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A practical procedure for the selective *N*-alkylation of 4-alkoxy-2-pyridones and its use in a sulfone-mediated synthesis of *N*-methyl-4-methoxy-2-pyridone

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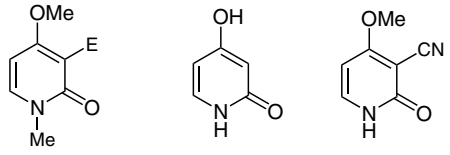
Abstract—It has been found that selective *N*-alkylation of 4-alkoxy-2-pyridones can be achieved under anhydrous, mild conditions with tetrabutylammonium iodide and potassium *tert*-butoxide being employed as the catalyst and the base, respectively. The procedure was applied to the preparation of 4-methoxy-1-methyl-2-pyridone, a valuable building block for heterocycle synthesis.

N-Alkylated 2-pyridones are important intermediates in the synthesis of polycyclic compounds of biological significance as illustrated by the recent synthetic approaches toward the camptothecin family of antitumor agents.[1] They are also structural subunits of naturally occurring products such as the heterocycle-annelated pyridone alkaloid cerpegin having analgesic, anti-ulcer, and anti-inflammatory activities[2] or the antibiotic 4-hydroxypyridones funiculosine, which possesses fungicide properties[3] and aurodox, an antimicrobial agent.[4] However, there is still a need for efficient methods allowing the selective *N*-alkylation of 2-pyridones, as known procedures generally suffer from low yields and/or competition between *N*- and *O*-alkylation.[5] In the course of a program aimed at evaluating the synthetic potential of *N*-alkylated-4-alkoxy-2-pyridones as precursors of new drug-like heterocycles,[6] we needed to prepare a series of such compounds and were particularly interested in 4-methoxy-*N*-methyl-2-pyridone **1**, a known building block in heterocycle synthesis,[3,6,7] and its phenylsulfonyl derivative **2**[8] as model substrates.



1 E = H
2 E = SO_2Ph
3 E = CN **(ricinin)**

4

5

To the best of our knowledge, no satisfactory procedure was available from the literature in terms of practicity and low cost for the large scale preparation of **1**. Alkylation of commercially available, but rather expensive, 4-hydroxy-2-pyridone (**4**) is rather difficult owing to its low solubility in common organic solvents. Direct bismethylation of **4** has been reported to proceed under phase transfer conditions (H_2O/benzene; 70 °C) using dimethylsulfate in the presence of benzyl triethylammonium bromide.[3] However, the process is sluggish, low yielding (50%), and isolation of the product rather tedious.

In 1917, Winterstein et al.[9] reported the isolation of *N*,*O*-dimethylpyridone **1** obtained upon heating of the alkaloid ricinin (**3**) in aq H_2SO_4. Ricinin was initially obtained from castor seeds (*Ricinus communis* L.)[10] but can now be prepared from malononitrile according

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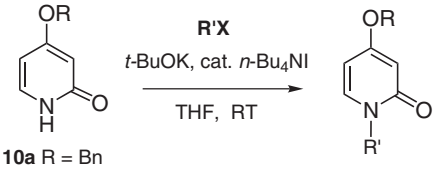
to a four-step procedure as reported by Junek and coworkers.[11] The most significant drawback of the method was again the low yield obtained for the *N*-methylation of the 3-cyanopyridone precursor **5** (Me_2SO_4, aq NaOH, rt, 53% yield). As it was predicted that sulfonylpyridone **2** would be available from phenylsulfonylacetonitrile (**6**) according to the same reaction sequence, and that pyridone **1** would then be accessible from desulfonylation of **2**, avoiding the *N*-methylation in the last step, we planned to exploit this approach as a common route to both compounds (Scheme 1).

Thus, preparation of the cyclic precursor **9** was accomplished in three high yielding steps by using slight modifications of Junek's procedure. Notably, in our modified method, cyclization of the enamine precursor was conducted in 80% aq acetic acid[12] instead of concd sulfuric acid. With pyridone **9** in hand, the next issue to address was the alkylation step. As expected, the aforementioned procedures involving aqueous systems proved unsatisfactory in terms of yields (<50%) and ease of product purification. We then decided to seek new conditions and were pleased to find that selective *N*-methylation of **9** could be accomplished under mild conditions by using the anhydrous system MeI/*t*-BuOK/cat. *n*-Bu_4NI[13] in THF at room temperature. The desired sulfonyl derivative **2** was obtained in nearly quantitative yield. It is of interest that the reaction sequence has been successfully scaled up to produce 45 g of **2** in a single batch.

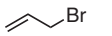
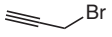
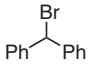
Removal of the phenylsulfonyl group in **2** was then investigated. A screening of desulfonylation conditions suggested from the literature were tried (Na/Hg, MeOH;[14] Mg/$HgCl_2$, EtOH;[15] H_2, Raney Ni, EtOH;[16] Bu_3SnH, AIBN, toluene;[17] *i*-PrMgBr, $Ni(acac)_2$, THF;[18] $NaBH_4$, DMF[19]) all of which failed to give the desired product in satisfactory yield, if any. Finally, the procedure developed by Julia for the desulfonylation of acyclic vinyl sulfones by sodium dithionite ($Na_2S_2O_4$) proved to be quite effective. In the original report,[20] the reductant was used in combination with $NaHCO_3$ and Adogen™ under phase transfer conditions (benzene–water, 80 °C). However, optimization studies established that desulfonylation of **2** proceeds faster and gives better yields (up to 93%) by substituting Adogen™ for *n*-Bu_4NI and by carrying out the reaction at 90 °C in aqueous toluene.

Having succeeded in developing a practical, alternative synthesis of pyridone **1**,[21] we then focused our attention on the general applicability of the *N*-alkylation of pyridones by mean of the *t*-BuOK/*n*-Bu_4NI system. Quite interestingly, methylation of cyanopyridone **5** under the same conditions used previously for **9** was shown to proceed smoothly to yield ricinin (**3**) in an improved 90% isolated yield compared to that reported by Junek (53%). The alkylation protocol applied also to other alkylating reagents as demonstrated by reaction of 4-alkoxypyridones **10a**,**b** (Table 1)[22] with methyl iodide,[23] benzyl bromide, allyl bromide, propargyl bromide, as well as the less activated *n*-butyl iodide. Even the secondary benzhydryl bromide[24] participated in the process but required higher temperature (50 °C) and prolonged reaction times.[25]

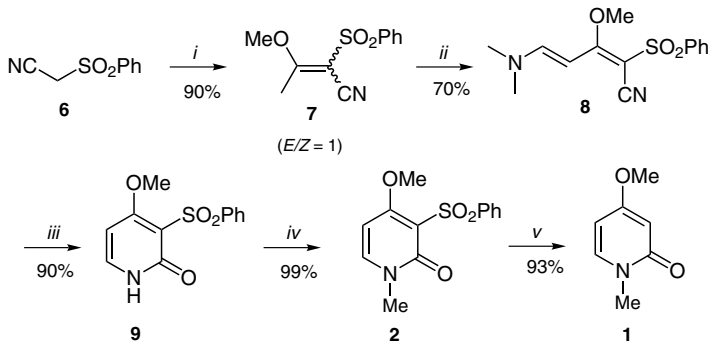
Table 1. Selective *N*-alkylation of 4-alkoxy-2-pyridones



10a R = Bn
10b R = Me

Entry	Pyridone	Alkyl halide	Yield (%)[a]
1	**10a**	MeI	91% (**11a**)
2	**10a**	BnBr	93% (**11b**)
3	**10a**		90% (**11c**)
4	**10a**		83% (**11d**)
5	**10a**		62%[b] (**11e**)
6	**10b**	MeI	91% (**1**)
7	**10b**	BnBr	95% (**11f**)
8	**10b**	*n*-BuI	94%[c] (**11g**)

[a] Yields refer to single runs. Reactions conducted overnight on a half-millimolar scale. Ratio **10**–R′X–*t*-BuOK–*n*-Bu_4NI = 1:1.5:1.1:0.05.
[b] 3 equiv of benzhydryl bromide were used (50 °C, 3 days).
[c] 3 equiv of butyl iodide were used.



Scheme 1. Reagents and conditions: (i) neat $MeC(OMe)_3$, Δ (–MeOH); (ii) neat $Me_2NCH(OMe)_2$, 140 °C; (iii) 80% aq AcOH reflux; (iv) MeI (1.5 equiv), *t*-BuOK, cat. *n*-Bu_4NI, THF, 0 °C to rt; (v) $Na_2S_2O_4$, $NaHCO_3$, *n*-Bu_4NI, toluene–H_2O, 90 °C.

In conclusion, we have described an efficient protocol for the selective *N*-alkylation of 4-alkoxy-2-pyridones, which also holds promise for the alkylation of 2-pyridones in general. We have also established an alternative, practical procedure for the synthesis of 4-methoxy-1-methyl-2-pyridone **1**, a versatile building block in heterocycle synthesis. The method is particularly well suited for the preparation of **1** in multi-gram scale.

Acknowledgments

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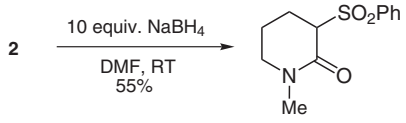
19. Wrobel, Z. *Tetrahedron* **1998**, *54*, 2607–2618. When applied to **2** the reaction afforded the corresponding saturated dealkoxylactam:



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21. Experimental part: Preparation of **1** from phenylsulfonylacetonitrile **6**:
3-Methoxy-2-(phenylsulfonyl)but-2-enenitrile (**7**): Phenylsulfonylacetonitrile (1.9 g, 10.5 mmol) and 1,1,1-trimethoxyethane (1.5 g, 12.5 mmol) were placed in a round-bottomed flask fitted with a distillation apparatus. The mixture was heated with stirring until a gentle distillation of methanol started (oil bath heated to approx. 100 °C). Once distillation ceased, the mixture was allowed to reach room temperature, at which point it solidified. The solid residue was taken up in methanol (20 mL) and the resulting suspension was cooled to −20 °C and let to stand for 2 h at that temperature. The precipitate was collected by suction filtration, washed with cold methanol (5 mL), and dried under vacuum to yield 2.2 g (90%) of **7** as a colorless solid (1:1 mixture of *E* and *Z* isomers). NMR data were in agreement with that reported in the literature.[26]
5-(Dimethylamino)-3-methoxy-2-(phenylsulfonyl)penta-2,4-dienenitrile (**8**): A neat mixture of **7** (1.02 g, 3.5 mmol) and *N*,*N*-dimethyl formamide dimethyl acetal (0.63 g, 5.3 mmol) was heated at 140 °C for 3 h. The mixture was then allowed to reach room temperature and concentrated in vacuo to give a reddish oil, which was subjected to column chromatography (silica gel; ethyl acetate–petroleum ether: 55:45) to afford **8** as a pale yellow solid (720 mg, 70%). Mp 147 °C. ^{1}H NMR (300 MHz, $CDCl_3$): 2.96 (s, 3H); 3.21 (s, 3H); 3.93 (s, 3H); 5.20 (br s, 1H); 7.49–7.51 (m, 4H); 7.96 (d, $J = 6.7$ Hz, 2H). ^{13}C NMR (75 MHz, $CDCl_3$): 37.61; 46.01; 61.17; 85.19; 86.91; 117.5; 126.90; 128.83; 132.58; 143.67; 154.32; 178.56.
4-Methoxy-3-(phenylsulfonyl)pyridin-2(1*H*)-one (**9**): A solution of **8** (5.04 g, 19.0 mmol) in 80% aq acetic acid (25 mL) was refluxed for 4 h. The reaction mixture, which solidified upon cooling to room temperature, was diluted with ethyl acetate (10 mL) and cooled in ice water. The

solid product was collected by suction filtration and washed with cold ethyl acetate (5 mL). Recrystallization from methanol gave **9** as a pale yellow solid (4.5 g, 90%). Mp 220 °C (dec). ^{1}H NMR (300 MHz, DMSO-d_6): 3.90 (s, 3H); 6.26 (d, $J = 7.5$ Hz, 1H); 7.50–7.60 (m, 3H); 7.69 (d, $J = 7.5$ Hz, 1H); 7.88 (dd, $J = 8.7$–1.7 Hz, 2H). ^{13}C NMR (75 MHz, DMSO-d_6): 58.30; 94.64; 112.01; 127.87; 129.27; 133.38; 143.24; 144.25; 159.52; 170.68.

4-Methoxy-1-methyl-3-(phenylsulfonyl)pyridin-2(1*H*)-one (**2**): To a stirred, cooled solution (0 °C) of pyridone **9** (1.78 g, 6.7 mmol) in THF (20 mL) were successively added *t*-BuOK (0.82 g, 7.3 mmol) and *n*-Bu_4NI (0.125 g, 0.34 mmol) under nitrogen and the resulting reaction mixture was held at 0 °C for 15 min. Methyl iodide (1.4 g, 9.9 mmol) was then added and the mixture was allowed to reach room temperature and stirred overnight. The reaction mixture was then quenched with water (20 mL) and THF was removed in vacuo to leave a white suspension in water, which was collected by suction filtration, washed with water, and dried over P_2O_5 to give **2** as an off-white solid (1.8 g, 99%). Mp 207–208 °C. ^{1}H NMR (200 MHz, $CDCl_3$): 3.44 (s, 3H, NMe); 4.01 (s, 3H, OMe); 6.07 (d, $J = 7.7$ Hz, 1H); 7.45–7.60 (m, 4H); 8.11 (d, $J = 6.8$ Hz 2H). ^{13}C NMR (75 MHz, $CDCl_3$): 37.95; 57.92; 94.21; 113.20; 128.50; 128.99; 133.30; 144.11; 144.98; 159.46; 169.82. Anal. Calcd for $C_{13}H_{13}NO_4S$: C, 55.90; H, 4.69; N, 5.01. Found C, 55.80; H, 4.79; N, 4.95.

4-Methoxy-1-methylpyridin-2(1*H*)-one (**1**): $Na_2S_2O_4$ decomposes rapidly in aqueous solution to chiefly thiosulfate and sulfite, and should therefore be added in portions as follows: To a mixture of pyridone **2** (2.0 g, 7.2 mmol), $NaHCO_3$ (6.0 g, 71.4 mmol), and *n*-Bu_4NI (0.53 g, 1.44 mmol) in toluene (80 mL) and water (80 mL) was added a first portion of $Na_2S_2O_4$ (2.5 g, 14.3 mmol) and the well-stirred mixture was warmed to 90 °C in the dark under an atmosphere of nitrogen. After 2 h an additional portion of $Na_2S_2O_4$ (2.5 g) was cautiously added and stirring was continued for an additional 2 h. This addition procedure was repeated until complete desulfonylation had occurred as judged from TLC analysis (silica gel, acetone). In general, 8–14 equiv of $Na_2S_2O_4$ were necessary to achieve complete conversion. The reaction mixture was then cooled to room temperature. The organic layer was collected and the aqueous phase was extracted with ethyl acetate and dichloromethane (5×20 mL each). The organic extracts were combined, dried over $MgSO_4$, and concentrated in vacuo. The residue was subjected to column chromatography (silica gel; acetone) to afford **1** as a white solid (0.93 g, 93%). Mp 112–114 °C (lit.[27] 113–114 °C). NMR data were in agreement with that reported in the literature.[3]

22. Compound **10a** is commercially available; **10b** was obtained by desulfonylation of **9** (51% yield) according to the same procedure as used for **2**.
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25. In a typical experiment, *t*-BuOK (0.55 mmol) and *n*-Bu_4NI (0.025 mmol) were added to a cooled solution (0 °C) of pyridone **10** (0.5 mmol) in THF (5 mL) and the resulting reaction mixture was held at 0 °C for 15 min. The alkyl halide (0.75 mmol) was added and the resulting mixture was left to stir overnight. The mixture was then concentrated in vacuo, and after usual workup with 1 N aq NaOH and dichloromethane, the crude product was purified by flash chromatography on silica gel eluting with an appropriate combination of ethyl acetate and dichloromethane. Selected data for *N*-alkylated pyridones **11**: ^{1}H NMR ($CDCl_3$, 300 MHz). **11b**: δ 7.42–7.27 (m, 10H); 7.13 (d, $J = 7.5$ Hz, 1H); 6.05 (d, $J = 2.6$ Hz, 1H); 5.95 (dd, $J = 2.6$ and 7.5 Hz, 1H); 5.09 (s, 2H); 4.99 (s, 2H). **11c**: δ 7.30–7.25 (m, 5H); 7.03 (d, $J = 7.35$ Hz, 1H); 5.90–5.75 (m, 3H); 5.16 (dd, $J = 1.3$ and 10.2 Hz, 1H); 5.10 (d, $J = 1.3$ and 17.1 Hz, 1H); 4.89 (s, 2H); 4.41 (dt, $J = 1.3$ and 5.65 Hz, 2H). **11d**: δ 7.48 (d, $J = 7.5$ Hz, 1H); 7.40–7.35 (m, 5H); 6.03 (dd, $J = 2.6$ and 7.5 Hz, 1H); 5.98 (d, $J = 2.6$ Hz, 1H); 4.98 (s, 2H); 4.68 (d, $J = 2.6$ Hz, 2H); 2.47 (t, $J = 2.6$ Hz, 2H). **11e**: δ 7.45 (s, 1H); 7.40–7.30 (m, 11H); 7.14 (m, 4H); 7.02 (d, $J = 7.9$ Hz, 1H); 6.08 (d, $J = 2.6$ Hz, 1H); 5.93 (dd, $J = 2.6$ and 7.9 Hz, 1H); 4.99 (s, 2H). **11f**: δ 7.35–7.25 (m, 5H); 7.11 (d, $J = 7.5$ Hz, 1H); 5.97 (d, $J = 2.8$ Hz, 1H); 5.89 (dd, $J = 2.8$ and 7.5 Hz, 1H); 5.09 (s, 2H); 3.76 (s, 3H). **11g**: δ 7.05 (d, $J = 8.3$ Hz, 1H); 5.81 (m, 2H); 3.79 (t, $J = 7.4$ Hz, 3H); 3.68 (s, 3H); 1.62 (m, 2H); 1.28 (m, 2H); 0.87 (t, $J = 7.4$ Hz, 3H).
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