A New Approach to the Synthesis of 2-Methyl-4*H*-furo[3,2-*c*][1]benzopyran-4-ones and 2*H*,5*H*-Pyrano[3,2-*c*][1]benzopyran-5-ones

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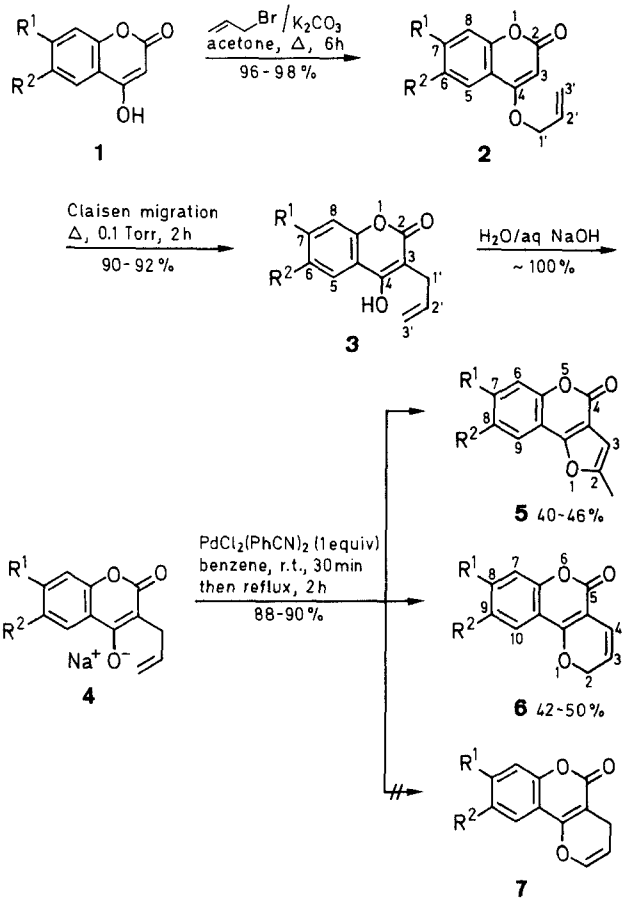
The reaction of various 4-hydroxy-2*H*-[1]benzopyran-2-ones **1** with allyl bromide and potassium carbonate in acetone yielded the corresponding 4-allyloxy derivatives **2**. Claisen rearrangement of **2** gave 3-allyl-4-hydroxy-2*H*-[1]benzopyran-2-ones **3**, which were reacted with sodium hydroxide to give the sodium benzopyranolate salt **4**. Oxidative cyclization of **4** with equimolar quantities of dichlorobis(benzonitrile)palladium gave a 1:1 mixture of 2-methyl-4*H*-furo[3,2-*c*][1]benzopyran-4-ones **5** and 2*H*,5*H*-pyrano[3,2-*c*]-[1]benzopyran-5-ones **6**, in 88–90% combined yield.

The syntheses of various 2-methylfuro fused benzopyranone systems have been described.[1–4] The starting materials are usually allyloxybenzopyranones, which by rearrangement and cyclization give the corresponding 2-methylfuro derivatives in yields ranging from 2–60%. 2-Methylbenzofurans were obtained quantitatively when 2-allylphenols were subjected to oxidative cyclization with dichlorobis(benzonitrile)palladium.[5] This method, with modifications, was previously used by us for the synthesis of 2-phenyl-8-methyl-4*H*-furo[2,3-*h*][1]benzopyran-4-ones,[6] 2-methyl-7*H*-furo[3,2-*g*][1]benzopyran-7-ones and 8-methyl-2*H*-furo[2,3-*h*][1]benzopyran-2-ones,[7] respectively.

We have extended this method, and report herein the oxidative cyclization of sodium 3-allyl-2-oxo-2*H*-[1]-benzopyran-4-olates **4** with dichlorobis(benzonitrile)-palladium (Scheme **A**).

4-Hydroxy-2*H*-[1]benzopyran-2-ones **1a–e** were prepared by the known procedures.[8] Compounds **1a–e** gave 4-allyloxy-2*H*-[1]-benzopyran-2-ones **2a–e** by reaction with allyl bromide/potassium carbonate in refluxing acetone. Claisen migration of **2a–e** by heating *in vacuo* afforded the corresponding 4-hydroxy-3-allyl-2*H*-[1]-benzopyran-2-ones **3a–e** which were converted into their sodium salts **4a–e** by treatment with dilute aqueous sodium hydroxide. The anhydrous sodium salts were suspended in benzene and an equimolar quantity of dichlorobis(benzonitrile)palladium[9] was added and the mixture stirred at room temperature for 30 min and then refluxed for 2 h, after which time metallic palladium began to precipitate. The crude reaction mixture contained: benzonitrile, 2-methyl-4*H*-furo[3,2-*c*][1]-benzopyran-4-ones **5a–e** and 2*H*,5*H*-pyrano[3,2-*c*][1]benzopyran-5-ones **6a–e**, which were separated by column chromatography. The total yield of the reaction products was 80% (calculated from **1a–e**), **5a–e** and **6a–e** are formed in 1:1 ratio. This is in contrast to our previous results where furo derivatives were the sole product.[5–7].

The compounds **2a–e**, **3a–e**, **5a–e** and **6a–e** were characterized by spectral data (Table). In the ^{1}H-NMR spectra of **5**, typified by **5a**,[6] the methyl group at C-2 appeared at $\delta = 2.55$ and H-3 at $\delta = 6.57$. The ^{1}H-NMR spectra of **6**, typified by **6a**, showed peaks at $\delta = 5.18$, 5.53 and 6.57 respectively due to H-2 methylene protons, H-3 and H-4 alkene protons respectively, correlating



1–7	R^1	R^2
a	H	H
b	H	Me
c	H	Cl
d	H	Br
e	Cl	H

Scheme A

well to the values observed in 2*H*-[1]benzopyran.[10] The ^{1}H-NMR data does not support the structure of the alternative compounds **7a–e**.[11] In addition, **6a–c** synthesized, by an alternative route,[12] had identical physical and spectroscopic data to those formed via the oxidative cyclization route presented herein.

The proposed mechanism for the formation of **5a–e** and **6a–e** is shown in Scheme **B**. The first step is the formation of the dimeric-*Π*-allyl complex **A**[6,13] which was converted into isomeric monomers **D** and **G**. Cyclization of the monomer **D** resulted in the compounds **5a–e** through **E**, while cyclization of monomer **G** gave rise to isomeric pyranocoumarins **6a–e** and **7a–e** through **H**. The exclusive formation of **6a–e** by route *a* in preference to **7a–e** by route *b* is due to the facile elimination of H-3 proton which is allylic in character.

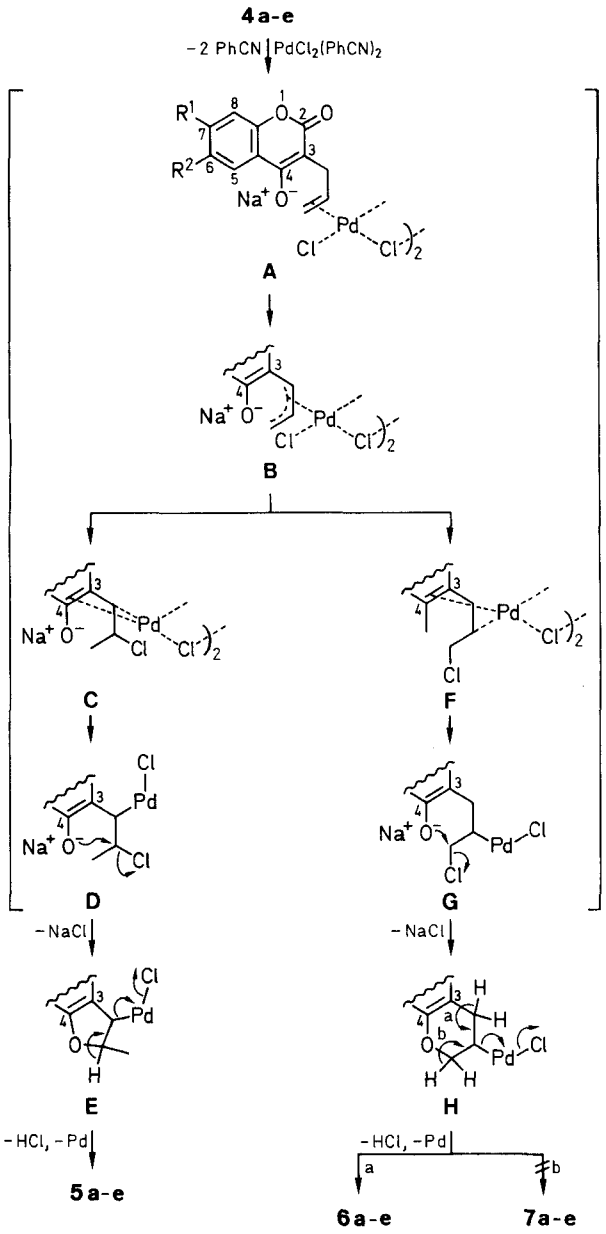
Table. Compounds **2a–e**, **3a–e**, **5a–e**, **6a–e** Prepared

Product	Yield[a] (%)	mp (°C) (solvent)	Molecular Formula[b] or Lit. mp (°C)	IR (KBr) $\nu_{C=O}$ (cm^{-1})	UV (MeOH) λ_{max} (nm) (log ε)	^{1}H-NMR (90 MHz, solvent[c]/TMS) δ, J(Hz)	MS (70 eV) m/z (%)
2a	98	115 (benzene)	115[14]	1720	219 (4.05), 264 (3.94), 275 (3.90), 302 (3.70)	4.63 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 5.50 (m, 2H, H-3′), 5.60 (s, 1H, H-3), 6.10 (m, 1H, H-2′), 7.12–7.60 (m, 3H, H-6, H-7, H-8), 7.86 (dd, 1H, H-5, $J_{5,6}=9$, $J_{5,7}=2$)	202 (M^+)
2b	97	108 (benzene)	108[14]	1715	220 (4.25), 266 (4.06), 276 (3.99), 308 (3.77)	2.40 (s, 3H, CH_3), 4.71 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 5.48 (m, 2H, H-3′), 5.62 (s, 1H, H-3), 6.10 (m, 1H, H-2′), 7.10–7.42 (m, 2H, H-7, H-8), 7.58 (d, 1H, H-5, $J_{5,7}=2$)	216 (M^+)
2c	98	148 (benzene)	$C_{12}H_9ClO_3$ (236.63)	1720	224 (4.24), 266 (3.82), 278 (3.73), 312 (3.57)	4.68 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 5.47 (m, 2H, H-3′), 5.68 (s, 1H, H-3), 6.08 (m, 1H, H-2′), 7.20–7.55 (m, 2H, H-7, H-8), 7.78 (d, 1H, H-5, $J_{5,7}=2$)	236 (M^+)
2d	98	151 (benzene)	$C_{12}H_9BrO_3$ (218.10)	1720	224 (4.29), 226 (3.87), 278 (3.78), 312 (3.59)	4.64 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 5.44 (m, 2H, H-3′), 5.61 (s, 1H, H-3), 6.07 (m, 1H, H-2′), 7.12–7.65 (m, 2H, H-7, H-8), 7.83 (d, 1H, H-5, $J_{5,7}=2$)	280 (M^+)
2e	96	118 (benzene)	$C_{12}H_9ClO_3$ (236.63)	1715	224 (4.22), 264 (3.80), 276 (3.74), 310 (3.56)	4.69 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 5.46 (m, 2H, H-3′), 5.64 (s, 1H, H-3),6.12 (m, 1H, H-2′), 7.12–7.30 (m, 2H, H-6, H-7), 7.72 (d, 1H, H-5, $J_{5,6}=9$)	236 (M^+)
3a	90	131 (Et_2O)	131[15]	1675	215 (4.22), 305 (3.95)	3.38 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 4.97 (m, 2H, H-3′), 5.90 (m, 1H, H-2′), 7.26–7.67 (m, 3H, H-6, H-7, H-8), 7.93 (m, 1H, H-5)	202 (M^+)
3b	90	171 (Et_2O)	$C_{13}H_{12}O_3$ (216.23)	1670	214 (4.22), 312 (3.89)	2.41 (s, 3H, CH_3), 3.37 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 5.01 (m, 2H, H-3′), 5.88 (m, 1H, H-2′), 7.14–7.39 (m, 2H, H-7, H-8), 7.72 (d, 1H, H-5, $J_{5,7}=2$)	216 (M^+)
3c	91	76.5 (Et_2O)	$C_{12}H_9ClO_3$ (236.63)	1670	218 (4.31), 320 (3.79)	3.38 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 4.94 (m, 2H, H-3′), 5.89 (m, 1H, H-2′), 7.27–7.67 (m, 2H, H-7, H-8), 7.87 (d, 1H, H-5, $J_{5,7}=2$)	236 (M^+)
3d	92	153 (Et_2O)	$C_{12}H_9BrO_3$ (281.10)	1675	218 (4.37), 316 (3.86)	3.48 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 4.96 (m, 2H, H-3′), 5.91 (m, 1H, H-2′), 7.14–7.79 (m, 2H, H-7, H-8), 7.85 (d, 1H, H-5, $J_{5,7}=2.0$)	280 (M^+)
3e	90	146 (Et_2O)	$C_{12}H_9ClO_3$ (236.63)	1670	216 (4.26), 314 (3.82)	3.48 (dd, 2H, H-1′, $J_{1',2'}=6$, $J_{1',3'}=2$), 4.98 (m, 2H, H-3′), 5.91 (m, 1H, H-2′), 7.16–7.79 (m, 2H, H-6, H-8), 7.78 (d, 1H, H-5, $J_{5,6}=9$)	236 (M^+)
5a	45	174 (benzene)	174[14]	1730	218 (3.89), 288 (3.75), 313 (3.88), 328 (3.79)	2.55 (d, 3H, furyl CH_3, $J_{CH_3,3}=1$), 6.57 (d, 1H, H-3, $J_{3,CH_3}=1$), 7.38–7.60 (m, 3H, H-6, H-7, H-8), 7.82 (dd, 1H, H-9, $J_{9,8}=8$, $J_{9,7}=2$)	200 (M^+, 100), 199 (M − H, 38), 172 (M − CO, 5), 171 (M − HCO, 15), 144 (M − C_3H_4O, 12), 120 (M − C_5H_4O, 5), 92 (M − $C_6H_4O_2$, 6)

Table. (continued)

Product	Yield[a] (%)	mp (°C) (solvent)	Molecular Formula[b] or Lit. mp (°C)	IR (KBr) $\nu_{C=O}$ (cm^{-1})	UV (MeOH) λ_{max} (nm) (log ε)	^{1}H-NMR (90 MHz, solvent[c]/TMS) δ, J(Hz)	MS (70 eV) m/z (%)
5b	40	156 (benzene)	155[14]	1725	221 (3.95), 292 (3.74), 318 (3.87), 334 (3.78)	2.44 (s, 3H, benzyl CH_3), 2.48 (d, 3H furyl CH_3, $J_{CH_3,3}=1$), 6.55 (d, 1H, H-3, $J_{3,CH_3}=1$), 7.26–7.28 (m, 2H, H-6, H-7), 7.60 (d, 1H, H-9, $J_{9,7}=2$)	214 (M^+, 100), 213 (M − H, 40), 186 (M − CO, 10), 185 (M − HCO, 22), 158 (M − C_3H_4O, 10), 134 (M − C_5H_4O, 8), 106 (M − $C_6H_4O_2$, 8)
5c	46	167 (benzene)	$C_{12}H_7ClO_3$ (234.61)	1740	220 (4.11), 292 (3.69), 318 (3.81), 335 (3.74)	2.49 (d, 3H, furyl CH_3, $J_{CH_3,3}=1$), 6.59 (d, 1H, $J_{3,CH_3}=1$), 7.25–7.40 (m, 2H, H-6, H-7), 7.81 (d, 1H, H-9, $J_{9,7}=2$)	234 (M^+, 100), 206 (M − CO, 10), 205 (M − HCO, 33), 178 (M − C_3H_4O, 10), 154 (M − C_5H_4O, 12), 126 (M − $C_6H_4O_2$, 10)
5d	43	178 (benzene)	$C_{12}H_7BrO_3$ (279.08)	1745	221 (4.02), 294 (3.65), 320 (3.72), 337 (3.70)	2.51 (d, 3H, furyl CH_3, $J_{CH_3,3}=1$), 6.59 (d, 1H, H-3, $J_{3,CH_3}=1$), 7.08 (dd, 1H, H-7, $J_{7,6}=9$, $J_{7,9}=2$), 7.34 (d, 1H, H-6, $J_{6,7}=9$), 7.96 (d, 1H, H-9, $J_{9,7}=2$)	278 (M^+, 100), 250 (M − CO, 5), 249 (M − HCO, 20), 222 (M − C_3H_4O, 10), 198 (M − C_5H_4O, 12), 170 (M − $C_6H_4O_2$, 8)
5e	45	148 (benzene)	$C_{12}H_7ClO_3$ (234.61)	1735	218 (3.70), 295 (3.60), 315 (3.62), 330 (3.54)	2.51 (d, 3H, furyl CH_3, $J_{CH_3,3}=1$), 6.58 (d, 1H, H-3, $J_{3,CH_3}=1$), 7.14–7.49 (m, 2H, H-6, H-8), 7.77 (d, 1H, H-9, $J_{9,8}=10$)	234 (M^+, 100), 206 (M − CO, 12), 205 (M − HCO, 28), 178 (M − C_3H_4O, 8), 154 (M − C_5H_4O, 10), 126 (M − $C_6H_4O_2$, 12)
6a	45	168 (benzene)	$C_{12}H_8O_3$ (200.19)	1705	206 (4.27), 210 (4.24), 250 (3.69), 345 (3.55)	5.18 (dd, 2H, H-2, $J_{2,3}=3$, $J_{2,4}=2$), 5.53 (dt, 1H, H-3, $J_{3,4}=10$, $J_{3,2}=3$), 6.57 (dt, 1H, H-4, $J_{4,3}=10$, $J_{4,2}=2$), 7.32–7.49 (m, 3H, H-7, H-8, H-9), 7.67 (dd, 1H, H-10, $J_{10,9}=10$, $J_{10,8}=2$)	200 (M^+, 88), 199 (M − H, 56), 172 (M − CO, 72), 171 (M − HCO, 44), 144 (M − C_3H_4O, 40), 120 (M − C_5H_4O, 34), 92 (M − $C_6H_4O_2$, 100)
6b	50	144 (benzene)	$C_{13}H_{10}O_3$ (214.21)	1700	205 (4.11), 210 (4.10), 270 (3.56), 310 (3.43)	2.40 (s, 3H, CH_3), 5.10 (dd, 2H, H-2, $J_{2,3}=3$, $J_{2,4}=2$), 5.63 (dt, 1H, H-3, $J_{3,4}=10$, $J_{3,2}=3$), 6.61 (dt, 1H, H-4, $J_{3,4}=10$, $J_{4,2}=2$), 7.11–7.27 (m, 2H, H-8, H-7), 7.52 (d, 1H, H-10, $J_{10,8}=2$)	214 (M^+, 100), 213 (M − H, 45), 186 (M − CO, 34), 185 (M − HCO, 42), 158 (M − C_3H_4O, 45), 134 (M − C_5H_4O, 50), 106 (M − $C_6H_4O_2$, 51)
6c	42	148 (benzene)	$C_{12}H_7ClO_3$ (234.61)	1710	215 (4.23), 270 (3.74), 315 (3.67)	5.13 (dd, 2H, H-2, $J_{2,3}=3$, $J_{2,4}=2$), 5.71 (dt, 1H, H-3, $J_{3,4}=10$, $J_{3,2}=3$), 6.58 (dt, 1H, H-4, $J_{4,3}=10$, $J_{4,2}=2$), 7.21 (d, 1H, H-7, $J_{7,8}=9$), 7.48 (dd, 1H, H-8, $J_{8,7}=9$, $J_{8,10}=2$), 7.68 (d, 1H, H-10, $J_{10,8}=2$)	234 (M^+, 100), 233 (M − H, 45), 206 (M − CO, 30), 205 (M − HCO, 43), 178 (M − C_3H_4O, 40), 154 (M − C_5H_4O, 45), 126 (M − $C_6H_4O_2$, 68)
6d	46	124 (benzene)	$C_{12}H_7BrO_3$ (279.08)	1710	210 (4.21), 265 (3.84), 315 (3.61)	5.13 (dd, 2H, H-2, $J_{2,3}=3$, $J_{2,4}=2$), 5.73 (dt, 1H, H-3, $J_{3,4}=10$, $J_{3,2}=3$), 6.58 (dt, 1H, H-4, $J_{4,3}=10$, $J_{4,2}=2$), 7.13 (d, 1H, H-7, $J_{7,8}=9$), 7.6 (dd, 1H, H-8, $J_{8,7}=9$, $J_{8,10}=2$), 7.87 (d, 1H, H-10, $J_{10,8}=2$)	278 (M^+, 100), 277 (M − H, 48), 250 (M − CO, 30), 249 (M − HCO, 45), 222 (M − C_3H_4O, 46), 198 (M − C_5H_4O, 47), 170 (M − $C_6H_4O_2$, 60)
6e	45	131 (benzene)	$C_{12}H_7ClO_3$ (234.61)	1705	208 (4.28), 210 (4.27), 270 (3.66), 310 (3.63)	5.28 (dd, 2H, H-2, $J_{2,3}=3$, $J_{2,4}=2$), 5.70 (dt, 1H, H-3, $J_{3,4}=10$, $J_{3,2}=3$), 6.56 (dt, 1H, H-4, $J_{4,3}=10$, $J_{4,2}=2$), 7.10–7.21 (m, 2H, H-7, H-9), 7.50 (d, 1H, H-10, $J_{10,9}=10$)	234 (M^+, 100), 233 (M − H, 46), 206 (M − CO, 32), 205 (M − HCO, 48), 178 (M − C_3H_4O, 48), 154 (M − C_5H_4O, 45), 126 (M − $C_6H_4O_2$, 64)

[a] Yield of pure isolated product.
[b] Satisfactory microanalyses obtained: C ± 0.26, H ± 0.12.
[c] Solvent: **2a–e**, **5a–e**, **6a–e** = $CDCl_3$; **3a–e** = acetone-d_6.



Scheme **B**

Thus the route presented offers a novel method for the synthesis of pyrano[3,2-*c*][1]benzopyrans, which are not easily obtained by other methods.[16-20]

Melting points (uncorrected) were determined on a sulfuric acid bath. IR (KBr) spectra were recorded on a Perkin-Elmer Infracord spectrophotometer model 337. UV spectra were recorded on a Shimadzu UV-VIS 200 spectrophotometer using MeOH as solvent, λ max in nm (log ϵ). ^{1}H-NMR spectra were recorded on Varian A60D with TMS as internal standard. Mass spectra were recorded on a Perkin-Elmer Hitachi RMU-6L mass spectrometer.

4-Allyloxy-2*H*-[1]benzopyran-2-ones 2; General Procedure:
A mixture of 4-hydroxy-2*H*-[1]benzopyran-2-one **1** (0.01 mol), allyl bromide (0.86 mL, 0.01 mol) and K_2CO_3 (5.5 g, 0.04 mol) in acetone (100 mL) is refluxed for 6 h on a steam bath. K_2CO_3 is filtered off and the filtrate concentrated. The product, 4-allyloxy-2*H*-[1]benzopyran-2-one **2** crystallized from benzene as colorless rods (Table).

3-Allyl-4-hydroxy-2*H*-[1]benzopyran-2-ones; General Procedure:
4-Allyloxy-2*H*-[1]benzopyran-2-one **2** is heated its melting point (Table) for 2 h under reduced pressure (0.1 Torr). The compound melts at first, but gradually solifies after 1 h. The product is extracted with 5% cold aq NaOH. Acidification with cold aq HCl gives 3-allyl-4-hydroxy-2*H*-[1]benzopyran-2-one **3** which crystallized from Et_2O as colorless needles (Table).

Sodium 3-Allyl-2-oxo-2*H*-[1]benzopyran-4-olates 4; General Procedure:
An aq suspension of 3-allyl-4-hydroxy-2*H*-[1]benzopyran-2-one **3** (0.001 mol) in H_2O (10 mL) is treated with aq NaOH (0.04 g in 10 mL H_2O, 0.001 m, 10 mL). The solution is concentrated under reduced pressure and throughly dried in a desiccator ($CaCl_2$) to yield a yellow solid.

2-Methyl-4*H*-furo[3,2-*c*][1]benzopyran-4-ones 5 and 2*H*,5*H*-Pyrano[3,2-*c*][1]benzopyran-5-ones 6; General Procedure:
A suspension of the sodium salt of **4** (0.005 mol) in benzene (200 mL) containing $PdCl_2(PhCN)_2$ (1.91 g, 0.005 mol) is stirred at r.t. for 30 min. The suspension becomes clear and develops an intense red colour during stirring. The clear solution is refluxed for 2 h when metallic Pd begins to precipitate and the solution turns colorless. Pd is filtered off and the filtrate concentrated. The reaction products benzonitrile, 2-methyl-4*H*-furo[3,2-*c*][1]benzopyran-4-one **5** and 2*H*,5*H*-pyrano[3,2-*c*][1]benzopyran-5-one **6** are separated by column chromatography on silica gel (25 g). On first elution with benzene (300 mL) gives benzonitrile and subsequent elution with benzene/EtOAc (9:1) gives 2-methyl-4*H*-furo-[3,2-*c*][1]benzopyran-4-one **5** followed by 2*H*,5*H*-pyrano[3,2-*c*]-[1]benzopyran-5-one (Table).

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