RECYCLIZATION OF 2-(2-PHENYLETHYL)PYRIDINIUM ALKIODIDES TO o-ALKYLAMINODIPHENYLMETHANES

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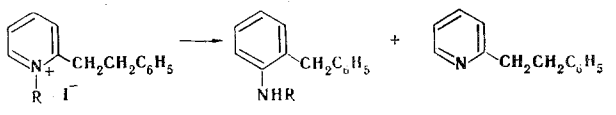
It is known that 1,2-dialkylpyridinium salts undergo recyclization to N-alkylanilines under the influence of nucleophilic agents [1].

We have established that 2-(2-phenylethyl)pyridinium alkiodides (Ia-c) undergo similar rearrangement to 2-benzyl-N-alkylanilines (IIa-c), which are diphenylmethane derivatives, under the influence of 50% aqueous solutions of alkylammonium sulfites (when the mixtures are heated at 190-200°C for 14-16 h).



I a-c II a-c III

I-II a R=CH_3; b R=C_2H_5; c R=н-C_4H_9

The only example of the preparation of o-methylaminodiphenylmethane known in the literature is a one-step synthesis [2]: Other o-alkylamino derivatives of diphenylmethane are unknown. The proposed method makes it possible to obtain these previously inaccessible compounds in one step.

Thus, o-methylaminodiphenylmethane (IIa), with mp 59-60°C (from benzene), was obtained in 82% yield from salt Ia and methylammonium sulfite. PMR spectrum (CCl_4): 2.75 (3H, s, CH_3N), 3.82 (2H, s, CH_2), 3.34 (1H, s, NH), and 6.51-7.31 ppm (9H, m, aromatic protons). o-Ethylaminodiphenylmethane (IIb), with mp 64-65°C (from benzene), was similarly obtained in 79% yield from salt Ib and ethylammonium sulfite. PMR spectrum (CCl_4): 1.07 (3H, t, $CH_2\mathbf{CH_3}$), 3.03 (2H, q, $\mathbf{CH_2}CH_3$), 3.83 (2H, s, CH_2), and 6.13-7.21 ppm (9H, m, aromatic protons). o-(n-Butylamino)-diphenylmethane (IIc), with mp 73-74°C (from benzene), was similarly obtained in 78% yield from salt Ic and n-butylammonium sulfite. PMR spectrum (CCl_4): 0.75-1.52 (7H, m, $CH_2CH_2CH_3$), 2.95 (2H, t, N—CH_2), 3.25 (1H, s, NH), 3.82 (2H, s, CH_2), and 6.08-7.21 ppm (9H, m, aromatic protons).

In the reaction of salts Ia-c with aqueous solutions of alkylammonium sulfites we isolated, in addition to II, the product of their dealkylation 2-(2-phenylethyl)pyridine (III) (in up to 10% yield), which is formed due to attack by the nucleophile at the nonring carbon atom bonded to the heteroatom.

The results of elementary analysis of IIb,c are in agreement with the calculated values, and IIa was identical to a genuine sample. A band of stretching vibrations of an NH group at 3365 cm^{-1} is observed in the IR spectra of amines IIa-c.

LITERATURE CITED

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