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INDIRECT OXIDATIVE DEAMINATION OF BENZYLAMINES TO BENZALDEHYDES

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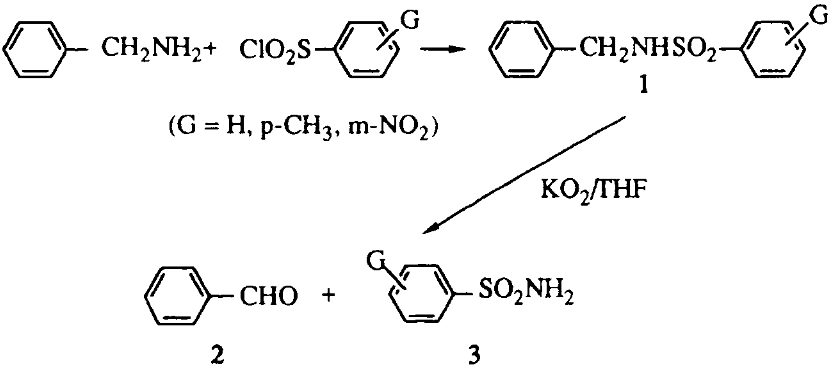
Abstract: Treatment of N-benzylarenesulfonamide (**1**) with potassium superoxide yields benzaldehyde (**2**) and arenesulfonamide (**3**).

Deamination of benzylamine to benzaldehyde is achieved, in most cases, indirectly using various oxidants to make corresponding imines which could then be hydrolyzed in acidic conditions.[1-3] We report a simple and convenient indirect method for the conversion of benzylamine to benzaldehyde (Scheme 1). As shown in Scheme 1, N-benzylarenesulfonamide (**1**)[4] prepared from benzylamine and arenesulfonyl chloride gives benzaldehyde (**2**), an oxidatively cleaved product, and arenesulfonamide (**3**) in the presence of potassium superoxide[5-7] at room temperature.

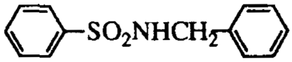
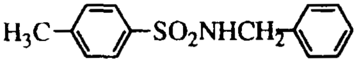
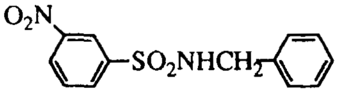
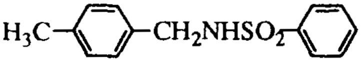
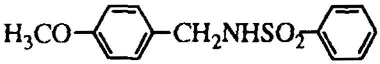
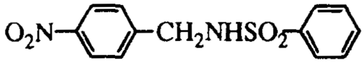
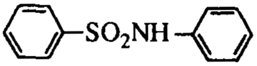
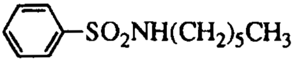
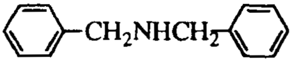
Scheme 1



A general procedure of the present methodology is as follows: In a round-bottomed flask (50 mL) containing dry tetrahydrofuran (25 mL), equipped with a condenser connected to gas bubbler and a magnetic stirrer, potassium superoxide (510 mg, 57 mmol) was introduced. While passing oxygen-free nitrogen from the bottom of the solution, N-benzylbenzenesulfonamide (470 mg, 19 mmol) was added. Soon vigorous formation of oxygen[8] ensued for about 15 seconds and then subsided. During the whole reaction period, no liberation of heat was observed. As the reaction proceeded, concentration of both benzaldehyde and benzenesulfonamide[9] increased at the expense of N-benzylbenzenesulfonamide, determined by G. C. and ^{1}H NMR spectra.

Two points can be noted from the results of this reaction (in Table1). One is that the present method (Scheme 1) is limited and unique only to N-benzylarenesulfonamides (entry 1-6, Table 1) in reaction with

Table 1. Synthesis of Benzaldehyde from the Reaction of N-Benzylbenzenesulfonamide and Related Compounds with KO_2

Entry	Substrate	Product	Reaction Time(hr)	Yield* (%)
1		PhCHO	24	88
2		PhCHO	24	90
3		PhCHO	24	19
4		p-CH_3-C_6H_4CHO	24	83
5		p-OCH_3-C_6H_4CHO	24	84
6		p-NO_2-C_6H_4CHO	24	85
7			20	0
8			20	0
9			20	0
10			8	0

*: Determined by either G. C. or ^1H NMR spectrum.

potassium superoxide to give benzaldehyde and arenesulfonamide. Other compounds such as N-phenylbenzenesulfonamide (entry 7), N-hexylbenzenesulfonamide (entry 8), N-benzylbenzamide (entry 9), and dibenzylamine (entry 10) are inert in these reaction conditions. The other is the facile and unusual N-C bond cleavage of compound **1** by potassium superoxide. It is described in the literature that structure of compound **1** is quite stable to acid or base. In forcing conditions, however, S-N bond is usually cleaved rather than N-C bond.[10,11] In conclusion the unusual cleavage of N-benzylarenesulfonamide (**1**) by potassium superoxide (Scheme 1) presents an alternative methodology for the indirect oxidative deamination of benzylamine.

Mechanistic view points of this method are currently under investigation.

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8. G. C. indicates that the gas is actually oxygen (more than 99%) with negligible amount of hydrogen (less than 1%).

9. Present in the solution either as potassium salt or benzensulfonamide itself.

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