Heterocycles. Part V [1a,b]. Reaction of α,β-Unsaturated Carbonyl Compounds with Arylacetamides. A Synthesis of 2-Pyridone Derivatives

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The reaction of 1,3-diaryl-2-propene-1-ones I with arylacetamides II, in the presence of sodium ethoxide under reflux, for two hours, gave the corresponding 3,4,6-triaryl-3,4-dihydro-2(1*H*)-pyridones IV. However, when the reaction of these ketones was carried out in the presence of sodium hydride, they gave the corresponding 3,4,6-triaryl-2(1*H*)-pyridones VI or a mixture of IV and VI. When 1,3-diaryl-2-propyne-1-ones V were reacted with arylacetamides, in the presence of sodium hydride, they yielded the corresponding 2-pyridones VI. Treatment of compounds IV with selenium produced the corresponding 2-pyridones VI. Acetylation of the latter compounds gave the corresponding 2-acetyl derivatives VIII. The structure of all products was confirmed by chemical and spectroscopic evidence, and the mechanism of the reactions was discussed.

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The reaction of α,β-unsaturated ketones with malonamide [3] and cyanoacetamide [4-9] have been reported to give the corresponding 2-pyridones. It was assumed that the reactants undergo Michael additions, followed by cyclization to form pyridone derivatives. The present investigation was intended to study the reaction of α,β-unsaturated ketones, ethylenic and acetylenic, with arylacetamides, to establish the structure of the products, and to throw further light on the reaction mechanism. Thus, when 1,3-diaryl-2-propene-1-ones Ia-ℓ were refluxed with arylacetamides

Scheme 1

Compound	Ar	Ar'	Ar''
I,IV,VIa	C_6H_5	C_6H_5	C_6H_5
I,IV,VIb	C_6H_5	p-$CH_3C_6H_4$	C_6H_5
I,IV,VIc	C_6H_5	p-ClC_6H_4	C_6H_5
I,IV,VId	C_6H_5	m-$CH_3OC_6H_4$	C_6H_5
I,IV,VIe	C_6H_5	p-BrC_6H_4	C_6H_5
I,IV,VIf	C_6H_5	C_6H_5	p-ClC_6H_4
I,IV,VIg	p-ClC_6H_4	C_6H_5	p-ClC_6H_4
I,IV,VIh	C_6H_5	C_4H_3S	C_4H_3S
I,IVi	C_6H_5	m-ClC_6H_4	C_6H_5
I,IVj	C_6H_5	p-$CH_3OC_6H_4$	C_6H_5
I,IVk	C_6H_5	3,4-$OCH_2C_6H_3$	C_6H_5
I,IVl	p-ClC_6H_4	p-ClC_6H_4	C_6H_5
IIa	C_6H_5	—	
IIb	p-ClC_6H_4	—	
IIc	p-$CH_3OC_6H_4$	—	
Va	—	C_6H_5	C_6H_5
Vb	—	C_6H_5	p-ClC_6H_4
Vc	—	C_6H_5	p-$CH_3OC_6H_4$
VIi	C_6H_5	p-$CH_3OC_6H_4$	C_4H_3S
VIj	p-$CH_3OC_6H_4$	C_6H_5	p-ClC_6H_4
VIk	p-ClC_6H_4	C_6H_5	C_6H_5
VIl	p-ClC_6H_4	C_6H_5	p-$CH_3OC_6H_4$
VIm	C_6H_5	C_4H_3S	p-ClC_6H_4
VIn	C_6H_5	p-ClC_6H_4	C_4H_3S
VIIIa	C_6H_5	C_6H_5	p-ClC_6H_4
VIIIb	C_6H_5	C_4H_3S	p-ClC_6H_4

Scheme I

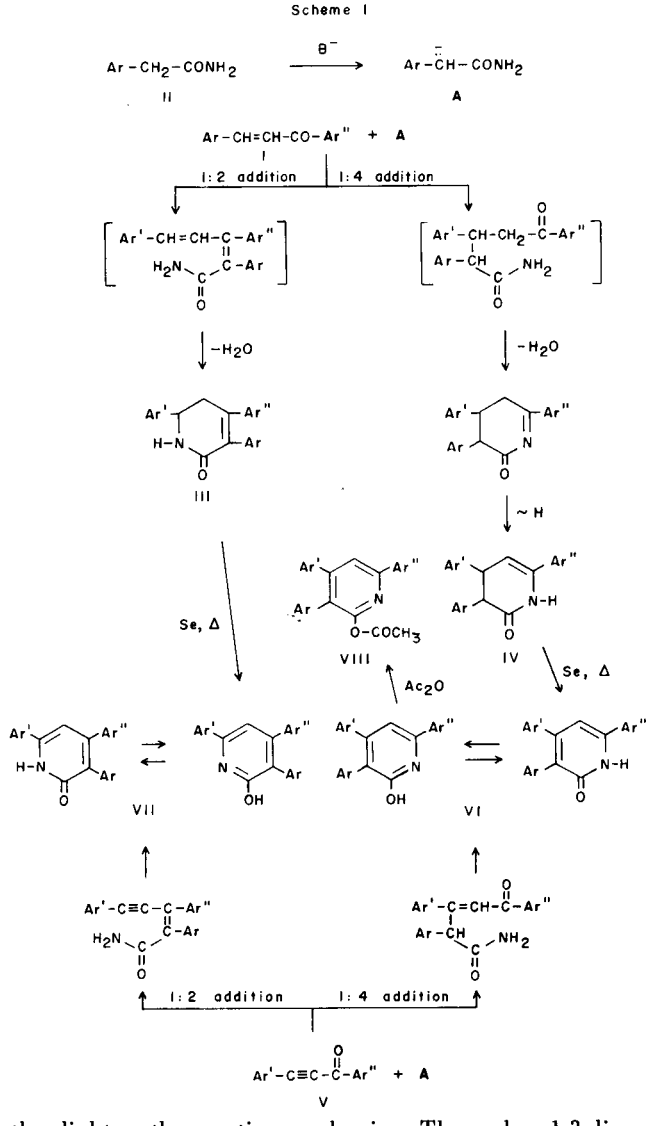


Table 1

Infrared, Electronic and Nuclear Magnetic Resonance Spectral Data of IVa-l

	Infrared Spectra (Potassium Bromide)		Electronic Spectra (Ethanol)		NMR (Deuteriochloroform + DMSO-d_6)	
Compound	cm^{-1}	ν	λ max (nm)	log ϵ	δ	Assignments (No. of Protons)
IVa	3215 (br)	NH	338-342	2.69	8.27 (br)	(1) NH
	1680 (s)	C=O	274-276	3.65	7.47-7.0 (m)	(15) ArH
	1655 (s)	C=C	216	4.20	5.57 (d)	(1) =CH
					3.93 (m)	(2) >CH-CH=
IVb	3250 (br)	NH	270	3.70	8.00 (br)	(1) NH
	1680 (s)	C=O	216-218	4.15	7.47-7.0 (m)	(14) ArH
	1649 (s)	C=C			5.53 (d)	(1) =CH
					3.90 (m)	(2) >CH-CH=
					2.23 (s)	(3) CH_3
IVc	3250 (br)	NH	342	2.91	10.07 (br)	(1) NH
	1680 (s)	C=O	258-274	3.64	7.62-7.0 (m)	(14) ArH
	1645 (s)	C=C	219	4.11	5.47 (d)	(1) =CH
					3.93 (m)	(2) >CH-CH=
IVd	3220 (br)	NH	275	3.82	8.0 (br)	(1) NH
	1678 (s)	C=O	210-212	4.31	7.5-6.57 (m)	(14) ArH
	1652 (m)	C=C			5.57 (d)	(1) =CH
					3.93 (m)	(2) >CH-CH=
					3.67 (s)	(3) OCH_3
IVe	3220 (br)	NH	342	3.09	8.07 (br)	(1) NH
	1685 (s)	C=O	250-270	3.89	7.50-6.93 (m)	(14) ArH
	1650 (m)	C=C	218	4.49	5.53 (d)	(1) =CH
					3.87 (m)	(2) >CH-CH=
IVf	3220 (br)	NH	284	3.63	10.27 (br)	(1) NH
	1678 (s)	C=O	244	3.86	8.0-7.13 (m)	(14) ArH
	1645 (m)	C=C			5.73 (d)	(1) =CH
					4.0 (m)	(2) >CH-CH=
VIg	3210 (br)	NH	280	3.98	10.17 (br)	(1) NH
	1680 (s)	C=O	242	4.29	8.00-6.87 (m)	(13) ArH
	1645 (m)	C=C			5.67 (d)	=CH
					4.13 (m)	>CH-CH=
IVh	3220 (br)	NH	363	3.24	8.22 (br)	(1) NH
	1680 (s)	C=O	290	4.14	7.92-6.94 (m)	(11) ArH
	1650 (m)	C=C	242	4.38	5.56 (d)	(1) =CH
					3.92 (m)	>CH-CH=
IVi	3230 (br)	NH	324	2.74	8.43 (br)	(1) NH
	1678 (s)	C=O	262-278	3.79	7.5-6.93 (m)	(14) ArH
	1650 (s)	C=C			5.5 (d)	(1) =CH
					3.87 (m)	(2) >CH-CH=
IVj	3220 (br)	NH	280-284	3.92	8.25 (br)	(1) NH
	1677	C=O	278	3.94	7.5-6.6 (m)	(14) ArH
	1645	C=C	220	4.64	5.57 (d)	(1) =CH
					3.87 (m)	(2) >CH-CH=
					3.67 (s)	(3) OCH_3
IVk	3225 (br)	NH	338	3.43	7.97 (br)	(1) NH
	1685 (s)	C=O	286	4.09	7.5-6.57 (m)	(13) ArH
	1652	C=C			5.87 (m)	(2) OCH_2O
					5.53 (d)	(1) =CH
					3.83 (m)	(2) >CH-CH=
IVl	3230 (br)	NH	274-278	3.76	10.3 (br)	(1) NH
	1678 (s)	C=O	240	4.03	8.0-6.93 (m)	(13) ArH
	1648 (m)	C=C			5.67 (d)	(1) =CH
					4.20 (m)	(2) >CH-CH=

IIa-c in the presence of sodium ethoxide for two hours, they gave the corresponding 3,4,6-triaryl-3,4-dihydro-2(1*H*)-pyridones IVa-*l*. However, when the reaction was carried out in the presence of sodium hydride, they afforded, in most cases, a mixture of IV and 3,4,6-triaryl-2(1*H*)-pyridones VI. In some cases the 2-pyridones VI were the sole product. Similarly, aroylarylacetylenes Va-c reacted with arylacetamides II, in the presence of sodium hydride

Table 2

Infrared, Electronic and Nuclear Magnetic Resonance Spectral Data of IVa-n and VIIIa,b

Compound	Infrared Spectra (Potassium Bromide) cm^{-1}	ν	Electronic Spectra (Ethanol) λ max (nm)	log ε	NMR (Deuteriochloroform + DMSO-d_6) δ	Assignments (No. of Protons)
VIa	3150-2700 (br)	NH	344	4.39	7.67-7.0 (m)	(16) ArH + NH
	1629 (s)	C=O	256	4.45	6.57 (s)	(1) =CH-
VIb	3100-2800 (br)	NH	344	4.42	8.0-7.0 (m)	(15) ArH + NH
	1634 (s)	C=O	258	4.54	6.63 (s)	(1) =CH-
VIc	3250-2500 (br)	NH	344	4.12	8.1-7.12 (m)	(15) ArH + NH
	1632 (s)	C=O	254	4.19	6.70 (s)	(1) =CH-
VId	3100-2800 (br)	NH	344	4.21	8.0-6.8 (m)	(15) ArH + NH
	1632 (s)	C=O	254	4.26	6.67 (s)	(1) =CH-
					3.57 (s)	(3) -CH_3
VIe	3200-2500 (Br)	NH	344	4.36	8.05-7.10 (m)	(15) Ar-H + NH
	1632 (s)	C=O	260	4.63	6.71 (s)	(1) =CH-
VIf	3150-2900 (br)	NH	350	4.00	8.0-7.17 (m)	(16) ArH + NH + =CH-
	1629 (s)	C=O	259	4.10		
VIg	3200-2200 (br)	NH	354	4.09	8.0-7.0 (m)	(15) ArH + NH + =CH-
	1622 (s)	C=O	260	4.19		
VIh	3050-2400 (br)	NH	369	4.09	7.33-6.67	(15) ArH + NH + =CH-
	1630 (s)	C=O	284	4.09		
VIi	3300-2500 (br)	NH	356	4.00	8.2-7.0 (m)	(15) ArH + NH + =CH-
	1623 (s)	C=O	262	4.12	4.0 (s)	(3) -OCH_3
VIj	3200-2400 (br)	NH	356	4.05	8.13-7.07 (m)	(15) ArH + NH + =CH-
	1620 (s)	C=O	244	4.22	4.13 (s)	(3) -OCH_3
VIk	3200-2800 (br)	NH	350	4.34	8.10-7.1 (m)	(14) ArH + NH + =CH-
	1622 (s)	C=O	250	4.37		
VIl	3200-2800 (br)	NH	350	4.06	7.67-7.17 (m)	(15) ArH + NH + =CH-
	1623 (s)	C=O	266	3.97	3.92 (s)	(3) -OCH_3
VIm	3250-2800 (br)	NH	355	4.14	8.33-7.07 (m)	(15) ArH + NH + =CH-
	1627 (s)	C=O	322	4.11		
			274	4.19		
VIn	3200-2700 (br)	NH	363	4.20	8.27-7.2 (m)	(15) ArH + NH + =CH-
	1629 (s)	C=O	276	4.14		
			246	4.11		
VIIIa	1770 (s)	C=O	350	3.66	8.33-7.07 (m)	(15) ArH
	1593 (s)	C=C	286	4.09	2.0 (s)	(3) $OCOCH_3$
			256	4.28		
VIIIb	1764 (s)	C=O	292	4.33	8.07-6.8 (m)	(14) ArH
	1590 (s)	C=C	286	4.39	1.93 (s)	(3) $OCOCH_3$
			266	4.51		

Table 3

3,4,6-Triaryl-3,4-dihydro-2(1*H*)-pyridones IVa-l

Compound No.	Yield (%)	Mp °C	Formula	Calcd. % C	H	N	Cl	Found % C	H	N	Cl
IVa	96	177-178	$C_{23}H_{19}NO$	84.89	5.89	4.30	—	85.00	5.86	4.17	—
IVb	92	205-206	$C_{24}H_{21}NO$	84.92	6.24	4.13	—	85.05	6.21	4.03	—
IVc	98	214-215	$C_{23}H_{18}ClNO$	76.77	5.04	3.89	9.85	77.01	5.01	3.79	9.77
IVd	91	169-170	$C_{24}H_{21}NO_2$	81.10	5.96	3.94	—	81.22	5.97	4.04	—
IVe	88	210-211	$C_{23}H_{18}BrNO$	68.33	4.49	3.46	19.76 (Br)	68.23	4.55	3.29	19.53 (Br)
IVf	92	180-181	$C_{23}H_{18}ClNO$	76.77	5.04	3.89	9.85	76.83	5.06	3.95	10.01
IVg	89	235-237	$C_{23}H_{17}Cl_2NO$	70.06	4.35	3.58	17.99	69.86	4.19	3.45	17.91
IVh	90	156-157	$C_{19}H_{14}NOS_2$	68.06	3.88	4.18	19.10 (S)	68.11	3.79	4.12	18.97 (S)
IVi	94	181-182	$C_{23}H_{18}ClNO$	76.77	5.04	3.89	9.85	76.78	5.01	3.81	9.92
IVj	89	171-172	$C_{24}H_{21}NO_2$	81.10	5.96	3.94	—	81.29	5.99	3.85	—
IVk	82	189-190	$C_{24}H_{17}NO_3$	78.46	4.66	3.81	—	78.20	5.16	3.72	—
IVl	94	175-177	$C_{23}H_{17}Cl_2NO$	70.06	4.35	3.55	17.99	70.09	4.29	3.48	17.90

Table 4

3,4,6-Triaryl-2(1*H*)-pyridones VIa-n and the 2-Acetylpyridines VIIIa,b

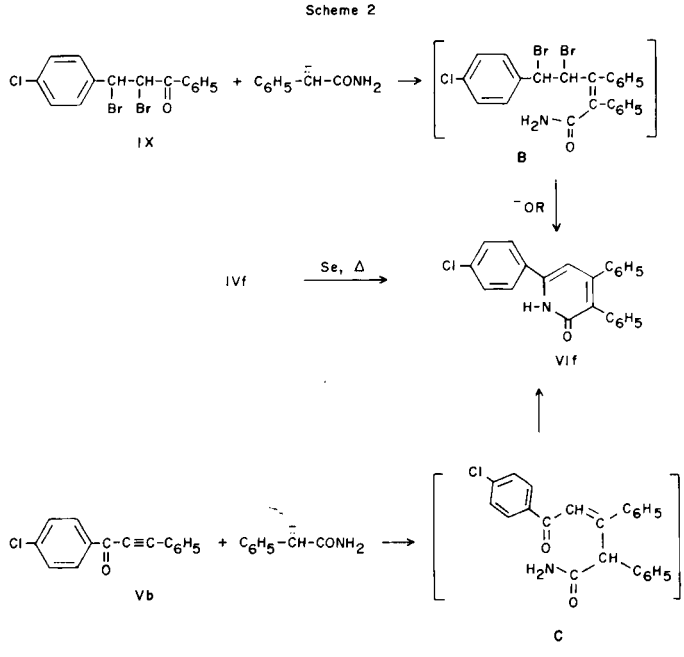
Compound No.	Yield (%)	Mp °C	Formula	Calcd. % C	H	N	Cl	S	Found % C	H	N	Cl	S
VIa	98	311-312	$C_{23}H_{17}NO$	85.42	5.30	4.33	—	—	85.48	5.41	4.21	—	—
VIb	93	272-273	$C_{24}H_{19}NO$	85.43	5.68	4.15	—	—	85.32	5.71	4.32	—	—
VIc	95	299-300	$C_{23}H_{16}ClNO$	77.20	4.51	3.91	9.91	—	77.32	4.55	3.93	10.12	—
VId	95	263-264	$C_{24}H_{19}NO_2$	81.56	5.42	3.96	—	—	81.71	5.40	3.78	—	—
VIe	94	312-313	$C_{23}H_{16}BrNO$	68.67	4.01	3.48	19.86	—	68.71	4.05	3.52	20.01	—
VIf	89	285-286	$C_{23}H_{16}ClNO$	77.20	4.51	3.91	9.91	—	77.28	4.50	3.86	10.21	—
VIg	95	298-300	$C_{23}H_{17}Cl_2NO$	70.06	4.34	3.55	17.98	—	70.21	4.29	3.48	17.85	—
VIh	93	298-299	$C_{19}H_{13}NOS_2$	68.03	3.90	4.17	—	19.18	68.04	3.81	4.10	—	19.26
VIi	91	286-287	$C_{22}H_{17}NO_2S$	73.51	4.76	3.90	—	8.92	73.48	4.59	3.82	—	8.89
VIj	87	316-317	$C_{24}H_{18}ClNO_2$	74.32	4.67	3.61	9.14	—	74.28	4.53	3.59	9.21	—
VIk	93	293-294	$C_{23}H_{16}ClNO$	77.20	4.50	3.91	9.90	—	77.12	4.43	3.82	9.75	—
VIl	92	296-297	$C_{24}H_{18}ClNO_2$	74.32	4.67	3.61	9.14	—	74.19	4.48	3.57	9.05	—
VIm	90	315-316	$C_{21}H_{14}ClNOS$	69.32	3.88	3.85	9.74	8.81	69.17	3.79	3.75	9.66	8.73
VIn	88	339-340	$C_{21}H_{14}ClNOS$	69.32	3.88	3.85	9.74	8.81	69.46	3.77	3.70	9.52	9.11
VIIIa	95	300-302	$C_{25}H_{18}ClNO_2$	75.09	4.53	3.50	8.86	—	74.92	4.50	3.48	8.79	—
VIIIb	93	267-269	$C_{23}H_{16}ClNO_2S$	68.06	3.97	3.45	8.73	7.90	67.93	3.88	3.41	8.62	—

to give the corresponding 2-pyridones VIa-c.

The structure of the 2-pyridone derivatives IV and VI was established by both spectroscopic and chemical evidence. Thus, the infrared spectra of the 2-pyridones IVa-ℓ (Table 1), show absorptions in the 3250-3215 cm^{-1} (broad), 1685-1677 cm^{-1} and 1655-1645 cm^{-1} regions which are correlated to the -NH, C=O, and the C=C stretching frequencies of these compounds [9]. Further support for the assigned structures comes from the nmr spectra (Table 1). They show a multiplet in the region δ 4.20-3.83 and a doublet in the region δ 5.73-5.47 which correspond to the protons at carbons 3, 4 and carbon 5, respectively. The spectra also show broad signals in the region δ 10.30-7.97 which can be attributed to the NH proton, and disappear after adding deuterium oxide [10]. The uv spectra of these compounds lend further support to the proposed structures (Table 1). They show absorption bands attributable to $\pi \rightarrow \pi^*$ transition of the styrene moiety [9]. The mass spectra of these compounds IVa-ℓ afforded further evidence and showed peaks corresponding to their molecular ions.

The chemical behaviour of these compounds IV is also in good agreement with the assigned structure. Thus, upon heating with Selenium at 200-210° for one hour, they gave the corresponding 3,4,6-triaryl-2(1*H*)-pyridones VIa-ℓ in good yields, (*cf.* Scheme 1). The latter products are identical with those obtained from the reaction of acetylenic ketones V with the arylacetamides II in the presence of sodium hydride as a base. The structure of compounds VI was also inferred from their spectral and chemical behaviour. Thus, their infrared spectra show strong bands in the region 1634-1620 cm^{-1}, which can be ascribed to the unsaturated pyridone system. The broad bands in the region 3250-2500 cm^{-1} are attributed to the bonded NH [10].

Scheme 2



Their nmr spectra, show a singlet in the region δ 6.70-6.3 which can be assigned to the proton at C_5. The ultraviolet spectra show strong resemblance to each other which reflects their structural identity [9]. The mass spectra of VI gave peaks which correspond to their molecular ions.

The structure of the triaryl-2-pyridones VI was also supported by their reaction with acetic anhydride to produce the corresponding 2-acetyl derivatives VIII, indicating the presence of the enol form of compounds VI.

Information concerning the mechanism of the above reactions are derivable from the reaction of 2,3-dibromo-3-*p*-chlorophenyl-1-phenylpropan-1-one (IX) with the sodium salt of phenylacetamide. This yielded the corresponding

3,4-diphenyl-6-chlorophenyl-2(1*H*)-pyridone (VIf). This reaction proceeds by a 1:2-addition to produce the intermediate **B**, followed by cyclization (*cf.* Scheme 2). The same product was also obtained by reacting Vb with phenylacetamide using sodium hydride as a base (*cf.* Scheme 2). This leads to the conclusion that the Michael acceptor Vb underwent a 1:4-addition to give VIf. The fact that VIf can be only obtained from the dehydrogenation of a compound having the structure IV and not III, indicates that α,β-ethylenic ketones reacted with arylacetamides, through the Michael addition (1:4-addition) of the carbanion **A** to the ketone, to produce the corresponding 2-pyridone (*cf.* Scheme 1).

It can be argued at this respect that both α,β-ethylenic or acetylenic ketones, undergo a Michael addition with the arylacetamide carbanions **A**, to produce the compounds IV and VI, respectively.

EXPERIMENTAL

Melting points are uncorrected. Ultraviolet and infrared spectra were measured on Cary 17 and Perkin Elmer 580 B spectrophotometers, respectively. The nmr spectra were run on Varian T 60 A, using TMS as the internal standard. The ms were carried out using Varian MAT 311A. The purity of the analytical samples was checked by tlc (silica gel). Microanalysis were determined by Alfred Bernhardt, West Germany and by Central Analytical Laboratory (KISR), Kuwait.

Reaction of 1,3-Diaryl-2-propen-1-ones I with Arylacetamides, II. General Procedure.

a) In the Presence of Sodium Ethoxide.

1,3-Diaryl-2-propene-1-one I (0.03 mole) was added to a solution of sodium ethoxide (0.03 mole) and arylacetamide (0.03 mole) in absolute ethanol (150 ml). The reaction mixture, which gradually acquired an orange colour, was heated on a boiling water-bath for two hours with occasional stirring. The solvent was evaporated and the residual product was dissolved in water, then acidified with dilute hydrochloric acid (10%). The solid, separated by filtration, was crystallised from methanol to give the corresponding 3,4,6-triaryl-3,4-dihydro-2(1*H*)-pyridones IVa-*ℓ*. These results are reported in Table 3.

b. In the Presence of Sodium Hydride.

A mixture of the amide II (0.1 mole) and sodium hydride (0.1 mole) in dry benzene (70 ml) was stirred until the amide dissolved in the solvent. After about 2 hours, the chalcone I (0.1 mole) was dissolved in 50 ml of dry benzene and added to the reaction mixture. The reaction was initiated by adding a few drops of absolute methanol and the mixture was stirred for a further 5 hours at room temperature. The benzene layer was washed with water and dried. Distillation of benzene left a product which upon crystallization from a hexane/benzene mixture (2:1) gave IV. The insoluble part was crystallized from ethanol to give VI.

Reaction of 1,3-Diaryl-2-propyne-1-ones V with Arylacetamides II. General Procedure.

A mixture of the amide II (0.1 mole) and sodium hydride (0.01 mole) in dry benzene (70 ml) was stirred until the amide was dissolved. The acetylenic ketone V, was dissolved in dry benzene (50 ml) and added to the above mixture, then worked up as mentioned above. The product was crystallized from ethanol to give VI. The results are tabulated in Table 4.

Dehydrogenation of 3,4,6-Triaryl-3,4-dihydro-2(1*H*)-pyridones Va-*ℓ*. General Procedure.

A mixture of the pyridine derivatives IVa-*ℓ* (2.0 g) and selenium (0.4 g) was heated on an oil-bath at 200-210° for one hour. The reaction product was cooled, boiled with ethanol and then filtered. The solid product was separated and crystallized from methanol to give the corresponding 2-pyridone derivatives VIa-*ℓ*. In general, the yield is nearly quantitative. The results are reported in Table 4.

Reaction of 2,3-Dibromo-3-*p*-chlorophenyl-1-phenylpropane-1-one (IX) with Phenylacetamide (Ia).

Phenylacetamide (0.01 mole) was treated with sodium ethoxide (0.01 mole) in ethanol (50 ml) to form the phenylacetamide carbanion (**A**). The latter was reacted with (0.01 mole) of the chalcone dibromide IX and the product was worked up as usual [11], to give VIf which was crystallized from methanol.

Action of Acetic Anhydride on the 2-Pyridones VIf,m. General Procedure.

The mixture of the pyridone derivative (1.0 g) and acetic anhydride (3 ml) was refluxed on a water bath for one hour. The cold reaction mixture was worked up as usual [12] to give the corresponding 2-acetoxy-3,4,6-triarylpyridine VIII. The results are reported in Table 4.

REFERENCES AND NOTES

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