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Synthesis of Arylmethylene-Butanimides and Butenamides and Evaluation of their Biological Activity

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Abstract: Condensation of 2-[methyl (4-methylphenyl) methylene]butandioic anhydride[1] (1) with different hydrazines, semicarbazide or different amines at different temperature in different solvents gives arylmethylenebutanimides and butenamides derivatives (2-16).Some of the products showed biological activity.

Keyword: Hydrazine hydrate, Amines, Unsaturated Anhydrides

INTRODUCTION

Itaconic anhydride reacts with hydrazine hydrate[1,2] to give itaconic hydrazide or its tautomer 3,6-dihydroxy-4-methylene-5,5-dihydropyridazine,whereas with phenylhydrazine it gives N-anilinoitaconimide, and with amines in ethanol at room temperature gives N-substituted itaconamic acids ,which on cyclization with acetic anhydride give N-substituteditaconimides. $^{\alpha}$, $^{\beta}$ -unsaturated anhydrides[3], react with hydrazine hydrate and phenylhydrazine in ethanol to give the hydrazide derivatives. Amidation of the anhydrides with amines gives the corresponding 5-alkyl-N-5-(p-chlorophenyl)-3-oxapent-4-en-imide, γ-phenylitaconic anhydride and its p-methoxy derivatives[4] react with hydrazine hydrate to give biscarbamic acids derivatives. With phenylhydrazine, N-anilinophenylitaconamic acids were obtained and with methyl amine gave itaconimide derivatives.Reaction of them with aromatic amines, gave the corresponding itaconamic acid derivatives, whereas p-toluidine gave p-tolylitaconimide derivatives. The thermal condensation of diphenylmethylenebutanedioic anhydride with aromatic or aliphatic amines[5] gave the corresponding butenamides, pyrrolidine-2,5-diones, respectively.

EXPERIMENTAL

General remark. Analysis were carried out in the Microanalytical Laboratories, Cairo University, and in the Central Laboratory, Ain Shams University.

- FTIR were measured using Nicolet, Magna-IR ™ Spectrophotometer at the Chemistry Department, and College of Women for Arts, Science, and Education of Ain Shams University.
- MS spectra were recorded using GCMS QP 1000 EX Shimadzu at Micro analytical Laboratories, Cairo University.
- ^{1}H-NMR spectra were recorded using Varian Gemini (300 MHz) at Micro analytical Laboratories, Cairo University.
- Biological activity: Antimicrobial screening was performed in the National Research Center, Cairo, Egypt.

General Procedure:

I-Condensation of 2-[methyl (4-methylphenyl) methylene]butandioic anhydride (1) with different hydrazines or amines at different temperatures in different solvents: A mixture of (20mmole) of anhydride(**1**) and (40mmole) of hydrazines or amines in 10 ml of solvent (xylene or ethanol) was stirred at room temperature. Completion of reaction was followed up by thin layer chromatography (TLC) where it was found that it takes place after 15 minutes in xylene and after 120 minutes in ethanol. Reactions also were carried out by heating the reaction mixture for 4 hours at the boiling point of xylene or ethanol.

The reaction mixture was then concentrated and precipitate was filtered and dissolved in chloroform. The chloroform layer was washed with dil. HCl to get rid of unreacted amines, then extracted with 10% ice cold sodium carbonate solution. Acidification the aqueous layer precipitated the carboxylic compounds which are 1,2-bis(3-carboxy-3-butenoyl)hydrazine(**2**) or N-(substituted)-3-carboxy-3-butenamide derivatives (**4,9-15**) whereas, evaporation of the organic layer gave N-(substituted)-2-methyl-enebutanimide derivatives (**3, 6,7,8,16**).

II-Condensation of 2-[methyl (4-methylphenyl) methylene]butandioic anhydride (1) with semicarb-azide hydrochloride (d):

A mixture of semicarbazide hydrochloride (**d**), (40mmol) and anhydrous sodium acetate (40mmol) in 10 ml ethanol was refluxed for 30 minutes. The mixture was filtered off from the precipitated sodium chloride, then 20 mmol of anhydride (**1**) in 5ml ethanol was added to the filtrate and the reaction mixture was refluxed of 2 hours. The separated solid was extracted and worked up similar to the procedure given in I to give N-carbmido-3- [methyl (4-methylphenyl) methylene]butanimide (**5**).Structure of the products were confirmed by spectral analyses, FTIR, ^{1}H-NMR,and MS.

Biological activity: Antimicrobial screening: The antimicrobial screening of the prepared compounds (**2-16**)using the disk diffusion method and inhibition zone diameter (5mm) in different solvents , showed that some derivatives were highly active toward gram-positive and gram-negative bacteria, *Bacillus subtiles* and *Escherichia coli*, respectively but low active toward fungi and yeast , *Asper-gillus niger* and *Candida alibicans*. Butanimides showed higher biological activity than their corresponding butenamides. Moreover, compounds containing the electron-withdrawing carboxylic group, nitro or naphthyl group showed higher biological activities than the others.

RESULTS AND DISCUSSION

In the present work the factors affecting the condensation of anhydride (**1**) with amines were studied such as: specific solvent, temperature effects, basicity of amines and structural effect of anhydride. Formation of 1,2-bis[3-carboxy-4-methyl-4-(4-methylphenyl) -3-butenoyl]hydrazine (**2**) resulted from the condensation of 2-[methyl(4-methylphenyl)methylene] butandioic anhydride (**1**) with hydrazine hydrate (**a**) in ethanol or xylene at room temperature or at their boiling points indicates that the two molecules of anhydride add to one molecule of amine.

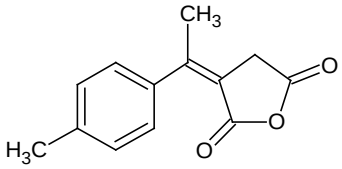
The difficulty of cyclization to bis-imide can be attributed to the steric factor and the electronic effect of electron donating methyl group in the anhydride. Condensation of anhydride (**1**) with phenyl- hydrazine (**b**) at room temperature , boiling ethanol or xylene gave N-anilino-3-[methyl(4-methylphenyl) methylene] butanimide (**3**) indicating the role of the basic nitrogen atom in the amido group towards intramolecular nucleophilic attack to form the cyclic imide. With 2, 4-dinitrophenylhydrazine(**c**), anhydride (**1**) gave N-(2,4-dinitroanilino)-3-carboxy-4-methyl-4-(4-methy- lphenyl) - 3-butenamide (**4**) in boiling xylene .

The formation of the butenamide derivative can be ascribed to the low nucleophilicity of the amido nitrogen atom toward further intramolecular nucleophilic attack on the carbonyl carbon to give the corresponding butanimide derivatives. The difference in reaction temperature is due to the difference in amines basicity. Condensation of 2-[methyl(4-methylphenyl)methylene]butandioic anhydride (**1**) with semicarbazide hydrochloride (**d**) take place only in presence of anhydrous sodium acetate and in boiling ethanol to give N-carba-mido-3-[methyl(4-methylphenyl)methylene]butanimide(**5**).

The reaction could not be carried out in xylene, due to the lack of solubility of semicarbazide hydrochloride in it. The results obtained from condensation of anhydride (**1**) with strong basic aliphatic amines, methyl-amine (**e**),n-butylamine (**f**) or isobutylamine (**g**) in ethanol or xylene either at room temperature or at their boiling point indicate to the high nucleophilicity of the amidonitrogen atom toward cyclization to form butanimide derivatives.

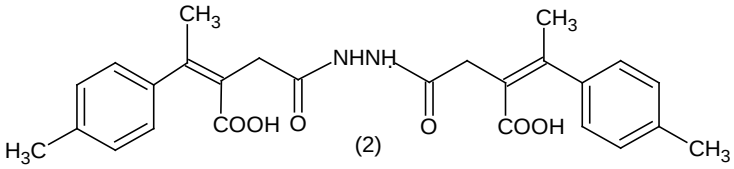
N-methyl-3-[methyl (4-methylphenyl) methylene]butanimide (**6**),N-butyl-3-[methyl(4-meth- y lphenyl) methylene]butanimide (**7**), and N-isobutyl-3-[methyl(4-methylphenyl) methylene] butanimide (**8**) were obtained respectively. With the weak basic aliphatic amine, benzyl amine (**h**), anhydride (**1**) gave N-benzyl-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide (**9**) where no intramolecular nucleophilic attack could take place. Reaction with benzylamine took place only in boiling xylene.

However, reaction with aniline (**i**) , p-toluidine (**j**), p-chloroaniline (**k**) and 1-naphthylamine (**l**) at room temperature, boiling ethanol or xylene ,and reaction with p-aminobenzoic acid (**m**) in boiling ethanol, or with p-nitroaniline (**n**) in boiling xylene gave N-substituted-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide(**10-15**) respectively. While with p-anisidine (**o**), N-(4-methoxphenyl)-3-[methyl (4-methyl phenyl) methy lene] butanimide (**16**) was obtained.



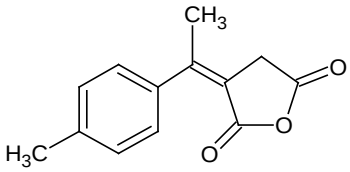
(1)

$NH_2NH_2.H_2O$
(a)



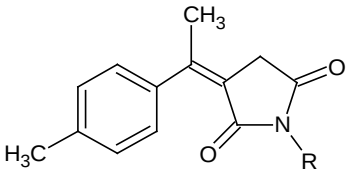
(2)

Scheme (1)

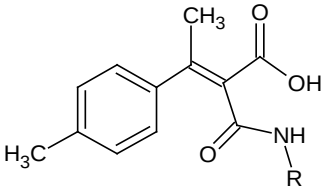


(1)

RNH_2
(b,d - g ,o)



RNH_2
(c,h - n)



Comp.	R
3	NHPh
5	$NHCONH_2$
6	Me
7	n-Bu
8	i-Bu
16	$4\text{-}CH_3OC_6H_4$

Comp.	R
4	$2,4\text{-}(NO_2)_2C_6H_3$
9	$C_6H_4CH_2$
10	C_6H_5
11	$4\text{-}CH_3C_6H_4$
14	$4\text{-}HOOCC_6H_4$
12	$4\text{-}ClC_6H_4$
15	$4\text{-}NO_2C_6H_4$
13	1-Naphthyl

Scheme (2)

Table-1: Elemental Analysis for compounds (**2-16**) and comparison of yields of products resulting in Ethanol and xylene.

Comp. No.	Name	Solvent of crystallization	Yield%		Analysis (Calculated/Found)			
			Ethanol	Xylene	C	H	N	Cl
2	1,2-Bis[3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenoyl]hydrazine	Benzene	28	60	67.20 67.80	6.07 7.02	6.03 6.03	- -
3	N-Anilino-3-[methyl(4-methylphenyl) methylene] butanimide	Ethanol	42	88	74.49 74.00	5.92 5.00	9.14 8.50	- -
4	N-(2,4-Dinitroanilino)-3-carboxy-4-methyl-4-(4-methylphenyl) 3-buten-amide	Ethanol	-	42	55.08 54.99	4.37 4.65	13.5 13.9	- -
5	N-Carbmido-3-[methyl(4-methylphenyl) methylene]butanimide	Ethanol	79	-	61.53 62.09	5.53 5.43	15.4 16.1	- -
6	N-Methyl-3-[methyl(4-methylphenyl)methylene]butanimide	Ethanol	44	85	73.35 74.30	6.59 6.46	6.11 5.51	- -
7	N-Butyl-3-[methyl(4-methylphenyl)methylene]butanimide	-	40	47	75.25 74.56	7.79 7.12	5.16 5.73	- -
8	N-Isobutyl-3-[methyl(4-methylphenyl)methylene] butanimide	-	50	55	75.25 74.55	7.79 7.35	5.16 5.30	- -
9	N-Benzyl-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Ethanol	-	36	74.29 74.50	6.54 5.70	4.33 4.27	- -
10	N-Phenyl-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Ethanol	20	28	73.77 73.80	6.19 5.40	4.52 4.00	- -

Comp. No.	Name	Solvent of crystallization	Yield%		Analysis (Calculated/Found)			
			Ethanol	Xylene	C	H	N	Cl
11	N-(4-Methylphenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Benzene	40	53	74.29 74.62	6.54 7.00	4.33 5.30	- -
12	N-(4-Chlorophenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Ethanol	63	88	66.38 67.20	5.27 5.10	4.07 4.12	10.3 10.8
13	N-(1-Naphthyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Ethanol	36	38	76.86 77.14	5.88 5.82	3.89 3.71	- -
14	N-(4-Carboxyphenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Ethanol	65	-	67.98 68.30	5.42 4.71	3.96 4.17	- -
15	N-(4-Nitrophenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide	Ethanol	-	17	64.41 64.65	5.12 4.30	7.91 7.20	- -
16	N-(4-Methoxyphenyl)-3-[methyl(4-methylphen-yl)methylene]butanimide	Ethanol	47	58	74.75 75.40	5.95 5.00	4.35 3.80	- -

Table.2: Characteristics of synthetic compounds

Compound 2	**1,2-Bis[3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenoyl]hydrazine:**
General property	White crystals, mp 182 ^{0}C, 28% yield in ethanol and 60% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3350 & 3280(NH,amide),3100-2500 (OH,acid), 1700(CO,acid) and 1650 (CO,hydrazide).
MS:m/z	464(M^+,0%,$C_{26}H_{28}N_2O_6$),280 (0.5,$C_{12}H_{12}O_6N_2$),201(0.4,$C_{12}H_{11}NO_2$), 115(4.8,C_9H_{11}) and 91(100,C_7H_7).
Compound 3	**N-Anilino-3-[methyl (4-methylphenyl) methylene]butanimide:**
General property	Yellow crystals, mp 189^{0}C, 42% yield in ethanol and 88% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3300(NH,imide),1700&1650(2CO,imide).

MS:m/z	306(M^+,53%,$C_{19}H_{18}N_2O_2$),215(2.8,$C_{12}H_{11}N_2O_2$),128 (42,$C_{10}H_8$), 93(100,C_6H_7N) and 91(17,C_7H_7).
^{1}H-NMR(DMSO-d_6)	δ(ppm) =2.35 (3H,s,CH_3), 6.7 (2H,d,CH_3ph), 7.15-7.17 (2H,d,CH_3Ph), 2.6(3H, s,CH_3), 3.4 (2H,s,CH_2), 6.7 (1H,t,NHPh), 7.2-7.3(2H,t,NHPh),and7.14-7.18(3H,m,NHPh).
Compound 4	**N-(2,4-Dinitroanilino)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	Yellow crystals, mp 180 ^{0}C ,0% yield in ethanol and 42% yield in xylene.
FTIR (KBr): υ(cm^{-1})	FTIR(KBr):υ(cm^{-1})=3360&3270(2NH,amide),3100-2500(OH,acid), 1695(CO,acid)and1675(CO,hydrazide)
MS:m/z	MS:m/z=414(M^+,0%,$C_{19}H_{18}N_4O_7$),396(13,$C_{19}H_{16}N_4O_6$),198 (14.9,$C_6H_6N_4O_4$), 171(40,$C_{12}H_{11}O$) and 129 (100,$C_{10}H_9$).
Compound 5	**N-Carbmido-3-[methyl(4-methylphenyl)methylene]butanimide:**
General property	White crystals, mp 246 ^{0}C ,79.4% yield in ethanol and 0 % yield in xylene.
FTIR (KBr): υ(cm^{-1})	3430 (NH, imide) , 3300&3170(NH_2,amide),1710&1690(2CO,imide).
MS:m/z	273(M^+, 0%,$C_{14}H_{15}N_3O_3$),230(100,$C_{13}H_{14}N_2O_2$),115(22, C_2H_5 N_3O_2) ,and 91(16.5,C_7H_7).
Compound 6	**N-Methyl-3-[methyl(4-methylphenyl)methylene]butanimide:**
General property	Pale yellow needles, mp 103 ^{0}C ,44.5% yield in ethanol and 85 % yield in xylene.
FTIR (KBr): υ(cm^{-1})	1690&1660(2CO,imide).
MS:m/z	229(M^+,100%,$C_{14}H_{15}NO_2$),144(76,$C_{11}H_{12}$),129(75.5,$C_{10}H_9$),91(10,C_7H_7)and85(17.7,$C_3H_3NO_2$)
^{1}H-NMR(DMSO-d_6)	δ(ppm)=2.3(3H,s,CH_3), 7.2-7.24(2H,d,CH_3Ph),7.27-7.3(2H,d,CH_3Ph),2.6(3H,s,CH_3),3.2(2H,s,CH_2) and 2.9 (3H,s,CH_3).
Compound 7	**N-Butyl-3-[methyl(4-methylphenyl)methylene]butanimide:**
General property	Yellowish brown vis-cous oil,40% yield in ethanol and 47 % yield in xylene
FTIR (KBr): υ(cm^{-1})	1698&1656(2CO,imide)
^{1}H-NMR($CDCl_3$)	δ(ppm)=2.37(3H,s,CH_3),7.1-7.14(2H,d,CH_3Ph),7.19-7.2(2H,d,CH_3Ph),2.6(3H,s,CH_3), 3.17(2H,s,CH_2),3.5-3.6(3H,t,CH_2),1.53-1.61(2H,quin,CH_2),1.27-1.4 (2H,imp.,CH_2), and (3H,t,CH_3)
Compound 8	**N-Isobutyl-3-[methyl(4-methylphenyl)methylene]butanimide:**
General property	Brown viscous oil,65% yield in ethanol and 77% yield in xylene.
FTIR (KBr): υ(cm^{-1})	1698&1656(2CO,imide)
^{1}H-NMR($CDCl_3$)	δ(ppm)=2.37(3H,s,CH_3),7.1-7.14(2H,d,CH_3Ph),7.23-7.3 (2H,d,CH_3Ph), 2.6 (3H,s,CH_3), 2.7(2H,s,CH_2),3.16-3.17(3H,d,CH_2),1.56-1.64(1H,m,CH),and0.91-0.94 (6H,d,2CH_3).
Compound 9	**N-Benzyl-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	White crystals, mp 180^{0}C ,0% yield in ethanol and 36% yield in xylene
FTIR (KBr): υ(cm^{-1})	3330(NH,amide), 3100-2500(OH,acid)1698(CO,acid)and1642 (CO,amide).
MS:m/z	323(M^+,0%,$C_{20}H_{21}NO_3$),305(9,$C_{20}H_{19}NO_2$),217(2.7,$C_{13}H_{13}O_3$),106(100,C_7H_8N) and 91 (71.4,C_7H_7).
Compound 10	**N-Phenyl-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	White crystals, mp 210^{0}C,36.5% yield in ethanol and 45% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3295(NH,amide),3100-2500(OH,acid)1690(CO,acid)and1650(CO,amide)

MS:m/z	309(M^+,3.5%,$C_{19}H_{19}NO_3$),291(7,$C_{19}H_{17}NO_2$),129(38,$C_{10}H_9$),and 93(100,C_6H_7N).
Compound 11	**N-(4-Methylphenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	White crystals,mp194^{0}C,40% yield in ethanol and 53% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3300(NH,amide),3100-2500(OH,acid),1685(CO,acid)and1660 (CO,amide)
MS:m/z	323(M^+,7.5%,$C_{20}H_{21}NO_3$),305(8,$C_{20}H_{19}NO_2$),129(41.5,$C_{10}H_9$),and 107(100,C_7H_9N).
Compound 12	**N-(4-Chlorophenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	Colorless crystals, mp 192^{0}C,63% yield in ethanol and 88% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3290(NH,amide),3100-2500(OH,acid) ,1690(CO,acid),and1670(CO,amide).
MS:m/z	343(M^+,5.6%,$C_{19}H_{18}NO_3Cl$),325(11.5,$C_{19}H_{16}NO_2Cl$),129 (85.2,$C_{10}H_9$),and 127(100,C_6H_6NCl).
Compound 13	**N-(1-Naphthyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	violet crystals, mp 183^{0}C,36% yield in ethanol and 38.5% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3290(NH,amide),3100-2500(OH,acid),1690(CO,acid),and1670(CO,amide).
MS:m/z	359(M^+,15%,$C_{23}H_{21}NO_3$),143(100,$C_{10}H_9N$),,and 216(22,$C_{13}H_{12}O_3$).
Compound 14	**N-(4-Carboxyphenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	Pale yellow needels ,mp 201^{0}C, 65 % yield in ethanol and 0% yield in xylene
FTIR (KBr): υ(cm^{-1})	3300(NH,amide),3100-2500(OH,acid),1695(CO,acid),and1660(CO,amide).
MS:m/z	353(M^+,0%,$C_{20}H_{19}NO_5$),335(2.4,$C_{20}H_{17}NO4$),120 (100,C_7H_8NO),and 137(77,$C_7H_7O_2N$).
Compound 15	**N-(4-Nitrophenyl)-3-carboxy-4-methyl-4-(4-methylphenyl)-3-butenamide:**
General property	yellow crystals , mp 210^{0}C,0% yield in ethanol and17% yield in xylene.
FTIR (KBr): υ(cm^{-1})	3290(NH,amide),3100-2500(OH,acid),1680(CO,acid),and1640(CO,amide).
MS:m/z	354(M^+,7.7%,$C_{19}H_{18}N_2O_5$),336(55,$C_{19}H_{16}N_2O_4$),138(63,$C_6H_6N_2O_2$),and 129 (100,$C_{10}H_9$).
Compound 16	**N-(4-Methoxyphenyl)-3-[methyl(4-ethylphenyl)methylene]butanimide**
General property	Gray needles, mp 176^{0}C ,47% yield in ethanol and 58% yield in xylene.
FTIR (KBr): υ(cm^{-1})	1700&1640(2CO,imide)
MS:m/z	321(M^+,92%,$C_{20}H_{19}NO_3$),293(43,$C_{18}H_{15}NO_3$),144(76.1,$C_{11}H_{12}$),and130(100,$C_{10}H_{10}$)
^{1}H-NMR (DMSO-d_6):	δ(ppm)=2.35(3H,s,CH_3),7.02-7.03(2H,d,CH_3Ph),7.2-7.21(2H,d,CH_3Ph), 2.6 (3H,s,CH_3),3.36(2H,s,CH_2),7.31-7.34(2H,d,CH_3OPh,7.25-7.28(2H,d,CH_3OPh) and 3.8 (3H,s,OCH_3).

CONCLUSION

In the present work, factors affecting the condensation of α ,β-unsaturated anhydride with amines were studied. These factors are: a) Specific solvent and temperature effects :Most of the reactions behave in similar ways , except few which show different trends and the condensation of the anhydride with different amines in xylene offered higher yields and purity than that reacted in ethanol . b)Basicity of amines : Some amines form 3-carboxy-3-butenamide derivatives , others give 2-methylenebutanimide derivatives. Hydrazine hydrate form the corresponding 1,2-bis (3-carboxy-3-butenoyl)hydrazine. c)Structural effect of anhydride on :i) Condensation reaction : The steric and electronic factor exerted by the presence of the sp^3 carbon of the methyl group in the anhydride decreased its reactivity toward

cyclization in case of weak basic amines. ii) Biological activity: Compounds containing the electron-withdrawing groups showed higher biological activities than the others.

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REFERENCES

1. S.M. El-Wahhab and L. El-Assal, *J. Indian Chem. Soc*. 1969, ***46***, 927-936.
2. A. Hiroyosh , *Kogyo Kagaku Zasshi* .1963**, 66**,124-126
3. M. El-Hashash , G.H. Sayed , M. Elkady , *Rev.Roum.Chim.*,1985 , **28 (6)**,659-666
4. S. M.Elwahab, Sh. M Abdallah , F. A Elmaria , E. A Mansour, Rev. *Roum. Chim.* 2003, ***48***,619-627
5. Sh. M Abdallah , H. M Hefny ,Turk J Chem. 2011 , **35**, 463 – 474

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