## Reaction of Benzofurazan N-Oxide with 1-Aza-1,3-Butadienes. Synthesis of a Novel Class of Quinoxaline N,N'-Dioxide Imines

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1-Aza-1,3-butadienes react with benzofurazan N-oxide (BFO) to give a new class of N,N'-dioxide imines.

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Benzofuran oxide 1 reacts with a variety of nucleophiles, enamines and enolates, and this chemistry has recently been reviewed [2]. Our earlier investigations on 1-aza-1,3-butadienes 2 had revealed that the carbon-carbon double bond does not participate in reactions and several dipoles [3], as well as nucleophiles [4], react with the carbon-nitrogen double bond only. In contrast, here we report reaction of BFO with azadienes 2 in which the carbon-carbon double bond has reacted, giving a novel class of quinoxaline N, N'-dioxide imines 3.

Equimolar quantities of 1, 1.36 g (0.01 mole) and 2 2.07 g (0.01 mole), on refluxing in dry benzene for one hour and then kept stirring for another 72 hours, on removal of benzene gives a dark brown residue. This mass was washed with dry ether ( $100 \times 5$  ml). The ether was removed under vacuum and the residue was crystallised from benzene-light petroleum ether to obtain yellow crystals, mp 189-190° in 60% yield [5]. The structural assignment of adduct 3a is supported by spectroscopic, as well as elemental data; ir (potassium bromide): 1625 and 1600 (C=N bonds) and 1350 (N  $\rightarrow$  O); nmr (deuteriochloroform, tetramethylsilane):  $\delta$  360 MHz 6.87 (2H, m), 7.27 (3H, m), 7.52 (5H, m), 7.93 (2H, m), 8.72 (2H, m), 8.86 (1H, s); ms: m/e 341 (3.2%), m/e 325 (12.1%), m/e 308 (100%).

Anal. Calcd. for  $C_{21}H_{15}N_3O$ : C, 73.90; H, 4.40; N, 12.32. Found: C, 73.88; H, 4.44; N, 12.30.

The two consecutive losses of 16 and 17 units are typical of di-N,N'-oxides. The absence of CH=CH trans and the presence of an azomethine proton at 8.86 (1H, s) proves beyond doubt that only the C=C site of 2 is involved in the reaction.

We have also observed similar products 3b and 3c when

we used N-(p-toluidine) (mp 189-191°, 65% yield) and N-(p-anisidine) (mp 193-194°, 68% yield) analogues of 2.

Compound **3b** had the following analytical data: *Anal.* Calcd. for  $C_{22}H_{17}N_3O_2$ : C, 74.37; H, 4.79; N, 11.83. Found: C, 74.29; H, 4.82; N, 11.88.

Compound **3c** had the following analytical data: *Anal.* Calcd. for  $C_{22}H_{17}N_3O_3$ : C, 71.16; H, 4.58; N, 11.32. Found: C, 71.04; H, 4.60; N, 11.42.

The N-methyl analogue of 2 had 3d, mp 172-173°, 70% yield. In the nmr (360 MHz, deuteriochloroform), the methyl group showed at  $\delta$  3.53 (3H, d) and the azomethine proton at  $\delta$  8.53 (1H, q). The mutual coupling was confirmed by spin-spin decoupling. The magnitude of this coupling (J = 1.6 Hz) clearly indicates a trans arrangement at the azomethine site [6].

Compound **3d** had the following analytical data: *Anal.* Calcd. for C<sub>16</sub>H<sub>13</sub>N<sub>3</sub>O<sub>2</sub>: C, 68.81; H, 4.66; N, 15.05. Found: C, 68.79; H, 4.60; N, 15.13.

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