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Microwave-Assisted Michael Addition of Sulfonamides to α,β-Unsaturated Esters: A Rapid Entry to Protected β-Amino Acid Synthesis

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An efficient, clean and very simple procedure for the synthesis of protected β-amino acids is described. Michael addition of sulfonamides to α,β-unsaturated esters in the presence of K_2CO_3 and tetrabutylammonium bromide (TBAB) under microwave irradiation affords the title compounds in good to high yields and short reaction times. This new method affords protected β-amino acids in high yields and short reaction times.

Keywords: Microwave, Michael addition, Sulfonamide, α,β-Unsaturated ester, β-Amino acid

INTRODUCTION

Microwave-assisted organic reactions have been applied as an effective technique in organic synthesis [1]. Microwave irradiation often leads to shorter reaction times, increased yields, easier workup, matches with green chemistry protocols, and can enhance the regio- and stereoselectivity of reactions [1]. Furthermore, its unique capabilities allow its application in reactions which are difficult or impossible to carry out by means of customary conventional methods [1]. In fact, the high usefulness of microwave-assisted synthesis encouraged us to increase the efficiency of several organic transformations and synthesis [2].

Aza-Michael addition of sulfonamides to α,β-unsaturated esters is significant as this reaction provides a direct and appealing route to protected β-amino acid synthesis. This class of compounds (β-amino acids) are essential components of many drugs and bioactive compounds such as β-peptides [3a], vitamin B_3 [3b], imeriamine (hypoglycemic and antiketogenic agent) [3c], cryptophycin (antitumor) [3d], and TAN-1057 A (antibiotic) [3e]. Moreover, *N*-protected β-amino acids have been applied in the synthesis of β-lactams [4a], 6-member heterocycles [4b] as well as diazepine [4c].

Different reagents and catalysts have been used for aza-conjugate addition of amines [5], amides [6] and imides [7] to α,β-unsaturated compounds, such as $ZrOCl_2.8H_2O$ [5a], aluminium dodecyl sulfate trihydrate [5b], micellar solution of sodium dodecyl sulfate [5c], polyacrylamide supported phenolate [5d], $LiClO_4$ [5e], SmI_2 [5f], kaolinetic clay [5g], β-cyclodextrin [5h], boric acid [5i], NaOH [6a], *t*-BuOK [6b], $Si(OEt)_4$-CsF [6c], 1,4-diazabicyclo[2,2,2]octane (DABCO) [2a], Na in absolute EtOH [7a] and $AlMe_2Cl$ [7b]. However, to the best of our knowledge, aza-Michael addition of sulfonamides to α,β-unsaturated compounds is scarce in literature. Reitz *et al*. have used alumina to achieve this

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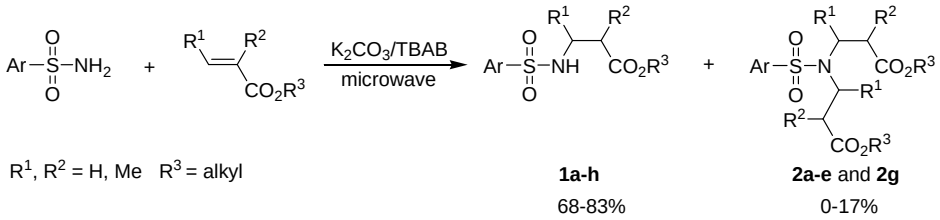

Scheme 1

reaction [8]. The methods established for Michael reaction of nitrogen compounds are usually associated with one or more of the following drawbacks: (i) formation of side products; (ii) using large excess of reagent; (iii) low yield and (iv) long reaction time.

Having the above objects in mind and also in extension of our previous study on application of microwave technique in Michael reaction published in this journal [2a], we now describe a new method for Michael addition of sulfonamides to various α,β-unsaturated esters in the presence of K_2CO_3 and TBAB under microwave irradiation (Scheme 1). The advantages of this method are high efficiency, high yield, short reaction time, low cast and compliance with green chemistry protocols.

EXPERIMENTAL

All chemicals were obtained from Merck or Fluka Chemical Companies. Some α,β-unsaturated esters were prepared from the corresponding acid chlorides by the reported method [9] and their structures were confirmed by IR and ^{1}H NMR spectra. The progress of the reactions was followed by TLC using silica gel SILG/UV 254 plates. IR spectra were run on a Shimadzu FTIR-8300 spectrophotometer. The ^{1}HNMR (250MHz) and ^{13}CNMR (62.5 MHz) were run on a Bruker Avanced DPX-250, FT-NMR spectrometer (δ in ppm, *J* in Hz). Mass spectra were recorded on a Shimadzu GC MS-QP 1000 EX apparatus. Melting points were recorded on a Büchi B-545 apparatus in open capillary tubes and are uncorrected. MB 245 domestic microwave oven from Butan Industrial Company was used for microwave irradiation.

General Procedure for Michael Addition of Sulfonamides to α,β-Unsaturated Esters Under Microwave Irradiation

To a well ground mixture of sulfonamide (2 mmol), K_2CO_3 (0.28 g, 2 mmol) and TBAB (0.32 g, 1 mmol) in a test tube was added α,β-unsaturated ester (2.4 mmol) and mixed thoroughly with a glass rod. The resulting mixture was irradiated in a microwave oven at 300 W for several one minute intervals (Table 4). Then, the reaction mixture was cooled to room temperature and suspended in chloroform (50 ml), filtered and the filtrate was washed with water (2 × 20 ml) and dried with $MgSO_4$. The solvent was evaporated and the crude product was purified by column chromatography on silica gel with EtOAc/*n*-hexane (1/3).

Physical and Spectroscopic Data of Isolated Products

(1a). Colorless oil; IR (neat): 3286, 3059, 2975, 1732, 1447, 1329 cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 1.16 (t, *J* = 7.0 Hz, 3H, *CH*$_3$), 2.48 (t, *J* = 5.0 Hz, 2H, O=C*CH*$_2$), 3.15 (t, *J* = 5.0 Hz, 2H, NH*CH*$_2$), 4.02 (q, *J* = 7.0 Hz, 2H, O*CH*$_2$), 5.56 (br, 1H, *NH*), 7.42-7.52 (complex, 3H), 7.80 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 14.0, 34.15, 38.7, 60.7, 127.0, 128.7, 132.6, 138.8, 171.7; MS m/z (%): 257 (M^+, 22.7).

(2a). Colorless oil; IR (neat): 3028, 2984, 1733, 1447, 1317 cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 1.22 (t, *J* = 7.1 Hz, 6H, 2*CH*$_3$), 2.59 (t, *J* = 5.0 Hz, 4H, 2O=C*CH*$_2$), 3.45 (t, *J* = 5.0 Hz, 4H, 2NH*CH*$_2$), 4.08 (q, *J* = 7.1 Hz, 4H, 2O*CH*$_2$), 7.46-7.57 (complex, 3H), 7.87 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 13.9, 32.8, 44.7, 60.4, 127.2, 129.1, 132.6, 138.9, 170.9; MS m/z (%): 357 (M^+, 31.0).

(1b). Colorless oil; IR (neat): 3271, 3048, 2960, 1733, 1447, 1330 cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 0.90 (t, *J* = 6.5 Hz, 3H, *CH*$_3$), 1.34 (m, 2H, CH_3*CH*$_2$), 1.56 (m, 2H, CH_3CH_2*CH*$_2$), 2.51 (t, *J* = 5.0 Hz, 2H, O=C*CH*$_2$), 3.19 (t, *J* = 5.0 Hz, 2H, NH*CH*$_2$), 4.03 (t, *J* = 7.0 Hz, 2H, O*CH*$_2$), 5.68 (br, 1H, *NH*), 7.48-7.57 (complex, 3H), 7.79 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 13.5, 18.9, 30.4, 34.1, 38.7, 64.6, 126.8, 129.0, 132.5, 139.9, 172.7; MS m/z (%): 285 (M^+, 24.8).

(2b). Colorless oil; IR (neat): 3036, 2966, 1732, 1447, 1317 cm^{-1}; ^1H NMR ($CDCl_3$): δ 0.92 (t, *J* = 6.5 Hz, 6H, 2*CH*$_3$), 1.33 (m, 4H, 2CH_3*CH*$_2$),1.56 (m, 4H, 2CH_3CH_2*CH*$_2$), 2.63 (t, *J* = 5.1 Hz, 4H, 2O=C*CH*$_2$), 3.45 (t, *J* = 5.1 Hz, 4H, 2NH*CH*$_2$), 4.06 (t, *J* = 6.9 Hz, 4H, 2O*CH*$_2$), 7.51-7.60 (complex, 3H), 7.82 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 13.5, 18.9, 30.5, 34.2, 44.4, 64.2, 126.9, 129.0, 132.4, 138.9, 171.1; MS m/z (%): 413 (M^+, 17.6).

(1c). Pale yellow oil; IR (neat): 3286, 3031, 2954, 1733, 1447, 1328 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.65 (t, *J* = 5.1 Hz, 2H, O=C*CH*$_2$), 3.17 (t, *J* = 5.1 Hz, 2H, NH*CH*$_2$), 5.00 (s, 2H, O*CH*$_2$), 5.71 (br, 1H, *NH*), 7.26-7.31 (complex, 5H), 7.45-7.51 (complex, 3H), 7.82 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 34.2, 38.7, 66.6, 126.9, 128.2, 128.4, 128.6, 129.2, 132.7, 135.4, 139.9, 171.7; MS m/z (%): 319 (M^+, 41.7).

(2c). Pale yellow oil; IR (neat): 3051, 2965, 1734, 1447, 1316 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.58 (t, *J* = 5.0 Hz, 4H, 2O=C*CH*$_2$), 3.36 (t, *J* = 5.0 Hz, 4H, 2NH*CH*$_2$), 5.00 (s, 4H, 2O*CH*$_2$), 7.22-7.25 (complex, 10H), 7.39-7.48 (complex, 3H), 7.72 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 33.3, 43.9, 65.5, 126.2, 127.7, 128.1, 128.9, 129.5, 131.8, 134.4, 137.8, 171.4; MS m/z (%): 404 (M^+-C_6H_5, 3.7), 340 (M^+-$C_6H_5SO_2$, 18.3).

(1d). Pale yellow oil; IR (neat): 3287, 3059, 2964, 1732, 1447, 1329 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.55 (t, *J* = 5.2 Hz, 2H, O=C*CH*$_2$), 3.19 (t, *J* = 5.2 Hz, 2H, NH*CH*$_2$), 4.69 (m, 2H, O*CH*$_2$), 5.50 (br, 1H, *NH*), 6.23 (m, 1H, PhCH=*CH*), 6.63 (d, *J* = 15.7 Hz, 1H, Ph*CH*), 7.29-7.35 (complex, 5H), 7.45-7.50 (complex, 3H), 7.95 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 34.2, 38.8, 65.8, 122.6, 122.9, 126.6, 126.9, 128.1, 128.6, 129.0, 132.7, 134.6, 139.9, 171.7; MS m/z (%): 345 (M^+, 26.5).

(2d). Pale yellow oil; IR (neat): 3026, 2981, 1732, 1448, 1317 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.51 (t, *J* = 5.1 Hz, 4H, 2O=C*CH*$_2$), 3.41 (t, *J* = 5.1 Hz, 4H, 2NH*CH*$_2$), 4.73 (m, 4H, 2O*CH*$_2$), 6.15 (m, 2H, 2PhCH=*CH*), 6.54 (d, 2H, *J* = 15.7 Hz, Ph*CH*), 7.28-7.36 (complex, 10H), 7.41-7.47 (complex, 3H), 7.94 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 33.8, 43.2, 65.4, 122.4, 123.0, 126.7, 126.9, 127.9, 128.5, 129.4, 132.4, 134.1, 139.3, 171.5; MS m/z (%): 392 (M^+-$C_6H_5SO_2$, 27.1).

(1e). Pale yellow oil; IR (neat): 3283, 3034, 2966, 1732, 1447, 1328 cm^{-1}; ^1H NMR ($CDCl_3$): δ 1.17-1.26 (complex, 6H, 2*CH*$_3$), 2.73 (m, 1H, O=C*CH*), 3.14-3.20 (complex, 2H, NH*CH*$_2$), 4.12 (q, *J* = 7.0 Hz, 2H, O*CH*$_2$), 5.65 (br, 1H, *NH*), 7.55-7.64 (complex, 3H), 7.93 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 14.0, 14.7, 39.6, 45.4, 60.8, 126.8, 129.1, 132.5, 139.9, 174.7; MS m/z (%): 271 (M^+, 36.4).

(2e). Pale yellow oil; IR (neat): 3042, 2982, 1732, 1447, 1316 cm^{-1}; ^1H NMR ($CDCl_3$): δ 1.09-1.19 (complex, 12H, 4*CH*$_3$), 2.78 (m, 2H, 2O=C*CH*), 3.22-3.27 (complex, 4H, 2NH*CH*$_2$), 4.03 (q, *J* = 7.0 Hz, 4H, 2O*CH*$_2$), 7.44-7.53 (complex, 3H), 7.75 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 14.1, 15.3, 39.2, 52.4, 60.7, 127.3, 129.1, 132.7, 139.0, 174.7; MS m/z (%): 385 (M^+, 18.7).

(1f). Pale yellow solid; mp 60-62 °C; IR (neat): 3285, 3044, 2953, 1732, 1447, 1329 cm^{-1}; ^1H NMR ($CDCl_3$): δ 1.14-1.23 (complex, 6H, 2*CH*$_3$), 2.38-2.43 (complex, 2H, O=C*CH*$_2$), 3.68 (m, 1H, NHC*H*), 4.04 (q, *J* = 7.1 Hz, 2H, O*CH*$_2$), 5.39 (br, 1H, *NH*), 7.49-7.59 (complex, 3H), 7.89 (m, 2H); ^{13}C NMR ($CDCl_3$): δ 14.0, 21.0, 40.7, 46.6, 60.7, 126.9, 129.0, 132.5, 140.9, 171.1; MS m/z (%): 271 (M^+, 44.2).

(1g). Pale yellow oil; IR (neat): 3286, 3044, 2960, 1732, 1330 cm^{-1}; ^1H NMR ($CDCl_3$): δ 0.89 (t, *J* = 6.8 Hz, 3H, CH_2*CH*$_3$), 1.33 (m, 2H, CH_3*CH*$_2$), 1.56 (m, 2H, CH_3CH_2*CH*$_2$), 2.40 (s, 3H, Ar*CH*$_3$), 2.51 (t, *J* = 5.3 Hz, 2H, O=C*CH*$_2$), 3.16 (t, *J* = 5.3 Hz, 2H, NH*CH*$_2$), 4.04 (t, *J* = 7.0 Hz, 2H, O*CH*$_2$), 5.76 (br, 1H, *NH*), 7.33 (d, *J* = 7.9 Hz, 2H), 7.73 (d, *J* = 7.9 Hz, 2H); ^{13}C NMR ($CDCl_3$): δ 13.5, 18.9, 21.3, 30.3, 34.1, 38.7, 64.5, 126.9, 129.6, 135.8, 143.2, 172.7; MS m/z (%): 299 (M^+, 28.1).

(2g). Pale yellow oil; IR (neat): 3056, 2975, 1734, 1317 cm^{-1}; ^1H NMR ($CDCl_3$): δ 0.92 (t, *J* = 6.9 Hz, 6H, 2CH_2*CH*$_3$), 1.32 (m, 4H, 2CH_3*CH*$_2$),1.57 (m, 4H, 2CH_3CH_2*CH*$_2$), 2.42 (s, 3H, Ar*CH*$_3$), 2.63 (t, *J* = 5.2 Hz, 4H, 2O=C*CH*$_2$), 3.43 (t, *J* = 5.2 Hz, 4H, 2NH*CH*$_2$), 4.02 (t, *J* = 7.0 Hz, 4H, 2O*CH*$_2$), 7.31 (d, *J* = 7.9 Hz, 2H), 7.72 (d, *J* = 7.9 Hz, 2H); ^{13}C NMR ($CDCl_3$): δ 13.6, 19.0, 21.4, 30.4, 33.9, 45.0, 64.5, 126.9, 129.5, 135.9, 143.5, 172.5; MS m/z (%): 427 (M^+, 9.2).

(1h). Yellow buff; IR (neat): 3289, 3052, 2961, 1732, 1591, 1448, 1329, 1289 cm^{-1}; ^1H NMR ($CDCl_3$): δ 1.15-1.22 (complex, 6H, 2*CH*$_3$), 2.42-2.49 (complex, 2H, O=C*CH*$_2$), 3.59 (m, 1H, NHC*H*), 4.08 (q, *J* = 7.0 Hz, 2H, O*CH*$_2$), 5.78 (br, 1H, *NH*), 7.39 (m, 1H), 7.86-7.97 (complex, 2H), 8.34 (m, 1H); ^{13}C NMR ($CDCl_3$): δ 13.7, 20.5, 41.6, 46.5, 60.9, 124.1, 131.3, 137.5, 142.6, 145.9, 149.1, 172.6; MS m/z (%): 316 (M^+, 33.6).

RESULTS AND DISCUSSION

Microwave-assisted Michael addition of benzenesulfonamide to *n*-butyl acrylate as a model reaction was carried out in the presence of TBAB and various organic and inorganic bases to evaluate their capabilities and selectivities. The reactions were performed at 300 W of microwave power. The results are summarized in Table 1. As Table 1 indicates, when K_2CO_3 was used, higher yield and selectivity as well as shorter reaction time were observed. The model reaction was also examined using conventional heating (110 °C) in the presence of TBAB and K_2CO_3; however, these conditions were not efficient (mono and disubstituted products were obtained in 52 and 19% respectively within 3 h).

In another study, Michael addition of benzenesulfonamide to *n*-butyl acrylate in the presence of TBAB and K_2CO_3 was examined at different microwave powers (100-700 W) (Table 2). As it is shown in Table 2, the best results were attained at

Table 1. The Effect of Bases Upon Michael Addition of Benzenesulfonamide with *n*-Butyl Acrylate in the Presence of TBAB under Microwave Irradiation (300 W)

Entry	Base	Time (min)	Yield (%)[a]	
			Monosubstituted product	Disubstituted product
1	K_2CO_3	5	82	16
2	Cs_2CO_3	5	71	23
3	Na_2CO_3	8	54	8
4	*t*-BuOK	5	62	9
5	CaO	8	67	9
6	NBu_3	8	23	5
7	DABCO	8	16	trace

[a]Isolated yield.

Table 2. The Influence of Different Microwave Powers (W) Upon Michael Addition of Benzenesulfonamide to *n*-Butyl Acrylate in the Presence of K_2CO_3 and TBAB

Entry	MW power (W)	Time (min)	Yield (%)[a]	
			Monosubstituted product	Disubstituted product
1	100	16	37	5
2	200	10	67	10
3	300	5	82	16
4	400	4	74	19
5	500	3	61	18
6	600	2	57	17
7	700	1.5	49	24

[a]Isolated yield.

300 W.

TBAB has an undeniable effect on progress of the reaction. The absence of TBAB in the reaction media gave low yield even by enhancing the reaction time and the microwave power. Thus, the presence of TBAB in our reaction is critically significant. TBAB absorbs the microwave irradiation as well as generates in situ heat and increases the temperature higher than its melting point (100-103 °C). In these conditions, TBAB creates homogeneous media whose resemblance is not far from that of ionic liquids [10]. Other quaternary ammonium salts were also examined in the reaction in which TBAB showed the best results (Table 3).

To establish the generality and applicability of this method, some sulfonamides were added to structurally diverse α,β-unsaturated esters to furnish the corresponding mono and disubstituted Michael adducts with high selectivity in short reaction times. The results are displayed in Table 4.

To study the structural influence of alkoxy group (-OR) of α,β-unsaturated esters (Michael acceptors) upon the reaction, the reaction of benzenesulfonamide with esters containing sterically hindered alkoxy groups was investigated in microwave conditions. As Table 4 shows, the bulkiness of alkoxy group had no significant effect on the yields, the reaction times as well as the selectivities (Table 4, entries 1-4). The structural effect of α,β-unsaturated esters on the Michael addition reaction was also studied. Lower yields of products and longer reaction times were obtained when benzenesulfonamide was introduced to sterically hindered α,β-unsaturated esters (ethyl methacrylate and ethyl crotonate) (Table 4, entries 5 and 6). However, in these cases, the selectivity increased (Table 4, entries 5 and 6). Interestingly, the reaction of benzenesulfonamide as well as *o*-nitrobenzenesulfonamide with ethyl crotonate affords only monosubstituted product (Table 4, entries 6 and 8). The effect of electron-withdrawing and electron-releasing substituents on the aromatic ring of sulfonamide on the reaction was also investigated. The electron-donating substituent (Me) decreased the yield of Michael adduct (Table 4, entries 7). The electron-withdrawing substituent (NO_2) had negligible effect on the reaction yield, but caused the Michael reaction to carry out in shorter reaction time in comparison to other reactions (Table 4, entries 8). This may be attributed to the microwave effect in which the more polar compounds react more readily than less polar compounds [1b].

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Table 3. The Effect of Various Quaternary Ammonium Salts Upon Michael Reaction of Benzenesulfonamide with *n*-Butyl Acrylate in the Presence of K_2CO_3 under Microwave Irradiation (300 W)

Entry	Salt	Time (min)	Yield (%)[a]	
			Monosubstituted product	Disubstituted product
1	-	15	21	trace
2	TBAB	5	82	16
3	TBAI	5	69	15
4	TBACl	5	73	16
5	TBAF	5	30	6
6	$TBAHSO_4$	5	37	8

[a]Isolated yield.

Table 4. Michael Adition of Slfonamides to α,β-Usaturated Eters in the Pesence of K_2CO_3 and TBAB under Mcrowave Iradiation (300 W)

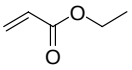
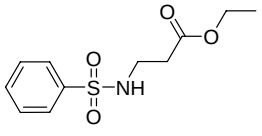
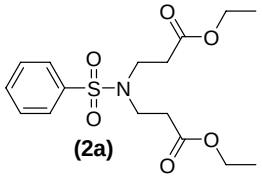
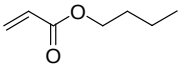
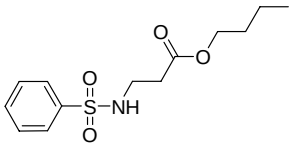
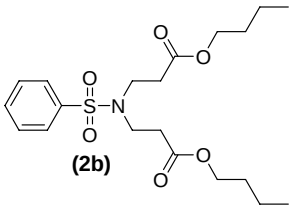
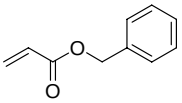
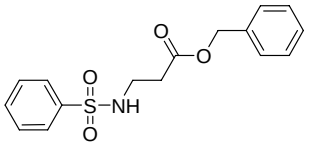
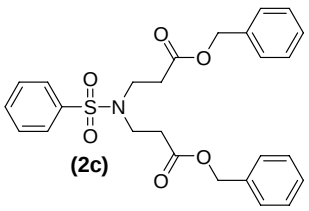
Entry	Ester	Product	Time (min)	Yield (%)[a]
1[b]		 **(1a)**	5	79
		 **(2a)**		17
2		 **(1b)**	5	82
		 **(2b)**		16
3		 **(1c)**	5	83
		 **(2c)**		17

Table 4. Continued

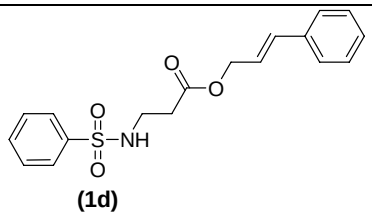
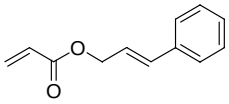
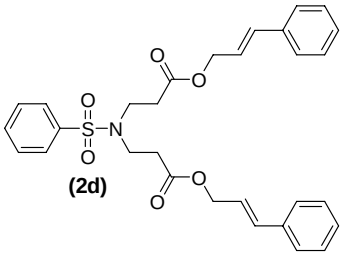
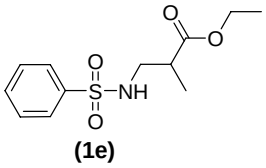
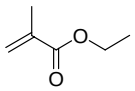
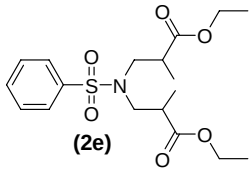
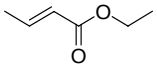
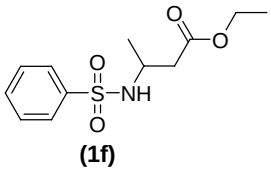
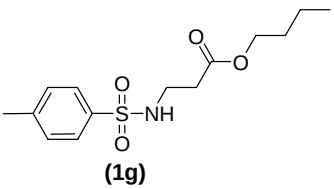
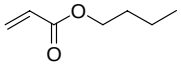
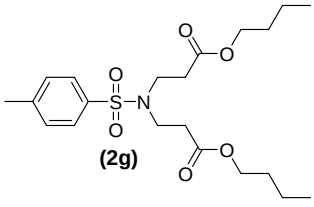
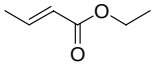
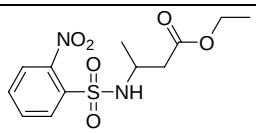
		 **(1d)**		81
4		 **(2d)**	5	16
		 **(1e)**		75
5		 **(2e)**	7	9
6[c]		 **(1f)**	8	70
		 **(1g)**		76
7		 **(2g)**	6	14

Table 4. Continued

8[c]		 **(1h)**	5	68

[a]Isolated yield. [b]In this reaction, the α,β-unsaturated ester/sulfonamide (mol/mol) ratio was 1.5/1. [c]In this case, only monosubstituted Michael adduct was obtained.

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