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Enhanced One-Pot Microwave Efficient Synthesis of Some Pyrrolidine-2, 5-Diones, Butenamides and their Sulfonamide Derivatives Versus Conventional Thermal Heating Technique

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DUE TO the highly important medical applications and wide biological activities of pyrrolidines, butenamides, and their sulfonamide derivatives, such as bacteriocides, fungicides, and insecticides, compounds (4-42) were synthesized from condensation of α, β-unsaturated anhydrides (1-3), with the corresponding amines, using either enhancing microwave irradiation heating or the conventional thermal heating technique in order to compare between their efficiency.

Keywords: Microwave, Synthesis, Condensation, Butenamides, Pyrrolidines and Sulfonamides.

The microwave irradiation comparable to conventional thermal heating proves to generate rapid intense heating with consequent significant reduction in reaction time, enhancement of yield and purity of products, in addition to its friendly environmental effect, known as green chemistry. It proves to be a convenient method of heating, comparable to conventional thermal technique. Moreover, some reactions that do not take place by conventional thermal heating technique or give very low yields, can be accomplished in high yields under microwave irradiation technique[1,2]. Amides, imides, and their derivatives are known to have wide medical applications, biological activities, and as starting materials in industry[3-7]. The aim of this work is the comparison between the two techniques and to synthesis amides and imides derivatives in high and pure yield.

Results and Discussion

Solvent-free microwave irradiation technique

Solvent-free microwave irradiation of 2-phenylmethylenebutanedioic anhydride (1), 2-(4-methoxyphenylmethylene)butanedioic anhydride (2), or 2-(methylphenylmethylene)butanedioic anhydride (3)[8,9] with different N-substituted 4-aminobenzenesulfonamides (a-g)[10], gave the corresponding N-(N'-substituted benzenesulfonamido)-3-carboxy-4-aryl-3-butenamide derivatives, and/or N-(N'-substituted benzenesulfonamido)-3-benzylidinepyrrolidine-2,5-dione derivatives. The microwave irradiation of anhydride (3) with some amines (h-m) gave the corresponding 3-carboxy-3-butenamide derivatives and/or pyrrolidine-2, 5-dione derivatives.

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The results obtained show that under solvent-free microwave irradiation heating of anhydrides (1and 2) with N-phenyl-4-aminobenzenesulfonamide (a), N-(4-methylphenyl)-4-aminobenzenesulfonamide (b), N-(4-methoxy- phenyl)-4-aminobenzenesulfonamide (c), and N-(4-chlorophenyl)-4-amino- benzenesulfonamide (d)[7] gave pyrrolidine-2,5-diones (4-7, 12-14 and 16), whereas anhydride (3) gave separable mixtures from the corresponding butenamides (21, 23, 25, and 27) and pyrrolidine-2,5-diones (22, 24, 26, and 28). This can be attributed to the steric effect exerted by the methyl group in anhydride (3).

However, microwave irradiation of anhydrides (1-3) with either N-(4-nitrophenyl-4-aminobenzenesulfonamide (e) or N-benzyl-4-aminobenzenesulfonamide (g), gave the corresponding butenamides (8, 11, 17, 20, 29, and 32). This is ascribed to low nucleophilicity of the amido nitrogen atom towards further intramolecular nucleophilic attack on the carbonyl carbon to give pyrrolidine-2,5-dione, due to the presence of the nitro group, and the steric effect exerted by the sp^3 methylene group in the benzyl moiety, respectively.

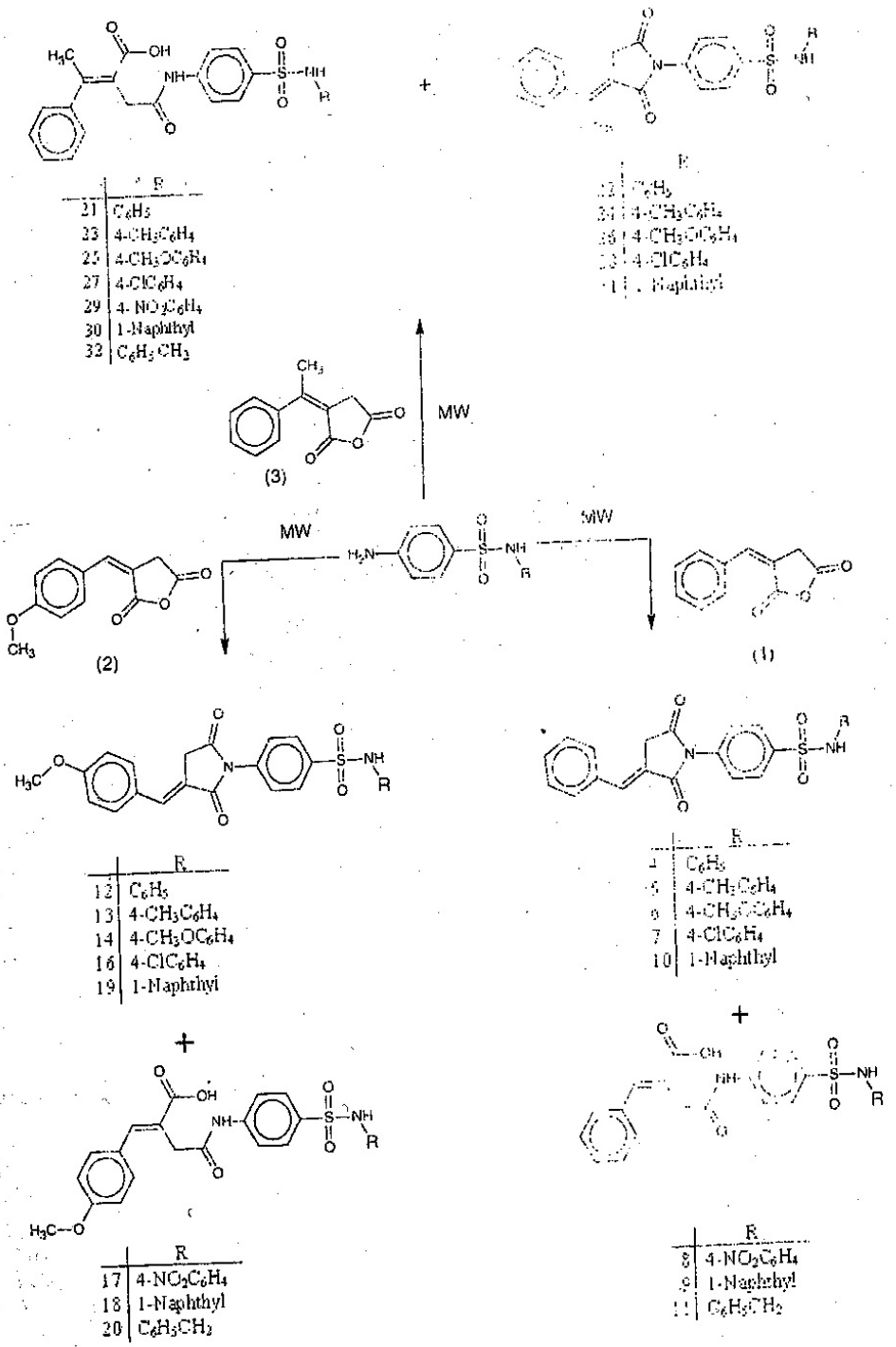
On the other hand, N-(1-naphthyl)-4-aminobenzenesulfonamide (f) gave with anhydrides (1-3), the respective separable mixtures from butenamides (9, 18, and 30) and pyrrolidine-2, 5-diones (10, 19, and 31). The formation of the cyclic pyrrolidine-2, 5-diones, irrespective to the low reactivity of the amido nitrogen atom in butenamides to form cyclic compounds can be attributed to the coplanarity of the naphthyl ring that facilitates the formation of the cyclic compounds (Scheme 1).

The results obtained from microwave irradiation of anhydride (3) with amines; 4-methylaniline (h), 4-methoxyaniline (i), 4-chloroaniline (j), 4-nitroaniline (k), 1-naphthylamine (l), and benzylamine (m), show that amines (h-k) gave the corresponding pyrrolidine-2,5-diones (34, 36, 38, and 40), whereas amine (l), gave a separable mixture from butenamide (41) and pyrrolidine- 2,5-dione (42), and amine (m) gave butenamide (43) as an only product (Scheme 2).

Comparison between the results obtained from reaction of anhydride (3) with N-substituted 4-aminobenzenesulfonamides (b-g) versus amines (h-m); shows that the presence of the benzenesulfonamido moiety decreases the reactivity of the amido nitrogen atom towards intramolecular nucleophilic attack to form the cyclic compounds.

Conventional thermal heating technique

Condensation of anhydrides (1-3) with different 4-amino-N-phenyl-benzenesulfonamide derivatives (a-g) (1:2 mole) in ethanol for at least 4 hr gave the corresponding N-(N'-substituted benzenesulfonamido)-3-carboxy-4-aryl-3-butenamide derivatives, and/or N-(N'-substituted benzenesulfonamido)-3-benzylidinepyrrolidine-2, 5-dione derivatives. Thermal heating of anhydride (3) with amines (h-m) gave the corresponding 3-carboxy-3-butenamide derivatives and/or pyrrolidine-2, 5-dione derivatives (Scheme 3).



	R
21	C_6H_5
23	4-$CH_3C_6H_4$
25	4-$CH_3OC_6H_4$
27	4-ClC_6H_4
29	4-$NO_2C_6H_4$
30	1-Naphthyl
32	$C_6H_5CH_2$

	R
12	C_6H_5
13	4-$CH_3C_6H_4$
14	4-$CH_3OC_6H_4$
16	4-ClC_6H_4
19	1-Naphthyl

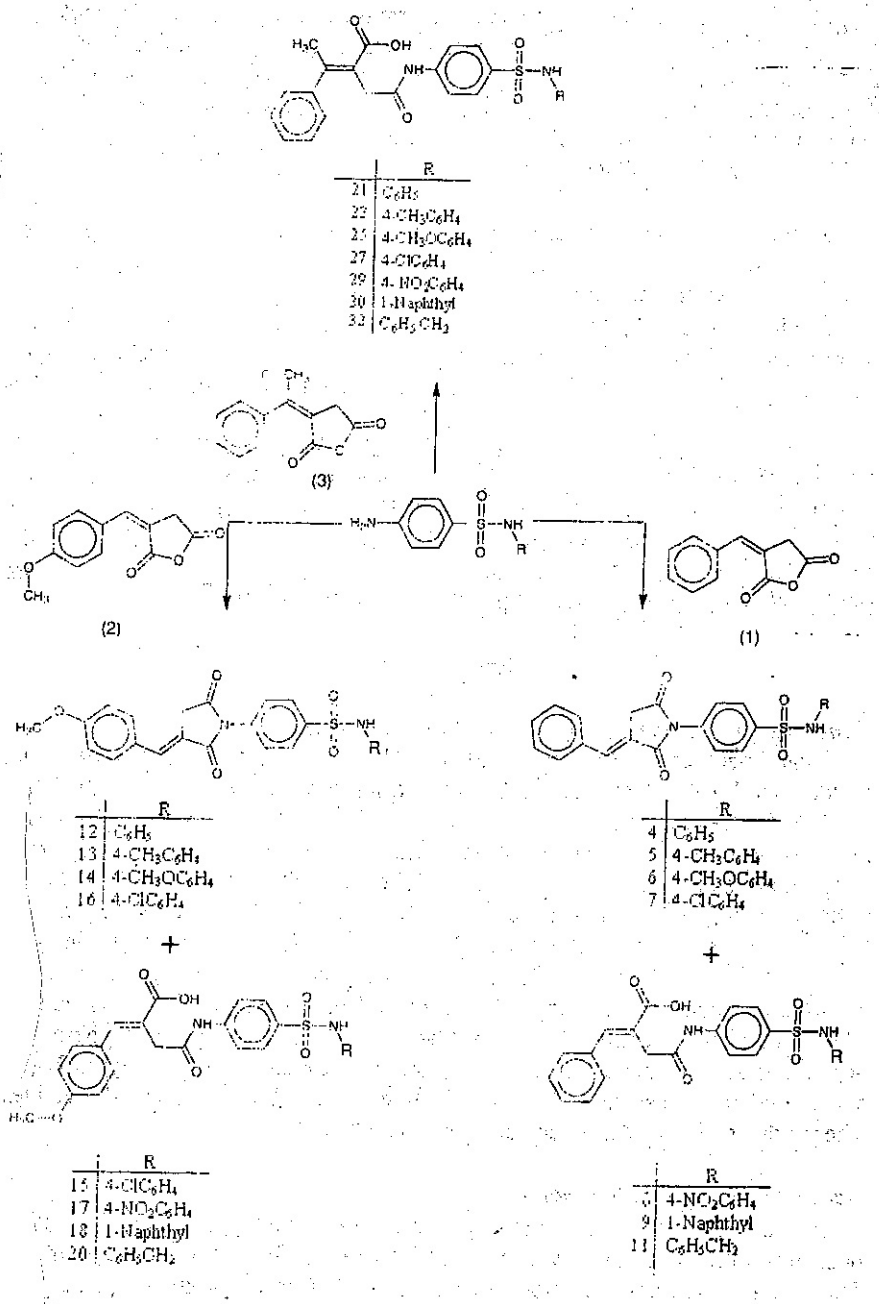
	R
4	C_6H_5
5	4-$CH_3C_6H_4$
6	4-$CH_3OC_6H_4$
7	4-ClC_6H_4
10	1-Naphthyl

	R
17	4-$NO_2C_6H_4$
18	1-Naphthyl
20	$C_6H_5CH_2$

	R
8	4-$NO_2C_6H_4$
9	1-Naphthyl
11	$C_6H_5CH_2$

Scheme 1.

Scheme 2.


R
21 C6H5
23 4-CH3C6H4
25 4-CH3OC6H4
27 4-ClC6H4
29 4-NO2C6H4
30 1-Naphthyl
32 C6H5CH2
(3)
(2)
(1)
R
12 C6H5
13 4-CH3C6H4
14 4-CH3OC6H4
16 4-ClC6H4
R
4 C6H5
5 4-CH3C6H4
6 4-CH3OC6H4
7 4-ClC6H4
R
15 4-ClC6H4
17 4-NO2C6H4
18 1-Naphthyl
20 C6H5CH2
R
8 4-NO2C6H4
9 1-Naphthyl
11 C6H5CH2

Scheme 3.

The condensation of anhydrides (1 and 2) with amines (a-c) using conventional thermal heating technique, gave pyrrolidine-2, 5-diones (4-6) and (12-14), respectively. With amine (d), thermal condensation of anhydride (1) gave pyrrolidine-2, 5-dione (7), whereas anhydride (2) gave a separable mixture from butenamide (15) and pyrrolidine-2, 5-dione (16). This can be attributed to the presence of mesomeric effect exerted by the methoxy group in anhydride (2) which decreases the susceptibility of the carbonyl carbon towards the anchimeric attack by the amido nitrogen atom.

On the other hand, the formation of only butenamides (8, 9) and (17, 18), from the thermal heating of anhydrides (1 and 2), respectively, with amines (e- f), can be ascribed to the low nucleophilicity of the amido nitrogen atom in the butenamides (8, 9, 17, and 18), whereas with amine (g), irrespective to the higher nucleophilicity of the amido nitrogen, its reactivity towards cyclization decreases due to the steric effect exerted by the sp^3 methylene group.

However, the formation of butenamides (21, 23, 25, 27, ,29, 30, 32, 33, 35, 37, 39, 41, and 43) as only products, resulted from thermal condensation of anhydride (3) with amines (a-m) can be attributed to the steric effect exerted by the tetrahedral sp^3 methyl carbon in anhydride (3), which decreases the susceptibility of the carbonyl carbon to be attacked by the amido nitrogen atom to form the corresponding pyrrolidine-2, 5-dione (Scheme 2)

Experimental

General solvent-free microwave irradiation technique

In an open vessel of the anhydride and N-substituted 4-aminobenzenesulfonamide was dry irradiated from 2-5 min in a microwave oven (1000 watt, 100% power). The reaction progress was monitored by TLC until no more unreacting materials were observed. The reaction mixture was then cooled down to the room temperature and dissolved in chloroform. The chloroform layer was then washed with dilute HCl to get rid of unreacted amines, and then extracted with 10% ice-cold sodium carbonate solution. Acidification of the aqueous layer with ice-cold concentrated hydrochloric acid, precipitated the carboxylic compounds; N-(N'-substituted benzenesulfonamido)-3-carboxy-4-aryl-3-butenamide derivatives. Thoroughly wash of the organic layer with water followed by its dryness over anhydrous sodium sulfate followed by organic solvent distillation gave N-(N'-substituted benzenesulfonamido)-3-substituted pyrrolidine-2, 5-dione derivatives. The products obtained were crystallized from ethanol.

General conventional thermal heating technique

A mixture of α, β-unsaturated anhydride (1-3) and N-substituted 4-amino-benzenesulfonamide (1:2) was heated under reflux for at least 4 hr in ethanol. The reaction progress was monitored by thin layer chromatography (TLC). The reaction mixture was then concentrated and the precipitate formed was filtered and dissolved in chloroform then worked up in the same way given in the

solvent-free microwave irradiation technique. Structures of products are confirmed by their spectral analyses, FTIR, ^{1}HNMR, and MS.

N-(N'-Phenylbenzenesulfonamido)-3-benzylidinepyrrolidine-2, 5-dione (4)

White crystals, m.p. 195^{o}C, 83% yield in m.w, and 45% in thermal. FTIR (KBr): υ (cm^{-1}) = 3288 (NH), 1775-1704 (2CO; imide), 1372 and 1157 (SO_2; asy. and sym.). MS: m/z = 418 (M^+, 86.5%, $C_{23}H_{18}N_2O_4S$), 327 (12.8, $C_{17}H_{13}NO_2S$), 326 (46.8, $C_{17}H_{12}NO_4S$) 263 (30, $C_{17}H_{13}NO_2$), 234(31.4, $C_{16}H_{12}NO$), 206 (18, $C_{15}H_{12}N$), 143 (6.5, $C_{10}H_7O$), 116 (34.5, C_9H_8), 115 (100, C_9H_7), and 97 (5.2, $C_4H_3NO_2$).

N-[N'-(4-Methylphenyl)benzenesulfonamido]-3-benzylidinepyrrolidine-2,5-dione (5)

Yellow crystals, m.p. 238^{o}C, 91% yield in m.w, and 64% in thermal. FTIR (KBr): υ(cm^{-1}) = 3271.5 (NH), 1772-1707 (2CO; imide), 1333 and 1160 (SO_2; asy. and sym.). MS: m/z = 432 (M^+, 8.3%, $C_{24}H_{20}N_2O_4S$), 326 (1.6, $C_{17}H_{12}NO_2S$), 262 (6.5, $C_{13}H_{14}N_2O_2S$), 116 (40.4, C_9H_8), 115(64, C_9H_7), 107 (8.8, C_7H_9N), 106 (100, C_7H_8N) and 91 (14.5, C_7H_7). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.62-7.73 (3H, m; Ph), 7.86-7.93 (2H, m; Ph), 7.61(1H, s; C<u>H</u> Ph), 3.87 (2H, s C<u>H</u>$_2$CO), 7.06 (2H, d; CH_3<u>Ph</u>), 7.50-7.53 (2H, d; NH<u>Ph</u>), 10.28 (1H, s, N<u>H</u>SO_2), 7.55-7.58 [(4H, s; NH<u>Ph</u>SO_2) and (1H, imp; N<u>H</u>CO)], and 2.15 (3H, s; C<u>H</u>$_3$Ph).

N-[N'-(4-Methoxyphenyl)benzenesulfonamido]-3-benzylidinepyrrolidine-2,5-dione (6)

Pale gray crystals, m.p. 185^{o}C, 92% yield in m.w, and 60% in thermal. FTIR (KBr): υ (cm^{-1}) =3264 (NH), 1774-1706 (2CO; imide), 1333 and 1160 (SO_2; asy. and sym.). MS: m/z = 448 (M^+, 48%, $C_{24}H_{20}N_2O_5S$), 292 (8.7, $C_{17}H_{10}NO_2S$), 123 (9, C_7H_9NO), 122 (100, C_7H_8NO or C_6H_4NS), 116 (5.8, C_9H_8) and 115 (22.4, C_9H_7). ^{1}HNMR (DMSO-d_6): δ (ppm)= 7.48-7.59 (3H, m; Ph), 7.70-7.82 (2H, m; Ph), 7.55 (1H, s; C<u>H</u> Ph), 3.87 (2H, s, C<u>H</u>$_2$CO), 7.59 [(4H, s; NH<u>Ph</u>SO_2) and (1H, imp; N<u>H</u>CO)], 10.09 (1H, s, N<u>H</u>SO_2), 7.01-7.05 (2H, d; NH<u>Ph</u>), 6.81-6.85 (2H, d; <u>Ph</u>OCH_3) and 3.68 (3H, s; OC<u>H</u>$_3$).

N-[N'-(4-Chlorophenyl)benzenesulfonamido]-3-benzylidinepyrrolidine-2,5-dione (7)

Pale yellow crystals, m.p. 203^{o}C, 94 % yield in m.w, and 54% in thermal. FTIR (KBr): υ (cm^{-1}) =3244 (NH), 1771-1711.5 (2CO; imide), 1373 and 1157 (SO_2; asy. and sym.). MS: m/z = 452 (M^+, 86.5%, $C_{23}H_{17}N_2O_4SCl$), 326 (38.9, $C_{17}H_{12}NO_4S$), 262(100, $C_{17}H_{12}NO_2$), 206 (12.7, $C_{15}H_{12}N$), 144 (6.2, $C_{10}H_8O$), 127 (10, C_6H_6NCl), 126 (32.2, C_6H_5NCl), 125 (3.2, C_6H_4NCl), 116(23.5 C_9H_8) and 115 (55, C_9H_7). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.70-7.74 (3H, m; Ph), 7.90-7.94 (2H, m; Ph), 7.59 (1H, s; C<u>H</u> Ph), 3.87 (2H, s, C<u>H</u>$_2$CO), 7.59-7.63 (2H, d; <u>Ph</u>NCO), 7.49-7.52 (2H, d; <u>Ph</u>SO_2), 10.60 (1H, s, N<u>H</u>SO_2), 7.14-7.18 (2H, d; <u>Ph</u>Cl) and 7.32-7.36 (2H, d; <u>Ph</u>NH).

N-[N'-(4-Nitrophenyl)benzenesulfonamido]-3-carboxy-4-phenyl-3-utenamide (8)

Yellow crystals, m.p. 180°C, 94% yield in m.w, and 47% in thermal. FTIR (KBr): υ (cm^{-1})= 3351-3224.5 (2NH), 3400-2400 (OH; acid), 1707.5(CO; acid), 1648.5(CO; amide), 1331 and 1151.5(SO_2; asy. and sym.). MS: m/z = 481 (M^+, 0%, $C_{23}H_{19}N_3O_7S$), 463 (42, $C_{23}H_{17}N_3O_6S$), 327 (9.3, $C_{17}H_{13}NO_4S$), 262 (46.2, $C_{12}H_{10}NO_3S$), 156 (100, $C_6H_6NO_2S$), 116 (17, C_9H_8), 115 (50.3, C_9H_7), 92 (55.6, C_6H_6N), and 65 (42, HO_2S).

N-[N'-(1-Naphthyl)benzenesulfonamido]-3-carboxy-4-phenyl-3-butenamide (9)

White crystals, m.p.150°C, 63 % yield in m.w, and 60% in thermal. FTIR (KBr): υ (cm^{-1}) =3300-3244 (2NH), 3400-2400 (OH; acid), 1700(CO; acid), 1678.1(CO; amide), 1344 and 1158(SO_2; asy. and sym.). MS: m/z = 468 (M^+, 100%, $C_{27}H_{22}N_2O_5S$), 326 (28, $C_{17}H_{12}NO_4S$), 298 (30, $C_{16}H_{14}NO_2S$), 143(37.5, $C_{10}H_7O$), 142 (93, $C_{10}H_8N$), 117 (9.6, C_9H_9), 116 (15.5 C_9H_8), 115 (74, C_9H_7) and 65 (8.4, HOS_2).

N-[N'-(1-Naphthyl)benzenesulfonamido]-3-benzylidinepyrrolidine-2,5-dione (10)

Brown crystals, m.p. 89°C, 19 % yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) = 3247 (NH), 1770.6-1710 (2CO; imide), 1374 and 1159 (SO_2; asy. and sym.). MS: m/z = 468 (M^+, 0%, $C_{27}H_{20}N_2O_4S$), 314 (18.5, $C_{16}H_{14}N_2O_3S$), 313 (79.5, $C_{16}H_{13}N_2O_3S$), 298 (7.7, $C_{16}H_{14}N_2O_2S$), 286(2.5, $C_{15}H_{14}N_2O_2S$), 285 (10.4, $C_{15}H_{13}N_2O_2S$), 144 (7, $C_{10}H_8O$), 143 (5, $C_{10}H_7O$), 116(64, C_9H_8) and 115 (100, C_9H_7).

N-(N'-Benzylbenzenesulfonamido]-3-carboxy-4-phenyl-3-butenamide (11)

White crystals, m.p. 170°C, 88 % yield in m.w, and 61% in thermal. FTIR (KBr): υ (cm^{-1}) = 3332.4-3203.3 (2NH), 3400-2400 (OH; acid), 1682.5(CO; acid), 1634.5(CO; amide), 1538 and 1156.5(SO_2; asy. and sym.). MS: m/z = 450 (M^+, 20.7%, $C_{24}H_{22}N_2O_5S$), 116 (26.4, C_9H_8), 115 (39, C_9H_7), 108 (12.7, $C_7H_{10}N$), 107 (11, C_7H_9N), 106(100, C_7H_8N), 91 (20, C_7H_7), 66 (7.4, H_2O_2S), 65 (19, HO_2S) and 64 (5, O_2S).

^{1}HNMR (DMSO-d_6): δ (ppm) = 7.77-7.27 (3H, m; Ph), 7.84-7.78 (2H, m; Ph), 7.48 (1H, s; C<u>H</u>Ph), 3.57 (2H, s; C<u>H$_2$</u>CO), 10.53 (1H, s; N<u>H</u>SO_2), 7.78 [4H, s; (4H, s; NH<u>Ph</u>SO_2) and N<u>H</u>CO)], 8.06-8.00 (1H, t; N<u>H</u>CH_2), 3.94-3.97 (2H, d; C<u>H$_2$</u>NH), 7.27 (5H, s; <u>Ph</u>CH_2) and 12.5 (1H, broad; COO<u>H</u>).

N-(N'-Phenylbenzenesulfonamido)-3-(4-methoxybenzylidine)pyrrolidine-2,5-dione(12)

Pale yellow crystals, m.p. 215°C, 91% yield in m.w, and 67% in thermal. FTIR (KBr): υ (cm^{-1}) =3242.5 (NH), 1767-1702 (2CO; imide), 1380 and 1161(SO_2; asy. and sym.). MS: m/z = 448 (M^+, 9.1%, $C_{24}H_{20}N_2O_5S$), 292 (13, $C_{18}H_{14}NO_3$), 265 (9, $C_{17}H_{15}NO$), 146(14.4, $C_{10}H_{10}O$), 145(14, $C_{10}H_9O$), 131 (12.3, C_9H_7O) and 92(100, C_6H_6N). ^{1}HNMR (DMSO-d_6): δ (ppm) = 3.84 (3H, s; OC<u>H$_3$</u>), 7.05-7.09 (2H, d; <u>Ph</u>OCH_3), 7.90-7.94 (2H, d; <u>Ph</u>CH=), 7.66 (1H, s;

CHPh), 3.2-3.6 (2H, imp.; CH_2CO), 7.70 [4H, s; N<u>Ph</u>SO_2)], 10.46 (1H, broad; $NHSO_2$) and 7.14-7.62 (5H, m; Ph).

N-[N'-(4-Methylphenyl)benzenesulfonamido]-3-(4-methoxybenzylidine) pyrrolidine -2,5-dione (13)

Yellow crystals, m.p. 220°C, 93% yield in m.w, and 70% in thermal. FTIR (KBr): υ (cm^{-1}) =3276.3 (NH), 1772.5-1706 (2CO; imide), 1375 and 1161(SO_2; asy and sym.). MS: m/z = 462 (M^+, 0%, $C_{25}H_{22}N_2O_5S$), 246 (6.5, $C_{13}H_{12}NO_2S$), 216 (3.2, $C_{12}H_{10}NO_3$), 146 (12, $C_{10}H_{10}O$), 145 (6.5, $C_{10}H_9O$), 132(33.6, C_9H_8O), 115 (18.4, C_9H_7), 105 (100, C_7H_7N) and 104 (97, C_7H_6N). ^{1}HNMR (DMSO-d_6): δ (ppm) = 3.83 (3H, s; OCH_3), 7.04-7.09 (2H, d; <u>Ph</u>OCH_3), 7.86-7.90 (2H, d; <u>Ph</u>CH=), 7.59 (1H, s; C<u>H</u>Ph), 3.2-3.6 (2H, imp.; C<u>H_2</u>CO), 7.55 (4H, s; (N<u>Ph</u>SO_2), 10.27 (1H, s; N<u>H</u>SO_2), 7.04-7.09 [4H,s; CH_3<u>Ph</u>NH and 2.20 (3H, s; PhC<u>H_3</u>).

N-[N'-(4-Methoxyphenyl)benzenesulfonamido]-3-(4-methoxybenzylidine) pyrrolidine-2,5-dione (14)

Pale yellow crystals, m.p. 175°C, 95% yield in m.w, and 71% in thermal. FTIR (KBr): υ (cm^{-1}) =3281 (NH), 1769-1706 (2CO; imide), 1377 and 1164 (SO_2; asy. and sym.). MS: m/z = 476 (M^+, 46%, $C_{25}H_{22}N_2O_6S$), 292 (9.1, $C_{18}H_{14}NO_3$), 146 (5.4, $C_{10}H_{10}O$), 145(9.8, $C_{10}H_9O$), 131 (4, C_9H_7O), 123 (9, C_7H_9NO), 122 (100, C_7H_8NO), 106 (2.6, C_6H_4NO), 103 (6.3, C_8H_7), and 65 (2, HOS_2).

N-[N'-(4-Chlorophenyl)benzenesulfonamido]-3-carboxy-4-(4-methoxyphenyl)- 3-butenamide (15)

White crystals, m.p. 179°C, 0% yield in m.w, and 47% in thermal. FTIR (KBr): υ (cm^{-1}) = 3261-3193 (2NH), 3400-2400 (OH; acid), 1690 (CO, acid), 1667.2 (CO; amide), 1336 and 1157.5(SO_2; asy. and sym.). MS: m/z= 500 (M^+, 0%, $C_{24}H_{21}N_2O_6SCl$), 327 (3.2, $C_{16}H_{11}N_2O_4S$), 282 (44, $C_{12}H_{11}N_2O_2SCl$), 218 (23.5, $C_{12}H_{10}O_4$), 217 (4.2, $C_{12}H_{11}NO_3$), 191 (3.3, $C_{11}H_{11}O_3$), 156 (100, $C_6H_6NO_2S$), 146 (54, $C_{10}H_{10}O$) and 126 (11.8, C_6H_5NCl). ^{1}HNMR (DMSO-d_6): δ (ppm) = 3.77 (3H, s; OCH_3), 7.08-7.11 (2H, d; <u>Ph</u>OCH_3), 7.47-7.43 (2H, d; <u>Ph</u>CH=), 3.55 (2H, s; C<u>H_2</u>CO), 10.35 (1H, s; N<u>H</u>CO), 7.77 [(4H, s; N<u>Ph</u>SO_2), and (1H,imp.;C<u>H</u>Ph)], 10.55 (1H, s; N<u>H</u>SO_2), 6.98-7.02 (2H, d; <u>Ph</u>NH) and 7.28-7.32 (2H, d; <u>Ph</u>Cl).

N-[N'-(4-Chlorophenyl)benzenesulfonamido]-3-(4methoxybenzylidine) pyrrolidine-2,5-dione (16)

Yellow crystals, m.p. 230°C, 86% yield in m.w, and 30% in thermal. FTIR (KBr): υ (cm^{-1}) =3254 (NH), 1768-1708 (2CO; imide), 1377 and 1161(SO_2; asy. and sym.). MS: m/z = 482 (M^+, 65.4%, $C_{24}H_{19}N_2O_5SCl$), 356 (39.1, $C_{18}H_{14}NO_5S$), 293 (27.6, $C_{18}H_{15}NO_3$), 292 (100, $C_{18}H_{14}NO_3$), 264 (33.7, $C_{12}H_7NO_2$), 146 (25.4, $C_{10}H_{10}O$), 145 (21.3, $C_{10}H_9O$), 131 (21.7, C_9H_7O), 126 (31, C_6H_5NCl) and 115 (6.5, C_9H_7). ^{1}HNMR (DMSO-d_6): δ (ppm) = 3.83 (3H, s; OCH_3), 7.13-7.17 (2H, d; <u>Ph</u>OCH_3) , 7.57-7.62 (2H, d; <u>Ph</u>CH=), 7.57-7.69 (1H,

imp.; C<u>H</u>Ph), 3.2-3.6 (2H, imp.; C<u>H</u>$_2$CO), 7.89 (2H, d; <u>Ph</u>N), 7.93 (2H, d; <u>Ph</u>SO$_2$), 10.60 (1H, s; N<u>H</u>SO$_2$), 7.05-7.09 (2H,d; <u>Ph</u>Cl) and 7.32-7.36 (2H, d; <u>Ph</u>NH).

N-[N'-(4-Nitrophenyl)benzenesulfonamido]-3-carboxy-4-(4-methoxyphenyl) -3-butenamide (17)

Yellowish brown crystals, m.p. 150 ^{o}C, 91% yield in m.w, and 43% in thermal. FTIR (KBr): υ (cm^{-1}) = 3295-3255 (2NH), 3400-2400 (OH, acid), 1690(CO; acid), 1672.3 (CO; amide), 1347 and 1160(SO$_2$; asy. and sym.). MS: m/z = 511(M$^+$, 0%, $C_{24}H_{21}N_3O_8S$), 493 (18.6, $C_{24}H_{19}N_3O_7S$), 293 (43.8, $C_{12}H_{11}N_3O_4S$), 218(23.4, $C_{12}H_{10}O_4$), 156 (100, $C_6H_6NO_2S$), 146 (28, $C_{19}H_{10}O$), 139 (6.6, $C_6H_7N_2O_2$), 131 (8, C_9H_7O), 115 (6.3, C_9H_7) and 92 (94.8, C_6H_6N).

N-[N'-(1-Naphthyl) benzenesulfonamido] -3-carboxy-4- (4-methoxyphenyl)-3-butenamide (18)

Pale brown crystals, m.p. 170^{o}C, 64% yield in m.w, and 68% in thermal. FTIR (KBr): υ (cm^{-1}) =3244-3203 (2NH), 3400-2400 (OH; acid), 1717(CO; acid), 1667(CO; amide), 1257 and 1176.8(SO$_2$; asy. and sym.). MS: m/z = 516 (M$^+$, 0%, $C_{28}H_{24}N_2O_6S$), 499(22.3, $C_{28}H_{23}N_2O_5S$), 498 (17, $C_{28}H_{22}N_2O_5S$), 218 (65, $C_{12}H_{10}O_4$), 191 (58.8, $C_{11}H_{11}O_3$), 147 (40, $C_{10}H_{11}O$), 146 (100, $C_{10}H_{10}O$), 143 (38.1 $C_{10}H_9N$), 131 (47.5, C_9H_7O), 115 (15.3, C_9H_7) and 92 (11.3, C_6H_6N).

N-[N'-(1-Naphthyl)benzenesulfonamido]-3-(4-methoxybenzylidine) pyrrolidine -2,5-dione (19)

Reddish brown crystals, m.p. 226^{o}C, 26% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) =3281(NH), 1704-1706 (2CO; imide), 1599 and 1126(SO$_2$; asy. and sym.). MS: m/z = 498 (M$^+$, 87.6%, $C_{28}H_{22}N_2O_5S$), 146 (12.6, $C_{10}H_{10}O$), 145 (9.1, $C_{10}H_9O$), 143 (24.3, $C_{10}H_7O$ or $C_{10}H_9N$), 142(100, $C_{10}H_8N$), 122(6.2, C_6H_4NS), 115(53.3, C_9H_7) and 103 (9, C_8H_7).

N-[N'-Benzylbenzenesulfonamido]-3-carboxy-4-(4-methoxyphenyl-3-butenamide (20)

Gray crystals, m.p. 190^{o}C, 88% yield in m.w, and 27% in thermal. FTIR (KBr): υ (cm^{-1}) =3371-3260(2NH), 3400-2400 (OH; acid), 1711(CO; acid), 1680 (CO; amide), 1323 and 1154(SO$_2$; asy. and sym.). MS: m/z = 480 (M$^+$, 9.1%, $C_{25}H_{24}N_2O_6S$), 293 (15, $C_{18}H_{15}NO_3$), 262 (12, $C_{13}H_{14}N_2O_2S$), 191 (10, $C_{11}H_{11}O_3$), 156 (15 $C_6H_6NO_2S$), 146 (56.5, $C_{10}H_{10}O$), 145(39.4, $C_{10}H_9O$), 106 (71.7, C_7H_8N), 91 (84, C_7H_7) and 77 (100, C_6H_5). ^{1}HNMR (DMSO-d$_6$): δ (ppm) = 3.79 (3H, s; OCH$_3$), 7.00-7.04 (2H, d; <u>Ph</u>OCH$_3$) , 7.47-7.50 (2H, d; PhCH=), 3.59 (2H, s; C<u>H</u>$_2$CO), 10.58 (1H, s; N<u>H</u>CO), 7.79 [4H, s; NH<u>Ph</u>SO$_2$) and (1H, imp., PhC<u>H</u>=)], 8.02 (1H, t; N<u>H</u>SO$_2$), 3.97 (2H, d; C<u>H</u>$_2$NH) and 7.28 (5H, s; <u>Ph</u>CH$_2$).

N-(N'-Phenylbenzenesulfonamido)-3-carboxy-4-methyl-4-phenyl-3-butenamide (21)

White crystals, m.p. 162^{o}C, 60% yield in m.w, and 20% in thermal. FTIR (KBr): υ (cm^{-1}) = 3300-3250(2NH), 3400-2400 (OH; acid), 1690(CO; acid), 1688 (CO; amide), 1337 and 1157(SO$_2$; asy and sym.). MS: m/z = 450 (M$^+$, 0%, $C_{24}H_{22}N_2O_5S$), 433 (40, $C_{24}H_{21}N_2O_4S$), 432 (100, $C_{24}H_{20}N_2O_4S$), 340 (37.3,

$C_{18}H_{14}NO_4S$), 277 (0, 4 $C_{18}H_{15}NO_2$), 276 (62.7, $C_{18}H_{14}NO_2$), 248(30, $C_{12}H_{12}N_2O_2S$), 156 (28, $C_6H_6NO_2S$), 130 (23.2, $C_{10}H_{10}$), 115 (28.4, C_9H_7) and 92 (40.4, C_6H_6N). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.66 (5H, s; Ph), 2.36 (3H, s; PhC(C<u>H</u>$_3$)=C), 3.19 (2H, s; C<u>H2</u>CO), 10.39 (1H, s; N<u>H</u>SO_2), 7.36 (4H, s; N<u>Ph</u>SO_2), 10.16 (1H, s; N<u>H</u>CO), 7.05-7.23 (5H, m; NH<u>Ph</u>) and 12.56 (1H, broad; COOH).

N-(N'-Phenylbenzenesulfonamido)-3-phenylethylidinepyrrolidine-2, 5-dione (22)

Pale yellow crystals, m.p. 136°C, 20% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) =3248.3(NH), 1766-1707 (2CO; imide), 1377and 1159 (SO_2; asy. and sym.). MS: m/z = 432 (M^+, 100%, $C_{24}H_{20}N_2O_4S$), 340 (47, $C_{18}H_{14}NO_4S$), 276 (93.8, $C_{18}H_{14}NO_2$), 248(29.6, $C_{12}H_{12}N_2O_2S$), 233 (10, $C_{12}H_{11}NO_2S$), 130 (29, $C_{10}H_{10}$), 129 (43.8, $C_{10}H_9$), 115 (32.4, C_9H_7) and 92 (44, C_6H_6N). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.52-7.66 (3H, m; Ph), 7.89-7.93 (2H, m; Ph), 2.34 (3H, s; PhC(C<u>H</u>$_3$)=C), 2.63 (2H, s; C<u>H</u>$_2$CO), 7.44 (4H, s; N<u>Ph</u>SO_2), 10.44 (1H, s; N<u>H</u>SO_2) and 7.05-7.32 (5H, m; Ph).

N-[N'-(4-Methylphenyl)benzenesulfonamido]-3-carboxy-4-methyl-4-phenyl-3-butenamide (23)

White crystals, m.p. 210°C, 67% yield in m.w, and 22% in thermal. FTIR (KBr): υ (cm^{-1}) = 3300-3255(2NH), 3400-2400 (OH; acid), 1705(CO; acid), 1688.5 (CO; amide), 1331 and 1157(SO_2; asy. and sym.). MS: m/z =464 (M^+, 0%, $C_{25}H_{24}N_2O_5S$), 447 (80.7, $C_{25}H_{23}N_2O_4S$), 446 (79, $C_{25}H_{22}N_2O_4S$), 277 (15.3, $C_{18}H_{15}NO_2$), 276(33, $C_{18}H_{14}NO_2$), 130(8, $C_{10}H_{10}$), 115 (22.5, C_9H_7), 107 (6.5, C_7H_9N), 106(100, C_7H_8N) and 91 (2.2, C_7H_7). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.63 (5H, s; Ph), 2.35 (3H, s; PhC(C<u>H</u>$_3$)=C), 3.18 (2H, s; C<u>H</u>$_2$CO), 10.24 (1H, s; N<u>H</u>SO_2), 7.36 (4H, s; NH<u>Ph</u>SO_2), 9.99 (1H, s; N<u>H</u>CO), 7.24 (2H,d; N<u>H</u>Ph), 7.00 (2H,d; CH_3<u>Ph</u>) and 2.18 (3H, s; C<u>H</u>$_3$Ph).

N-[N'-(4-Methylphenyl)benzenesulfonamido]-3-phenylethylidinepyrrolidine-2,5-dione (24)

Yellow crystals, m.p. 130°C, 25% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) =3250(NH), 1767-1705 (2CO; imide), 1378 and 1158 (SO_2; asy. and sym.). MS: m/z = 446 (M^+, 90.8%, $C_{25}H_{22}N_2O_4S$), 277 (20, $C_{18}H_{15}NO_2$), 276 (28.5, $C_{18}H_{14}NO_2$), 340(9.8, $C_{18}H_{14}NO_4S$), 130 (12, $C_{10}H_{10}$), 115 (23, C_9H_7), 107 (13, C_7H_9N), 106 (100, C_7H_8N), 102 (3, C_8H_6) and 79 (30, HNO_2S). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.49-7.64 (3H, m; Ph), 7.78-7.89 (2H, m; Ph), 2.36 (3H, s; PhC(C<u>H</u>$_3$)=C), 2.6 (2H, s; C<u>H</u>$_2$CO), 7.44 (4H, s; N<u>Ph</u>SO_2), 10.28 (1H, s; N<u>H</u>SO_2), 7.00 [4H,s; NH<u>Ph</u>CH_3), and 2.13 (3H, s; C<u>H</u>$_3$Ph).

N-[N'-(4-Methoxyphenyl) benzenesulfonamido]-3-carboxy-4-methyl-4-phenyl- 3-butenamide (25)

Pale yellow crystals, m.p. 130°C, 52% yield in m.w, and 72% in thermal. FTIR (KBr): υ (cm^{-1}) =3300-3243(2NH), 3400-2400 (OH; acid), 1705(CO; acid), 1665.5 (CO; amide), 1333 and 1158(SO_2; asy. and sym.). MS: m/z = 480(M^+, 2.3%, $C_{25}H_{24}N_2O_6S$), 278 (33, $C_{13}H_{13}N_2O_3S$), 156 (4.3, C_6H_6NO), 123 (20.4,

C_7H_9NO), 131 (1, $C_{10}H_{11}$), 122(100, C_6H_4NS or C_7H_8NO), 108 (9, C_7H_8O), 92 (10.3, C_6H_6N) and 79 (4, HNO_2S). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.61 (5H,s; Ph), 2.26 (3H, s; PhC(C<u>H</u>$_3$)=C), 3.17 (2H, s; CH2CO), 10.21 (1H, s; NHSO_2), 7.36 (4H, s; NH<u>Ph</u>SO_2)], 9.78 (1H, s; NHCO), 6.94(2H, d; <u>Ph</u>NH), 6.81 (2H, d; <u>Ph</u>OCH_3) and 3.67(3H, s; OCH_3).

N-[N'- (4-Methoxyphenyl) benzenesulfonamido]-3- phenylethylidinepyrrolidine -2,5-dione (26)

Yellow crystals, m.p. 178^{o}C, 40% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) =3254(NH), 1767-1705 (2CO; imide), 1376 and 1160(SO_2; asy. and sym.). MS: m/z = 462 (M^+, 35%, $C_{25}H_{22}N_2O_5S$), 130 (4, $C_{10}H_{10}$), 129 (5.1, $C_{10}H_9$), 123 (8.1, C_7H_9NO), 122 (100, C_7H_8O or C_6H_4NS), 115(8, C_9H_7), 102 (2, C_8H_6) and 96 (1.9, $C_4H_2NO_2$). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.33-7.55 (3H, m; Ph), 7.80-7.85 (2H, m; Ph), 2.33 (3H, s; PhC(C<u>H</u>$_3$)=C), 2.62 (2H, s; C<u>H</u>$_2$CO), 7.44 (4H, s; N<u>Ph</u>SO_2), 10.08 (1H, s; N<u>H</u>SO_2), 7.01-7.06 (2H, d; <u>Ph</u>NH),6.81-6.86 (2H, d; PhOCH_3) and 3.68 (3H, s; OC<u>H</u>$_3$).

N-[N'-(4-Chlorophenyl)benzenesulfonamido]-3-carboxy-4-methyl-4-phenyl- 3-butenamide (27)

White crystals, 200^{o}C, 30% yield in yield in m.w, and 37% in thermal. FTIR (KBr): υ (cm^{-1}) =3300-3243(2NH), 3400-2400 (OH; acid), 1705(CO; acid), 1665.5 (CO; amide), 1333 and 1158(SO_2; asy. and sym.). MS: m/z = 484 (M^+, 5%, $C_{24}H_{21}N_2O_5SCl$) 466 (100, $C_{24}H_{19}N_2O_4SCl$), 340 (55.5, $C_{18}H_{14}NO_4S$), 277 (19, $C_{18}H_{15}NO_2$), 276 (99.4, $C_{18}H_{14}NO_2$), 220 (6.6, $C_{16}H_{14}N$), 157 (9.6, $C_{11}H_9O$), 126 (46, C_6H_5NCl), 129 (25.6, $C_{10}H_9$), 115 (18.8, C_9H_7) and 79 (2, HNO_2S).

N-[N'-(4-Chlorophenyl) benzenesulfonamido]-3-phenylethylidinepyrrolidine-2, 5-dione (28)

Pale yellow crystals, m.p. 110^{o}C, 63% yield in m.w, and 37% in thermal. FTIR (KBr): υ (cm^{-1}) =3241(NH), 1766-1706 (2CO; imide), 1375and 1160(SO_2; asy. and sym.). MS: m/z = 466 (M^+, 100%, $C_{24}H_{19}N_2O_4SCl$), 340 (55.5, $C_{18}H_{14}NO_4S$), 277 (19, $C_{18}H_{15}NO_2$), 276 (99.4, $C_{18}H_{14}NO_2$), 220 (6.6, $C_{16}H_{14}N$), 157 (9.6, $C_{11}H_9O$), 126 (46, C_6H_5NCl),129 (25.6, $C_{10}H_9$), 115 (18.8, C_9H_7) and 79 (2, HNO_2S).

N-[N'-(4-Nitrophenyl) benzenesulfonamido]-3-carboxy-4-methyl-4-phenyl-3-butenamide (29)

Pale yellow crystals, m.p. 119^{o}C, 79% yield in m.w, and 61% in thermal. FTIR (KBr): υ (cm^{-1}) = 3360-3355(2NH), 3400-2400 (OH; acid), 1700(CO; acid), 1690 (CO; amide), 1344 and 1160(SO_2; asy. and sym.). MS: m/z =495 (M^+, 1.8%, $C_{24}H_{21}N_3O_7S$), 294 (4, $C_{18}H_{17}NO_3$), 293 (19, $C_{12}H_{11}N_3O_4S$), 292 (2.5, $C_{12}H_{10}N_3O_4S$), 202(9.2, $C_{12}H_{10}O_3$), 156(100, $C_6H_6NO_2S$), 137(0.9, $C_6H_5N_2O_2$), 131 (3, $C_{10}H_{11}$), 130 (13, $C_{10}H_{10}$), 115 (16.3, C_9H_7) and 92 (87, C_6H_6N).

N-[N'-(1-Naphthyl) benzenesulfonamido]-3-carboxy-4-methyl-4-phenyl-3-butenamide (30)

Pale violet crystals, m.p. 129°C, 53% yield in m.w, and 41% in thermal. FTIR (KBr): υ (cm^{-1}) =3300-3248 (2NH), 3400-2400 (OH; acid), 1700(CO; acid), 1698 (CO; amide), 1399.9 and 1159 (SO_2; asy. and sym.). MS: m/z = 500(M^+, 0%, $C_{28}H_{24}N_2O_5S$), 328 (1.3, $C_{17}H_{16}N_2O_3S$), 327(4, $C_{17}H_{15}N_2O_3S$), 298 (12.7, $C_{16}H_{14}N_2O_2S$), 297 (2.4, $C_{16}H_{13}N_2O_2S$), 143 (45, $C_{10}H_9N$), 142 (27.7, $C_{10}H_8N$), 130 (65, $C_{10}H_{10}$) , 129 (92, $C_{10}H_9$), 115 (100, C_9H_7) and 79 (4, HNO_2S). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.75-7.87 (3H,m; Ph), 8.07(2H, m; Ph), 2.36 (3H, s; PhC(C<u>H</u>$_3$)=C), 3.19 (2H, s; CH_2CO), 10.49 (1H, s; N<u>H</u>SO_2), 7.44 (4H, s; NH<u>Ph</u>SO_2), 10.20 (1H, s; N<u>H</u>CO) and 7.80-7.10 (7H, m; C_8<u>H</u>$_7$).

N-[N'-(1-Naphthyl) benzenesulfonamido]-3-phenylethylidinepyrrolidine-2,5-dione (31)

Violet crystals, m.p. 98°C, 15% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) =3251(NH), 1766.5-1706 (2CO; imide), 1372and 1159(SO_2; asy. and sym.). MS: m/z = 482 (M^+, 0%, $C_{28}H_{22}N_2O_4S$), 328 (14.1, $C_{17}H_{16}N_2O_3S$), 327 (64.7, $C_{17}H_{15}N_2O_3S$), 298 (13.5, $C_{16}H_{14}N_2O_2S$), 297 (4, $C_{16}H_{13}N_2O_2S$), 202(23.8, $C_{12}H_{10}O_3$), 143 (13, $C_{10}H_9N$), 142 (11.4, $C_{10}H_8$), 130 (90.3, $C_{10}H_{10}$), 129 (100, $C_{10}H_9$) and 115 (88, C_9H_7). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.80-7.88 (3H, m; Ph), 8.07 (2H, m; Ph), 2.34 (3H, s; PhC(C<u>H</u>$_3$)=C), 3.01 (2H, s; C<u>H</u>$_2$CO), 7.45 (4H, s; N<u>Ph</u>SO_2), 10.40 (1H, s; N<u>H</u>SO_2) and 7.17-7.62 (7H, m; C_8H_7).

N-(N'-Benzylbenzenesulfonamido)-3-carboxy-4-methyl-4-phenyl-3-butenamide (32)

White crystals, m.p. 153°C, 85% yield in m.w, and 57% in thermal. FTIR (KBr): υ (cm^{-1}) = 3321-3267(2NH), 3400-2400 (OH; acid), 1700(CO; acid), 1680 (CO; amide), 1328 and 1157(SO_2; asy. and sym.). MS: m/z =464 (M^+, 0%, $C_{25}H_{24}N_2O_5S$), 446 (22.4, $C_{25}H_{22}N_2O_4S$), 262 (32, $C_{13}H_{14}N_2O_2S$), 202 (36.5, $C_{12}H_{10}O_3$), 157(9.2, $C_{11}H_9O$), 156(15.2, $C_6H_6NO_2S$), 130 (41.4, $C_{10}H_{10}$), 129 (74, $C_{10}H_9$), 115 (46, C_9H_7), 106 (100, C_7H_8N), 92 (66.4, C_6H_6N) and 79 (14, HNO_2S).

N-(4-Methylphenyl)-3-carboxy-4-methyl-4-phenyl-3-butenamide (33)

White crystals, m.p. 162°C, 0% yield in m.w, and 26% in thermal. FTIR (KBr): υ (cm^{-1}) = 3300 (NH), 3100-2500 (OH acid), 1680 (CO, acid), and 1650 (CO, amide). MS: m/z = 309 (M^+, 25%, $C_{19}H_{19}NO_3$), 291 (22, $C_{19}H_{17}NO_2$), 202 (30, $C_{12}H_{10}O_3$), 130 (25, $C_{10}H_{10}$), 129 (33, $C_{10}H_9$), and 107 (100, C_7H_9N).

N-(4-Methylyphenyl)-3-phenylethylidinepyrrolidine-2, 5-dione (34)

Pale brown crystals, m.p. 118°C, 93% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) = 1756.8-1699 (2CO; imide). MS: m/z = 307 (M^+, 100%, $C_{19}H_{17}NO_3$), 292 (2.6, $C_{18}H_{14}NO_3$), 202 (1.7, $C_{12}H_{12}NO_2$), 149 (33, $C_8H_7NO_2$), 130 (91.2, $C_{10}H_{10}$), 129 (75, $C_{10}H_9$), 115 (53, C_9H_7) and 108 (16, C_7H_8O).

N-(4-Methoxyphenyl)-3-carboxy-4-methyl-4-phenyl-3-butenamide (35)

Gray crystals, m.p. 172°C, 0% yield in m.w, and 45% in thermal. FTIR (KBr): υ (cm^{-1}) = 3298 (NH), 3100-2500 (OH acid), 1698 (CO, acid), and 1663

(CO, amide). MS: m/z = 325 (M^+, 8%, $C_{19}H_{19}NO_4$), 307 (8%, $C_{19}H_{17}NO_3$), 202 (13%, $C_{12}H_{10}O_3$), 130 (31%, $C_{10}H_{10}$), 129 (53%, $C_{10}H_9$), and 123 (100%, C_7H_9NO). ^{1}HNMR (DMSO-d_6): δ (ppm) = 7.31-7.43 (5H, m; Ph), 2.33 (3H, s; PhC(C<u>H</u>$_3$)=C), 3.13 (2H, s; C<u>H</u>$_2$CO), 9.57 (1H, s; N<u>H</u>CO), 7.23-7.25 (2H, d; NH<u>Ph</u>), 6.82 – 6.85 (2H, d; <u>Ph</u>OCH$_3$), and 3.70 (3H, s; OC<u>H</u>$_3$).

N-(4-Methoxyphenyl)-3-phenylethylidinepyrrolidine-2, 5-dione (36)

Gray crystals, m.p. 144°C, 97% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) = 1756.8-1699 (2CO; imide). MS: m/z = 307 (M^+, 100%, $C_{19}H_{17}NO_3$), 292 (2.6, $C_{18}H_{14}NO_3$), 202 (1.7, $C_{12}H_{12}NO_2$), 149 (33, $C_8H_7NO_2$), 130 (91.2, $C_{10}H_{10}$), 129 (75, $C_{10}H_9$), 115 (53, C_9H_7) and 108 (16, C_7H_8O).

N-(4-Chlorophenyl)-3-carboxy-4-methyl-4-phenyl-3-butenamide (37)

White crystals, m.p. 176°C, 0% yield in m.w, and 49% in thermal. FTIR (KBr): υ (cm^{-1}) = 3290 (NH), 3100-2500 (OH; acid), 1695 (CO; acid) and 1660 (CO; amid). MS: m/z = 329 (M^+, 4%, $C_{18}H_{16}NO_3Cl$), 311 (6%, $C_{18}H_{14}NO_2Cl$), 202 (9%, $C_{12}H_{10}O_3$), 130 (27%, $C_{10}H_{10}$), 129 (64%, $C_{10}H_9$), and 127 (100% C_6H_6NCl).

N-(4-Chlorophenyl)-3-phenylethylidinepyrrolidine-2, 5-dione (38)

Brown crystals, m.p. 146°C, 80% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) = 1755-1704 (2CO; imide). MS: m/z = 311 (M^+, 68.7%, $C_{18}H_{14}NO_2Cl$), 158 (9.2, $C_{11}H_{10}O$), 131 (12.5, $C_{10}H_{11}$), 130 (100, $C_{10}H_{10}$), 129 (83.2, $C_{10}H_9$), 127(19, C_6H_6NCl), 125 (7.8, C_6H_4NCl), 115 (70.2, C_9H_7), 111 (5.3, C_6H_4Cl) and 102 (8, C_8H_6).

N-(4-Nitrophenyl)-3-carboxy-4-methyl-4-phenyl-3-butenamide (39)

Yellow crystals, m.p. 182°C, 0% yield in m.w, and 42% in thermal. FTIR (KBr): υ (cm^{-1}) = 3291 (NH), 3100-2500 (OH; acid), 1684 (CO; acid) and 1640 (CO; amid). MS: m/z = 340 (M^+, 0%, $C_{18}H_{16}N_2O_5$), 322 (29%, $C_{18}H_{14}N_2O_4$), 202 (21%, $C_{12}H_{10}O_3$), 138 (54%, $C_6H_6N_2O_2$), and 129 (100%, $C_{10}H_9$)

N-(4-Nitrophenyl)-3-phenylethylidinepyrrolidine-2,5-dione (40)

Brown crystals, m.p. 150°C, 89% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) = 1754.4-1703 (2CO; imide). MS: m/z = 322 (M^+, 5%, $C_{18}H_{14}N_2O_4$), 202 (7, $C_{12}H_{12}NO_2$), 158 (6.6, $C_{11}H_{10}O$), 157 (11.2, $C_{11}H_9O$), 136 (2.3, $C_6H_4N_2O_2$), 130 (100, $C_{10}H_{10}$), 129 (84.6, $C_{10}H_9$), 122 (2.3, $C_6H_4NO_2$), and 115 (85, C_9H_7).

N-(1-Naphthyl)-3-carboxy-4-methyl-4-phenyl-3-butenamide (41)

Pale violet crystals, m.p. 180°C, 19% yield in m.w, and 35% in thermal. FTIR (KBr): υ (cm^{-1}) = 3270 (NH), 3100-2500 (OH; acid), 1700(CO; acid) and 1640 (CO; amid). MS: m/z = 345 (M^+, 5%, $C_{22}H_{19}NO_3$), 327 (3%, $C_{22}H_{17}NO_2$), 202 (5%, $C_{12}H_{10}O_3$), 143 (100%, $C_{10}H_9N$), 130 (15%, $C_{10}H_{10}$), and 129 (27%, $C_{10}H_9$)

N-(1-Naphthyl)-3-phenylethylidinepyrrolidine-2, 5-dione (42)

Dark gray crystals, m.p. 135°C, 71% yield in m.w, and 0% in thermal. FTIR (KBr): υ (cm^{-1}) = 1762-1703 (2CO; imide). MS: m/z =327(M^+, 96.4%, $C_{22}H_{17}NO_2$), 270 (4.1, $C_{20}H_{16}N$), 202 (2, $C_{12}H_{12}NO_2$), 169 (25.5, $C_{11}H_7NO$), 157

(9.5, $C_{11}H_9O$), 158 (7.6, $C_{11}H_{10}O$), 131 (7, $C_{10}H_{11}$), 130 (100, $C_{10}H_{10}$)129 (64, $C_{10}H_9$), 128 (46, $C_{10}H_8$), 127 (23, $C_{10}H_7$) and 115 (53, C_9H_7).

N-Benzyl-3-carboxy-4-methyl-4-phenyl-3-butenamide (43)

White crystals, m.p. 142°C, 92% yield in m.w, and 71% in thermal. FTIR (KBr): υ (cm^{-1}) = 3263 (NH), 3100-2500 (OH; acid), 1700(CO; acid) and 1640 (CO; amid). MS: m/z = 309 (6%, $C_{19}H_{19}NO_3$), 291 (20% $C_{19}H_{17}NO_2$), 202(23.8, $C_{12}H_{10}O_3$), 143 (13, $C_{10}H_9N$), 142 (11.4, $C_{10}H_8$), 130 (90.3, $C_{10}H_{10}$), 202 (9, $C_{12}H_{10}O_3$) 130 (35, $C_{10}H_{10}$), 129 (46, $C_{10}H_9$), 107 (18, C_7H_9N), and 91 (100, C_7H_7).^{1}HNMR (DMSO-d_6): δ (ppm) = 7.13-7.40 (10H, m; 2 Ph), 2.2 (3H, s; C<u>H</u>$_3$), 2.9 (2H, s; C<u>H</u>$_2$CO), 8.99 (1H, s; N<u>H</u>CO), and 4.24 – 4.26 (2H, d, C<u>H</u>$_2$NH).

Conclusion

- In the present work, the comparison shows that the microwave irradiation furnishes the formation of the products in excellent yields, shorter time, enhances cyclization, significantly improves the reaction products quantitatively, qualitatively, economically, and environmentally (green chemistry), much more than thermal heating. Moreover, microwave irradiation reactions show regiospecific property, more than thermal ones.

- The results obtained from microwave irradiation of N-substituted 4-aminobenzenesulfonamides with α, β-unsaturated anhydrides show low reactivity than the corresponding non-sulfonated amines. This low reactivity can be attributed to the presence of bezenesulfonamido group.

General Remarks

Microwave irradiation was carried out in a Galanz Microwave Oven, WP1000AP30-2, Chemistry Department, University College of Women for Arts, Science and Education, Ain Shams University. Spectral measurements were carried out at Micro Analytical Centre, Faculty of Science, Cairo University using:

(a) FTIR PERKIN-ELMER-1430 infrared spectrophotometer; IR spectra.
(b) GCMS QP 1000 EX Shimaedzy; MS spectra.
(c) Varian Gemmi (200 MHz); ^{1}HNMR spectra.

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استخدام كفاءة أشعة الميكروويف فى تشييد بعض البيروليدينات ـ٢، ٥ـ دايون والبيوتينيميدات ومشتقاتهم من السالفوناميد فى وعاء واحد ومقارنتها بالطريقة التقليدية بالتسخين الحرارى

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تم تشييد بعض البيروليدينات ـ٢ ، ٥ ـ دايون والبيوتينيميدات ومشتقاتهم من السالفوناميد نظرا لأهميتهم فى التطبيقات الطبية الواسعة ونشاطهم البيولوجى كمبيدات للبكتريا والفطريات وكمبيدات حشرية ، وذلك بإستخدام طريقة أمنة اقتصادية وصديقة للبيئة (الكيمياء الخضراء) . الهدف من هذا البحث تشييد المركبات (٤-٤٢) عن طريق تكاثف الأنهيدرايدات غير المشبعة فى الوضع ألفا-بيتا (١-٣) مع بعض الأمينات بإستخدام أشعة الميكروويف ومقارنة كفاءتها مع الطريقة التقليديه بالتسخين الحرارى ، وقد أوضحت المقارنة أن تميز طريقة استخدام أشعة الميكروويف على الطريقة التقليدية من حيث قصر مدة إتمام التفاعل بالاضافة إلى تحسن واضح جدا فى الناتج كما وكيفا كما أنها أثبتت قدرتها على انتقائية الناتج ولها دورا كبيرا فى تكوين نواتج حلقية لم يمكن تكوينها بطريقة التسخين التقليدية .

وقد أثبتت مقارنة نتائج تكاثف الأنهيدريدات غير المشبعة فى الوضع ألفا ـ بيتا (١-٣) مع N ـ مشتقات ٤-أمينو بنزين سالفونميد أو الأمينات العادية أن التكاثف مع N ـ مشتقات ٤-أمينو بنزين سالفوناميد أقل نشاطا من الأمينات العادية وأرجع ذلك إلى وجود مجموعة البنزين فى N ـ مشتقات ٤-أمينو بنزين سالفونميد .

ولقد تم اثبات تركيب المركبات الناتجة عن طريق التحليل الطيفى (طيف الكتلة ـ الأشعة تحت الحمراء والرنين النووى المغناطيسى) .