-NO_2 benzaldehydes gave higher yields of chalcones than other substituents. The substituents in benzaldehydes like H, 4-Br, 4-OCH_3, 4-CH_3, 2-, 3- and 4-NO_2 gave higher yields of chalcones than other substituents in methanol medium. In dichloromethane medium, the aldehydes containing H, 4-Cl, 4-OCH_3, 4-CH_3, 2-, 3- and 4-NO_2 substituents gave higher yields than other substituents. The higher yields of chalcones were obtained for the aldehydes containing H, 4-I, 4-OCH_3, 4-CH_3, 2-, 3- and 4-NO_2 substituents in toluene solvent medium. The H, 4-I, 4-OCH_3, 4-CH_3, 2-, 3- and 4-NO_2 substituted benzaldehydes gave higher yields in tetrahydrofuran solvent medium.

Table 2. The effect of solvents on the synthesis of 4′-X-phenyl chalcones in percentage.

(X = H, Phenyl chalcones, entries A1-A11)

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	68	66	66	66
4-NH_2	60	64	62	63	63	64
4-Br	67	64	65	64	62	60
4-Cl	67	63	66	65	60	63
4-$N(CH_3)_2$	62	60	63	61	60	62
4-F	62	61	64	61	61	60
4-OH	63	62	62	60	60	63
4-I	61	62	65	64	65	66
4-OCH_3	65	67	68	67	67	67
4-CH_3	66	66	67	66	67	67
4-NO_2	68	67	68	67	66	68

(X = NO_2, Phenyl chalcones, entries B1-B12)

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	66	67	66	67
3-Br	65	66	65	66	66	65
4-Br	65	66	64	64	67	66
3-Cl	64	65	63	62	65	66
4-Cl	65	66	66	66	64	66
4-F	63	62	61	60	61	60
2-OCH_3	67	65	60	61	63	63
4-OCH_3	68	67	65	67	66	65
4-CH_3	66	68	66	66	66	67
2-NO_2	63	61	60	62	65	63
3-NO_2	65	66	62	66	65	66
4-NO_2	66	66	65	67	66	65

(HOH=Water; EtOH=Ethanol; MeOH= Methanol; DCM=Dichloromethane;
TE= Toluene; THF=Tetrahydrofuran)

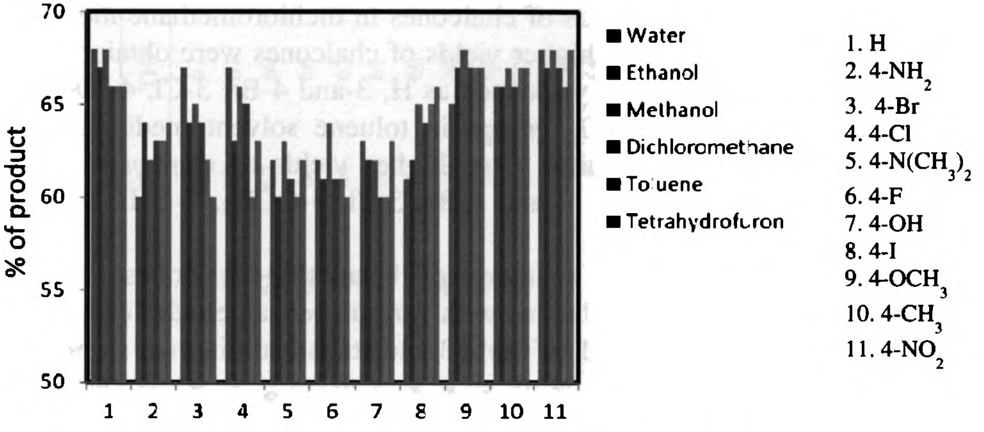


Fig. 2. The effect of solvents on the synthesis of phenyl chalcones (entries A1-A11).

The effect of solvents on synthesis of 4′-nitrophenyl chalcones (entries B1-B12)are presented in Table 2 and the statistical diagram is shown in Fig. 3.

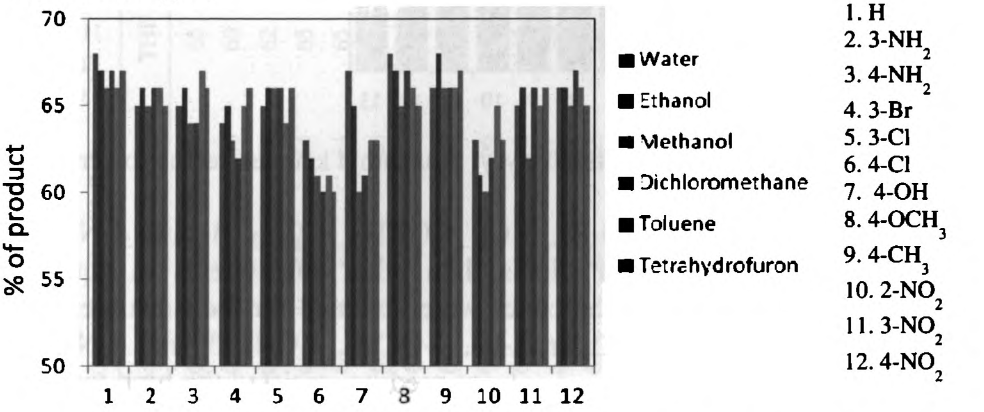


Fig. 3. The effect of solvents on the synthesis of 4′-nitrophenyl chalcones (entries B1-B12).

In aqueous phase, the substituents in benzaldehydes H, halogens, 2-OCH_3, 4-CH_3, 3- and 4-NO_2 gave higher yields than other substituents. In ethanol medium more than 65% of products were obtained except 4-F and 2-NO_2 substituents. Substituents in benzaldehydes such as H, 3-Br, 4-Cl, 2-and 4-OCH_3, 4-CH_3 and 4-NO_2 gave higher yields of chalcones than other substituents in methanol medium. The benzaldehydes containing H, 3-Br, 2-and 4-OCH_3, 4-CH_3, 3- and 4-NO_2

2-, 3- and 4-NO_2 substituted benzaldehydes gave higher yields in tetrahydrofuran solvent medium.

Table 2. The effect of solvents on the synthesis of 4′-X-phenyl chalcones in percentage.

(X = H, Phenyl chalcones, entries A1-A11)

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	68	66	66	66
4-NH_2	60	64	62	63	63	64
4-Br	67	64	65	64	62	60
4-Cl	67	63	66	65	60	63
4-$N(CH_3)_2$	62	60	63	61	60	62
4-F	62	61	64	61	61	60
4-OH	63	62	62	60	60	63
4-I	61	62	65	64	65	66
4-OCH_3	65	67	68	67	67	67
4-CH_3	66	66	67	66	67	67
4-NO_2	68	67	68	67	66	68

(X = NO_2, Phenyl chalcones, entries B1-B12)

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	66	67	66	67
3-Br	65	66	65	66	66	65
4-Br	65	66	64	64	67	66
3-Cl	64	65	63	62	65	66
4-Cl	65	66	66	66	64	66
4-F	63	62	61	60	61	60
2-OCH_3	67	65	60	61	63	63
4-OCH_3	68	67	65	67	66	65
4-CH_3	66	68	66	66	66	67
2-NO_2	63	61	60	62	65	63
3-NO_2	65	66	62	66	65	66
4-NO_2	66	66	65	67	66	65

(HOH=Water; EtOH=Ethanol; MeOH= Methanol; DCM=Dichloromethane; TE= Toluene; THF=Tetrahydrofuran)

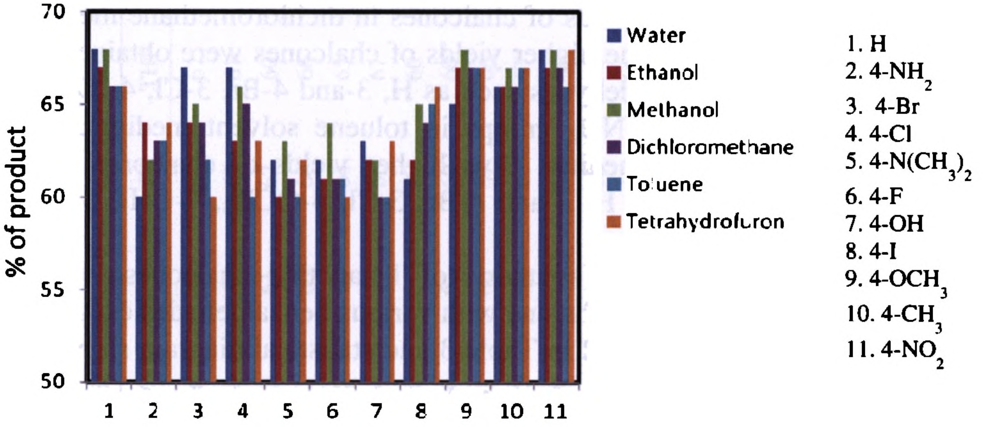


Fig. 2. The effect of solvents on the synthesis of phenyl chalcones (entries A1-A11).

The effect of solvents on synthesis of 4′-nitrophenyl chalcones (entries B1-B12)are presented in Table 2 and the statistical diagram is shown in Fig. 3.

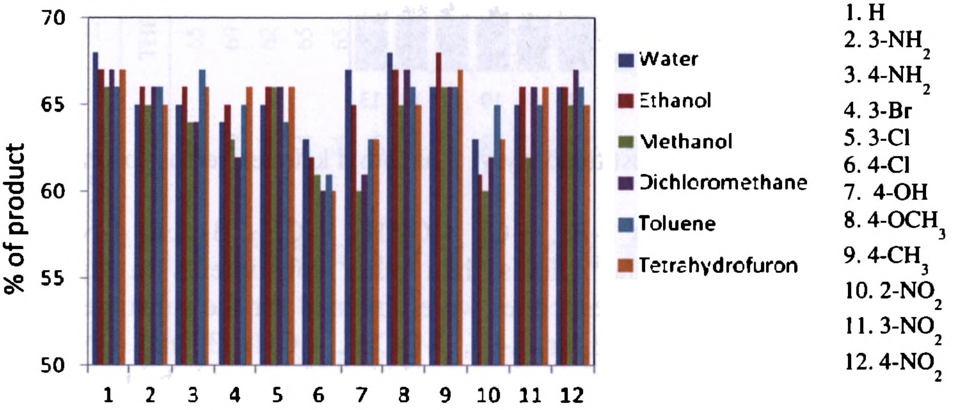


Fig. 3. The effect of solvents on the synthesis of 4′-nitrophenyl chalcones (entries B1-B12).

In aqueous phase, the substituents in benzaldehydes H, halogens, 2-OCH_3, 4-CH_3, 3- and 4-NO_2 gave higher yields than other substituents. In ethanol medium more than 65% of products were obtained except 4-F and 2-NO_2 substituents. Substituents in benzaldehydes such as H, 3-Br, 4-Cl, 2-and 4-OCH_3, 4-CH_3 and 4-NO_2 gave higher yields of chalcones than other substituents in methanol medium. The benzaldehydes containing H, 3-Br, 2-and 4-OCH_3, 4-CH_3, 3- and 4-NO_2

substituents gave higher yields of chalcones in dichloromethane medium than other substituents. The higher yields of chalcones were obtained for the substituents in benzaldehydes such as H, 3-and 4-Br, 3-Cl, 4- OCH_3, 4-CH_3 and 2-, 3- and 4-NO_2 groups in toluene solvent medium. The solvent tetrahydrofuran medium gave higher yields of chalcones from benzaldehydes containing H, 3-and 4-Br, 3-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 substituents.

The solvent effects on synthesis of 1-naphthyl chalcones (entries C1-C13) from 1-acetyl naphthalene with various benzaldehydes catalyzed by fly-ash: water is presented in Table 3 and statistical diagram shown in Fig. 4.

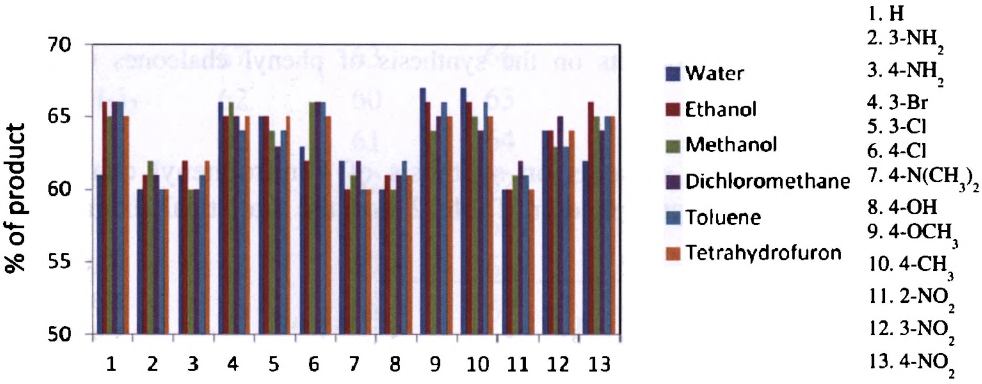


Fig. 4. Percentage of substituted styryl 1-naphthyl ketones versus solvents.

Substituents such as 3-Br, 3-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 in benzaldehydes gave higher yields than other substituents in aqueous medium. Higher yields of chalcones were obtained for the substituents in benzaldehydes such as H, 3-Br, 4-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 in ethanol medium. The methanol solvent medium assisted the high yield of chalcones with aldehydes containing H, 3-Br, 4-Cl, 4-CH_3 and 4-NO_2 substituents. Dichloromethane medium is favorable for obtaining high yields of chalcones with benzaldehydes containing H, 3-Br, 4-Cl, 4-OCH_3, 3- and 4-NO_2 substituents. The substituents H, 4-Cl, 4-OCH_3, 4-CH_3 and 4-NO_2 in benzaldehydes gave high yields of chalcones in tetrahydrofuran solvent medium.

The effect of solvent on synthesis of 4-bromo-1-naphthyl chalcones (entries D1-D13) is presented in Table 3 and the statistical diagram is shown in Fig. 5.

Table 3. The effect of solvents on the synthesis of 4-X-1-naphthyl and 6-X-2-naphthyl chalcones in percentage.

	(X = 4-H, 1-naphthyl chalcones, entries C1-C13)						(X = 4-Br, 4-Bromo-1-naphthyl chalcones, entries D1-D13)					
Subst.	HOH	EtOH	MeOH	DCM	TE	THF	HOH	EtOH	MeOH	DCM	TE	THF
H	61	66	65	66	66	65	60	67	66	68	67	67
3-NH_2	60	61	62	61	60	60	61	60	60	62	60	61
4-NH_2	61	62	60	60	61	62	62	61	61	61	62	60
3-Br	66	65	66	65	64	65	64	65	63	65	66	66
3-Cl	65	65	64	63	64	65	65	66	65	65	64	65
4-Cl	63	62	66	66	66	65	65	65	64	66	66	67
4-$N(CH_3)_2$	62	60	61	62	60	60	64	62	60	61	60	60
4-OH	60	61	60	61	62	61	62	61	60	62	60	60
4-OCH_3	67	66	64	65	66	65	66	65	63	66	67	66
4-CH_3	67	66	65	64	66	65	67	66	65	68	66	67
2-NO_2	60	60	61	62	61	60	61	60	60	61	60	60
3-NO_2	64	64	63	65	63	64	63	64	63	64	63	65
4-NO_2	62	66	65	64	65	65	66	65	65	66	65	66

	(X = 4-Cl, 4-Chloro-1-naphthyl chalcones, entries E1-E13)						(X = 4-OCH_3, 4-Methoxy-1-naphthyl chalcones, entries F1-F13)					
Subst.	HOH	EtOH	MeOH	DCM	TE	THF	HOH	EtOH	MeOH	DCM	TE	THF
H	65	68	67	68	68	67	65	67	68	68	68	68
3-NH_2	62	60	61	60	60	60	61	60	62	61	62	60
4-NH_2	62	60	62	60	60	60	61	60	60	60	60	60
3-Br	66	64	65	63	64	63	65	64	65	63	62	66
3-Cl	65	63	64	65	65	64	66	65	67	64	65	63

cont. Table 3.

	(X = 4-Cl, 4-Chloro-1-naphthyl chalcones, entries E1-E13)						(X = 4-OCH$_3$, 4-Methoxy-1-naphthyl chalcones, entries F1-F13)					
Subst.	HOH	EtOH	MeOH	DCM	TE	THF	HOH	EtOH	MeOH	DCM	TE	THF
4-Cl	64	66	66	65	66	65	68	67	66	68	65	66
4-N(CH$_3$)$_2$	63	60	61	60	61	61	60	60	62	61	62	60
4-OH	62	60	61	62	60	60	60	61	60	62	60	61
4-OCH$_3$	66	61	64	66	68	66	67	66	67	68	68	67
4-CH$_3$	67	67	66	67	68	67	66	65	66	68	67	66
2-NO$_2$	61	60	61	63	60	61	62	60	61	63	62	60
3-NO$_2$	63	65	63	66	65	64	65	64	66	65	63	66
4-NO$_2$	66	66	67	67	67	67	68	68	67	66	65	68
	(X = 4-CH$_3$, 4-Methyl-1-naphthyl chalcones, entries G1-G13)						(X = 6-H, 2-naphthyl chalcones, entries H1-H13)					
Subst.	HOH	EtOH	MeOH	DCM	TE	THF	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	68	68	67	67	60	61	62	60	64	62
3-NH$_2$	62	61	60	63	62	60	60	60	61	61	60	61
4-NH$_2$	61	60	61	60	60	63	64	64	65	66	65	65
3-Br	66	65	63	64	66	66	65	66	64	64	66	64
3-Cl	65	66	65	66	64	63	66	65	66	65	63	66
4-Cl	66	65	66	66	65	66	61	60	60	60	60	60
4-N(CH$_3$)$_2$	61	60	61	62	60	61	60	60	60	61	60	60
4-OH	60	62	60	60	62	60	67	66	67	65	68	61
4-OCH$_3$	68	67	67	66	63	65	66	66	65	64	66	66
4-CH$_3$	67	66	68	67	67	66	60	60	62	63	60	61

cont. Table 3.

	(X = 4-CH_3, 4-Methyl-1-naphthyl chalcones, entries G1-G13)						(X = 6-H, 2-naphthyl chalcones, entries H1-H13)					
Subst.	HOH	EtOH	MeOH	DCM	TE	THF	HOH	EtOH	MeOH	DCM	TE	THF
2-NO_2	60	60	61	62	63	60	64	65	66	65	64	65
3-NO_2	65	66	64	65	64	65	66	65	67	67	66	68
4-NO_2	68	67	66	68	67	65	60	61	62	60	64	62

	(X = 6-OCH_3, 6-Methoxy-2-naphthyl chalcones, entries I1-I13)						(X = 6-CH_3, 6-Methyl-2-naphthyl chalcones, entries J1-J13)					
Subst.	HOH	EtOH	MeOH	DCM	TE	THF	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	68	68	68	68	67	68	67	68	68	68
3-NH_2	63	61	60	62	63	60	60	60	60	61	62	61
4-NH_2	64	63	62	60	61	62	60	61	62	61	60	60
3-Br	63	64	63	62	65	64	65	65	64	63	65	64
3-Cl	61	64	62	64	64	66	65	66	65	64	65	63
4-Cl	64	65	64	63	66	65	66	66	67	65	66	64
4-$N(CH_3)_2$	60	61	60	60	60	60	60	61	60	60	60	60
4-OH	60	60	61	61	61	60	60	62	60	61	60	61
4-OCH_3	64	66	67	68	68	66	67	67	66	67	68	68
4-CH_3	65	63	66	67	65	66	68	67	65	66	67	67
2-NO_2	61	60	61	62	60	60	60	61	60	62	61	60
3-NO_2	64	64	66	65	67	66	62	62	62	63	60	62
4-NO_2	65	67	68	66	67	68	62	66	65	67	67	68

(HOH = Water; EtOH = Ethanol; Mech = Methanol; DCM = Dichloromethane; TE = Toluene; THF = Tetrahydrofuran).

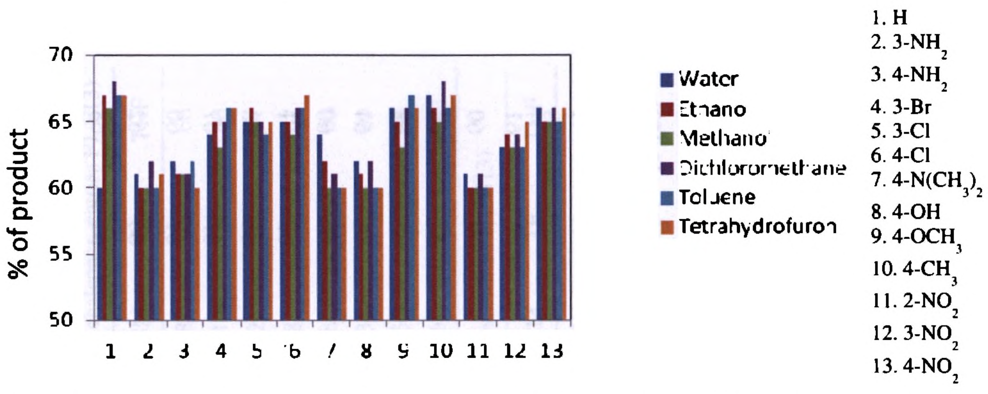


Fig. 5. Percentage of substituted styryl 4-bromo-1-naphthyl ketones versus solvents.

Substituents 4-Cl, 4-OCH_3, 4-CH_3 and 4-NO_2 in benzaldehydes gave high yields of chalcones in aqueous phase reaction. The ethanol medium is favored for obtaining high yields of products for H, halogens, 4-OCH_3, 4-CH_3 and 4-NO_2 substituted benzaldehydes than other substituted benzaldehydes. Benzaldehydes containing H, 3-Cl, 4-CH_3 and 4-NO_2 substituents gave higher yields of products in methanol medium than other substituted benzaldehydes. In dichloromethane solvent medium H, halogens, 4-OCH_3, 4-CH_3 and 4-NO_2 benzaldehydes gave higher yield percentage of chalcones than other substituted benzaldehydes. High yield of chalcones obtained for the substituents in benzaldehydes H, 3-Br, 4-Cl, 4-OCH_3, 4-CH_3 and 4-NO_2 in toluene solvent medium. Tetrahydrofuran solvent medium is favorable for obtaining high yield of chalcones, benzaldehydes containing H, 3-and 4-Cl, 4-CH_3, 3- and 4-NO_2 substituents than others.

Table 3 and Fig. 6 show the effect of solvent on synthesis of chalcones from 4-chloro-1-naphthyl methyl ketones and substituted benzaldehydes. Aqueous medium is favorable for obtaining higher yield of chalcones from benzaldehydes containing H, 3-Br, 3-Cl, 4-OCH_3, 4-CH_3 and 4-NO_2 substituents. The substituents H, 4-Cl, 4-CH_3, 3-and 4-NO_2 in benzaldehydes gave high yields percentage of products in ethanol medium. In methanol solvent the percentage of products obtained from the benzaldehydes with H, 3-Br, 4-Cl, 4-CH_3 and 4-NO_2 substituents is higher than other. Dichloromethane gave higher yield of product from H, 3- and 4-Cl, 4-OCH_3, 4-CH_3, 3-and 4-NO_2 substituted benzaldehydes than other substituted aldehydes. Toluene solvent medium is favored for obtaining high yields of chalcones for benzaldehydes with H, 3- and 4-Cl,4-OCH_3, 4-CH_3, 3- and 4-NO_2 substituents. The same trend have been observed for obtaining high yield of products in tetrahydrofuran solvent medium.

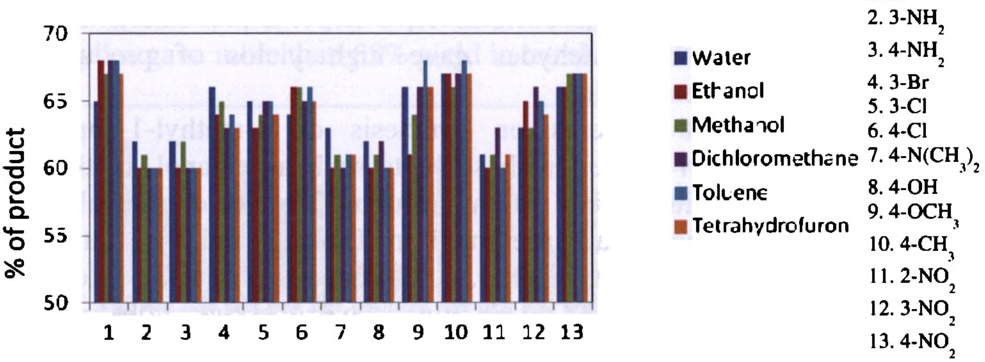


Fig. 6. Percentage of substituted styryl 4-chloro-1-naphthyl ketones versus solvents.

The influence of solvents on synthesis of 4-methoxy-1-naphthyl chalcones was studied with ethanol, methanol, dichloromethane, toluene and tetrahydrofuran and presented in Table 3 and the statistical diagram are shown in Fig. 7.

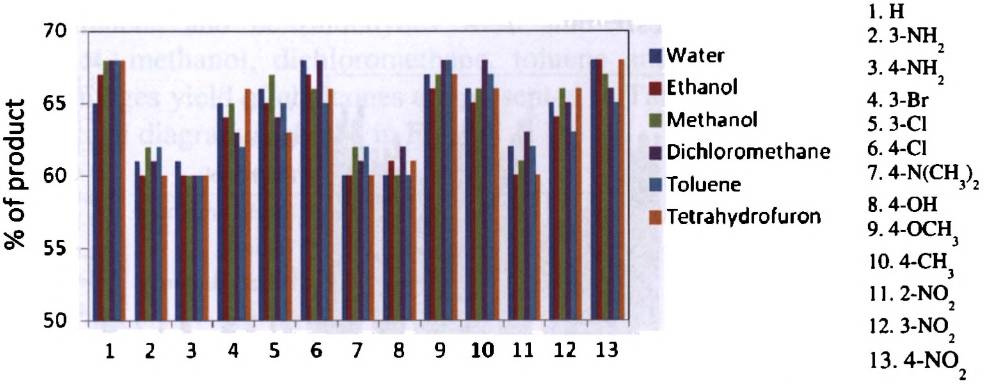


Fig. 7. Percentage of substituted styryl 4-methoxy-1-naphthyl ketones versus solvents.

Benzaldehydes with H, 3-Br, 3- and 4-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 substituents gave high yield of products in aqueous phase. The substituents H, 3- and 4-Cl, 4-OCH_3, 4-CH_3, and 4-NO_2 in benzaldehydes yields high percentage yield of products in ethanol medium. Higher yield of products obtained in methanol medium for the substituents H, halogens, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 in benzaldehydes. Dichloromethane is favorable for obtaining high yield of chalcones from the benzaldehydes with H, 4-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2

substituents. High yield of chalcones was produced by the benzaldehydes containing H, 3- and 4-Cl, 4-OCH$_3$, 4-CH$_3$, and 4-NO$_2$ substituents in toluene solvent. Substituents such as H, 3-Br, 4-Cl, 4-OCH$_3$, 4-CH$_3$, 3- and 4-NO$_2$ in benzaldehydes gave high yields of product in tetrahydrofuran solvent media.

The effects of solvents on synthesis of 4-methyl-1-naphthyl chalcones have been studied with solvents such as ethanol, methanol, dichloromethane, toluene and tetrahydrofuran. The yield of chalcones with respect to solvents are presented in Table 3 and the statistical correlation diagram is shown in Fig. 8. In aqueous phase, H, halogens, 4-OCH$_3$, 4-CH$_3$, 3- and 4-NO$_2$ substituted chalcones were obtained at higher percentage. The similar trend was observed on the effects of ethanol solvent in this reaction. The solvent methanol is favorable for obtaining high percentage yield of H, 3- and 4-Cl, 4-OCH$_3$, 4-CH$_3$, and 4-NO$_2$ substituted chalcones. The same trend was observed in dichloromethane solvent medium with 3-NO$_2$ substituent. Toluene medium gave high yield of chalcones with H, 4-Br, 4-Cl, 4-CH$_3$ and 4-NO$_2$ substituents in aldehydes. The same trend was observed with the effect of tetrahydrofuran solvent including 4-OCH$_3$ and 3-NO$_2$ substituents in aldehydes.

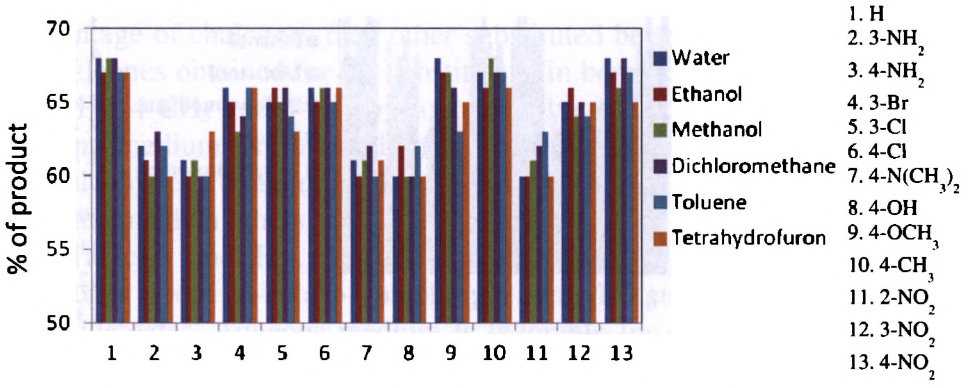


Fig. 8. Percentage of substituted styryl 4-methyl-1-naphthyl ketones versus solvents.

The influence of solvents on the synthesis of 2-naphthyl chalcones is presented in Table 3 and the statistical correlation diagram is shown in Fig. 9. In aqueous phase and ethanol medium, reaction gave more than 65% yield for the substituents H, 3-and 4-Cl, 4-OCH$_3$, 4-CH$_3$, 3- and 4-NO$_2$ in the benzaldehydes. The benzaldehydes containing the substituents H, 3-Br, 4-Cl, 4-OCH$_3$, 4-CH$_3$, 3- and 4-NO$_2$ gave higher yield of chalcones in methanol and dichloromethane media. In toluene medium, the aldehydes containing H, 3-Br, 3-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 substituents gave the high percentage yield of products. Substituents such as H, 3-Br, 4-Cl, 4-CH_3, 3- and 4-NO_2 present in aldehydes gave more than 65% yield in tetrahydrofuran medium.

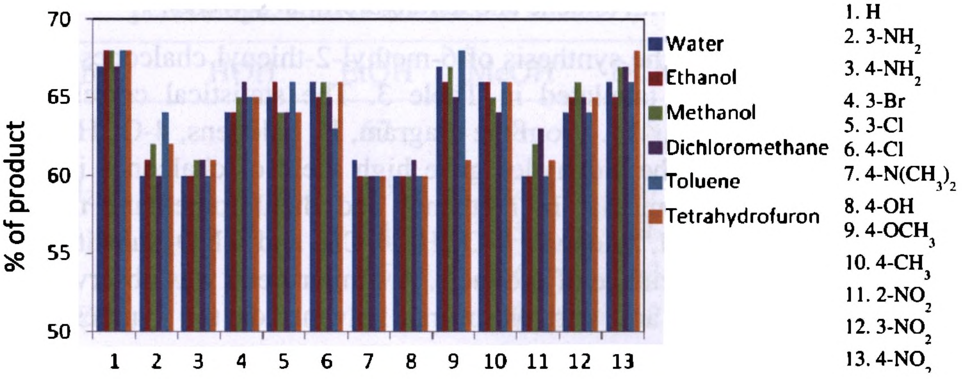


Fig. 9. Percentage of substituted styryl 2-naphthyl ketones versus solvents.

The solvent effects on the synthesis of 6-methoxy-2-naphthyl chalcones by aqueous phase aldol condensation of 6-methoxy-2-acetyl naphthalene and benzaldehydes were studied with solvents such as ethanol, methanol, dichloromethane, toluene and tetrahydrofuran. The percentages yield of chalcones are presented in Table 3 and the correlated statistical diagram is shown in Fig. 10.

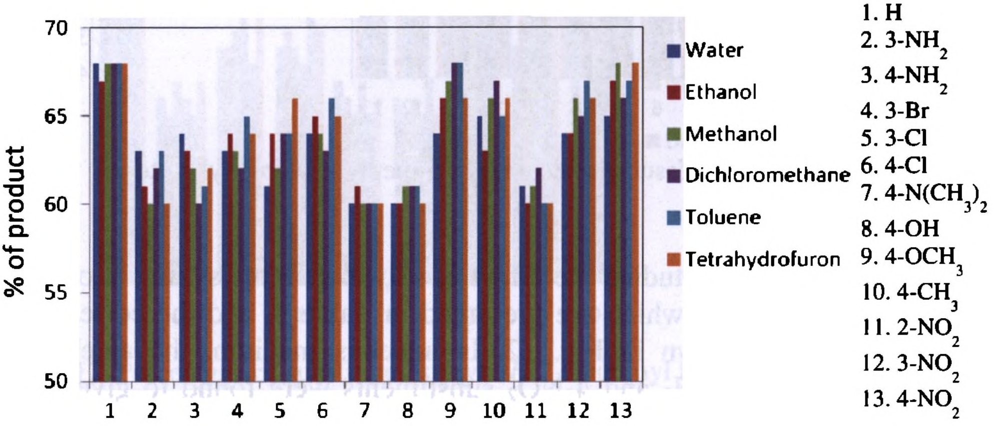


Fig. 10. Percentage of substituted styryl 6-methoxy-2-naphthyl ketones versus solvents.

Good percentage yields of chalcones were obtained for the aldehyde moiety containing H, 4-Cl, 4-OCH_3, 4-CH_3 and 4-NO_2 substituents in aqueous and ethanol medium. In methanol and dichloromethane medium, the aldehydes containing H, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 substituents gave high yields of chalcones. High percentage yield of products are obtained with H, 3-Br, 3- and 4-Cl, 4-OCH_3, 4-CH_3, 3- and 4-NO_2 substituents in benzaldehydes moiety with toluene and tetrahydrofuran solvents.

Solvent effect on the synthesis of 6-methyl-2-thienyl chalcones have been studied and are tabulated in Table 3. The statistical correlation diagram is shown in Fig.11. From the diagram, H, halogens, 4-OCH_3 and 4-CH_3substituents in the aldehydes gave high yield of chalcones in the aqueous and ethanol media. In methanol and dichloromethane media aldehydes containing H, halogens, 4-OCH_3, 4-CH_3 and 4-NO_2 substituents gave higher yield percentage of products. A similar trend was observed in the effects of toluene and tetrahydrofuran solvents on the synthesis of chalcones.

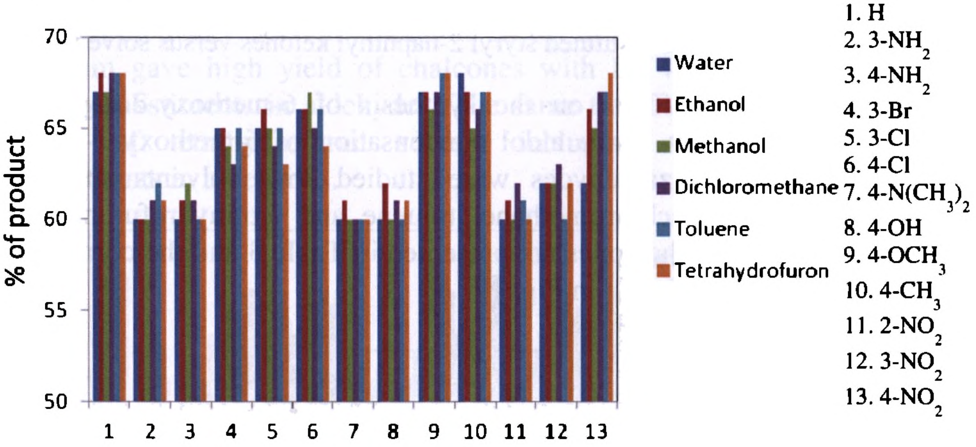


Fig. 11. Percentage of substituted styryl 6-methyl-2-naphthyl ketones versus solvents.

The author has studied the effect of solvents on the synthesis of 9*H*-flyorenyl chalcones, which are presented in Table 4, and the correlation bar diagram is shown in Fig. 12. In aqueous medium, the aldehydes containing H, 4-OCH_3 and 4-NO_2 substituents were found to give high yields of chalcones. Ethanol medium is favored for obtaining high yield of products, benzaldehydes containing H, 4-Cl, 4-OCH_3 and 4-NO_2 substituents. Benzaldehydes with H, 3-Cl, 4-OCH_3 and 4-NO_2 substituents were found to be good for higher yields of chalcones in methanol and dichloromethane solvent media. Toluene and tetrahydrofuran solvents are favorable for obtaining high yields of chalcones with H, 3- and 4-Cl, 4-OCH_3 and 4-NO_2 substituents in benzaldehydes.

Table 4. The solvent effects on synthesis of 9*H*-2-fluorenyl chalcones in percentage, entries K1-K9.

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	66	67	68	68	68	68
3-NH_2	61	60	62	61	60	62
3-Cl	64	64	65	66	63	66
4-Cl	64	65	64	63	66	65
4-$N(CH_3)_2$	62	61	60	62	60	61
4-OH	61	60	61	60	62	60
4-OCH_3	65	68	68	68	67	68
4-CH_3	64	65	64	66	65	66
4-NO_2	68	60	67	67	66	67

(HOH = Water; EtOH = Ethanol; MeOH = Methanol; DCM = Dichloromethane; TE = Toluene; THF = Tetrahydrofuran)

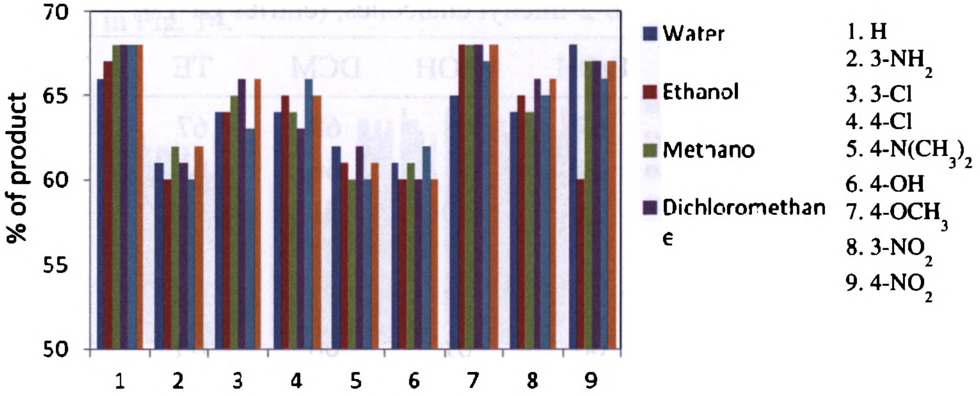


Fig. 12. Percentage of substituted styryl 9*H*-2-fluorenyl ketones versus solvents.

The percentage yield of products obtained from the aldol condensation of 5-bromo-2-thienyl and benzaldehydes with various solvents are presented in Table 5 and the correlation chart is shown in Fig. 13. The substituents H, 4-Br, 4-Cl, 4-OCH_3 and 4- CH_3 with the benzaldehydes gave higher yields of products in water, ethanol, methanol, dichloromethane, toluene and tetrahydrofuran solvents. The 2-chlorobenzaldehydes gave high yields in tetrahydrofuran solvent only. Similarly, 4-I benzaldehydes gave high yield of chalcones only in ethanol and dichloromethane solvents.

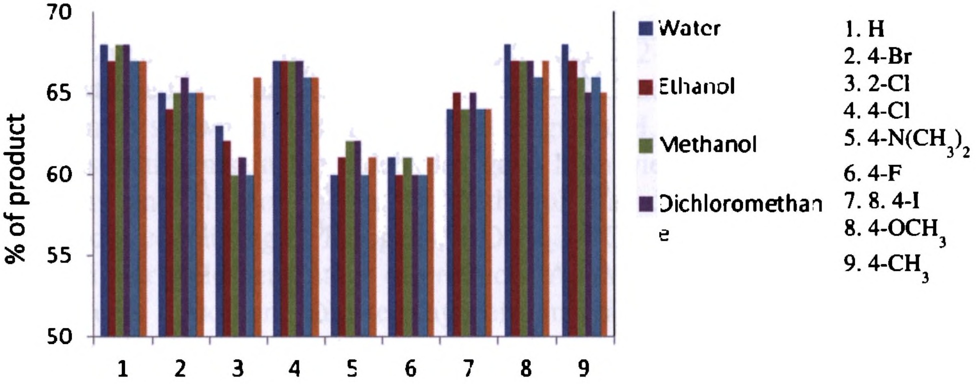


Fig. 13. Percentage of substituted styryl 5-bromo-2-thienyl ketones versus solvents.

Table 5. The solvent effects on synthesis of 5-X-2-thienyl chalcones in percentage

(X = Br, 5-Bromo-2-thienyl chalcones, (entries L1-L9)						
Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	68	67	68	68	67	67
4-Br	65	64	65	66	65	65
2-Cl	63	62	60	61	60	66
4-Cl	67	67	67	67	66	66
4-N(CH$_3$)$_2$	60	61	62	62	60	61
4-F	61	60	61	60	60	61
4-I	64	65	64	65	64	64
4-OCH$_3$	68	67	67	67	66	67
4-CH$_3$	68	67	66	65	66	65
(X = Cl, 5-Chloro-2-thienyl chalcones, entries M1-M12)						
H	68	67	67	66	65	68
3-Br	66	65	64	65	65	64

cont. Table 5.

(X = Cl, 5-Chloro-2-thienyl chalcones, entries M1-M12)

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
4-Br	65	63	65	64	63	66
3-Cl	63	62	60	61	60	60
4-Cl	62	63	64	60	61	60
4-F	60	60	61	61	60	60
2-OCH_3	67	67	66	67	66	67
4-OCH_3	68	67	67	66	65	66
4-CH_3	68	68	67	66	65	68
2-NO_2	67	67	67	66	63	66
3-NO_2	66	67	66	65	66	68
4-NO_2	67	64	64	64	64	67

(HOH = Water; EtOH = Ethanol; Mech = Methanol; DCM = Dichloromethane; TE = Toluene; THF = Tetrahydrofuran).

The influence of solvent on the synthesis of 5-chloro-2-thienyl ketones are presented in Table 5 and the statistical correlation diagram is shown in Fig. 14.

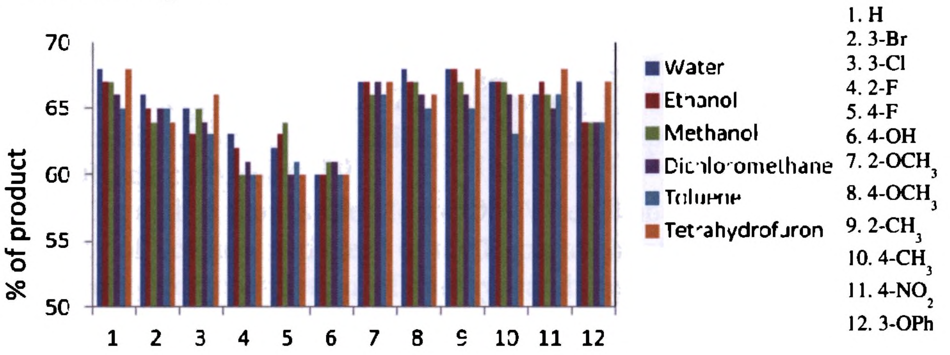


Fig. 14. Percentage of substituted styryl 5-chloro-2-thienyl ketones versus solvents.

In aqueous medium, more than 65% yield of chalcones obtained for the aldehydes containing H, 3-Br, 3-Cl, 2- and 4-OCH_3, 2- and 4-CH_3, 4-NO_2 and 3-OC_6H_5 as substituents. Aldehydes containing H, 3-Br, 4-OCH_3, 2- and 4-CH_3, 4-NO_2 substituents gave high yield of chalcones in ethanol medium. In methanol medium, the substituents H, 3-Cl, 4-OCH_3, 2- and 4-CH_3, 4-NO_2 with the benzaldehydes gave higher percentage yield of chalcones.

The same trend was observed for the yields of chalcones in dichloromethane and toluene solvents. Tetrahydrofuran was favorable for obtaining high yield of chalcones with H, 4-OCH_3, 2- and 4-CH_3, 4-NO_2 and 3-OC_6H_5 substituents in aldehyde moieties.

The effect of solvents on the synthesis of substituted styryl 1-pyrenyl ketones have been studied with various solvents in the fly-ash catalyzed aqueous phase aldol condensation. The percentage yield of the obtained ketones is presented in Table 6 and the correlation chart is shown in Fig. 15. As can be observed in the the chart, more than 65% of chalcones yield was obtained for the benzaldehydes with the substituents H, 3-Br, 3- and 4-Cl, 4-CH_3, 3- and 4-NO_2 in aqueous, ethanol, methanol and dichloromethane solvents. Good yields of chalcones were obtained with substituted benzaldehydes except 2, 3 and 4-Cl and 4-F substituents in toluene medium. In tetrahydrofuran medium, the benzaldehydes containing the substituents H, 3-Cl , 4- CH_3 and 3- and 4-NO_2 gave higher yields of chalcones.

Table 6. The solvent effects on the synthesis of 1-pyrenyl chalcones, entries N1-N10.

Subst.	HOH	EtOH	MeOH	DCM	TE	THF
H	67	67	68	68	68	68
3-Br	64	63	65	66	65	64
2-Cl	62	63	61	64	63	61
3-Cl	65	65	66	65	64	65
4-Cl	66	66	67	65	64	64
4-F	61	60	62	61	62	60
4-CH_3	67	68	67	67	66	68
3-NO_2	65	65	66	67	66	66
4-NO_2	67	66	68	67	68	67

(HOH = Water; EtOH = Ethanol; Mech = Methanol; DCM = Dichloromethane; TE = Toluene; THF=Tetrahydrofuran).

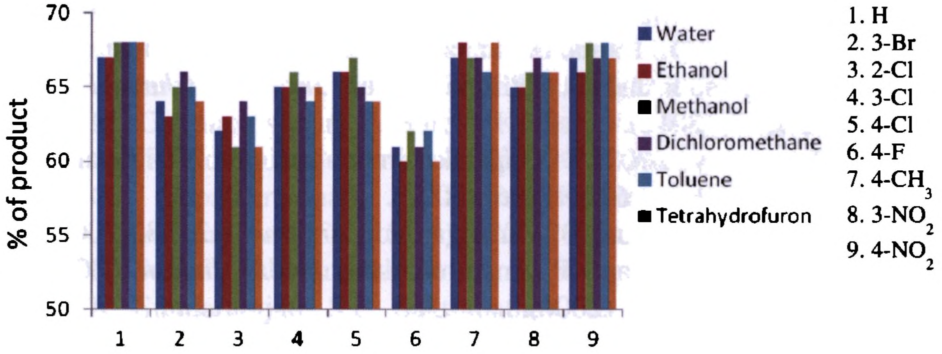


Fig. 15. Percentage of substituted styryl 1-pyrenyl ketones verses solvents.

4. CONCLUSIONS

The author has synthesized good yield of some series of aryl chalcones in aqueous phase crossed aldol condensation between aryl methyl ketone and substituted benzaldehydes catalyzed by fly-ash: H$_2$O. The synthesized chalcones were characterized by their physical constants and spectral data. The effects of solvents on the synthesis of chalcones have been studied with ethanol, methanol, dichloromethane, toluene and tetrahydrofuran solvents.

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