TABLE II

Amine	Derivatives: Dipicrate M. p., °C.	Dipicrate N, % Calcd.	Dipicrate N, % Found	Phenylurea M. p., °C.	Phenylurea N, % Calcd.	Phenylurea N, % Found
N,N'-Dibutylethylenediamine	148–149.5[a,6]	17.78	17.43	174–174.5	13.65	13.31
N,N'-Dioctylethylenediamine	158–159.5[b,7]	15.09	14.73	129.5–130	10.72	10.30
N,N'-Didodecylethylenediamine	152–154.5[c,7]	13.11	13.11	235 (subl.)	8.83	8.64
N,N'-Dicyclohexylethylenediamine	210 (dec.)	16.42	16.04	206	12.11	12.45
N,N'-Dibenzylethylenediamine	208–210[d] (dec.)	14.93	14.71	182[8]	11.71	11.73
N,N'-Dibutylpiperazine	155–156	17.07	16.89	.....	...	...

[a] King and McMillan reported 188°. [b] Linsker and Evans reported 108°. [c] Linsker and Evans reported 112°. [d] Monopicrate.

isolation. This was not possible in the case of dibenzylethylenediamine, which did not form a solid monohydrate under the conditions employed. The by-products listed in Table I were obtained by fractional distillation of the dialkyl ethylenediamines.

Acknowledgment.—The authors are indebted to F. C. Bersworth, of the Bersworth Laboratories, Framingham, Massachusetts, for financial support for this research.

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(7) Linsker and Evans, *ibid.*, **68**, 1432 (1946).
(8) Van Alphen, *Rec. trav. chim.*, **54**, 93 (1935).

DEPARTMENT OF CHEMISTRY
CLARK UNIVERSITY
WORCESTER, MASSACHUSETTS RECEIVED JUNE 17, 1949

Acetyldesoxycellulose Quaternary Salts

BY F. N. HAYES AND CHAO-HAN LIN

A cellulose acetate tosylate was prepared by the method of Malm, Tanghe and Laird.[1] Separate portions of it were heated with pyridine, 3-picoline and isoquinoline to give quaternary salts, resulting from displacement of tosylate ions. Similar reactions of *p*-toluenesulfonate esters with tertiary amines are well known.[2,3] Table I gives analytical data and the calculated values for free hydroxyls, unreacted tosylate ester groups, and quaternary salt groups per glucose unit for each of the three products.

The average over-all percentage conversion of the original hydroxyl groups to quaternary salt groups may be used as an estimate of the per cent. primary free hydroxyl in the original cellulose acetate. Our value of 28% compares favorably with a reported value of 25% on a similar sample,[1] determined by the method of tosylation and iodination.

TABLE I

Sample	Percentage N[a]	Percentage S[a]	Amount per g. u. Free[b] OH	Amount per g. u. Tos.[b] ester	Amount per g. u. Quat.[b] salts	Percentage Conv. of OH to Tos. replaced	Percentage Conv. of OH to quat. salt
I	..	6.85	0.96	0.69	..	..	..
II	1.93	5.81	1.02	.14	0.49	71	30
III	1.77	5.96	0.97	.26	.42	61	26
IV	1.72	5.74	0.96	.22	.47	68	29

[a] Analyses by Micro-Tech Laboratories, Skokie, Ill. [b] Calculated from the analyses, assuming that the acetyl content remains unchanged.

A sample of cellulose acetate, EK-102893,[4] was tosylated[1] and the product (I) was reprecipitated from acetone by alcohol. Pyridine, 3-picoline and isoquinoline were dried and redistilled.

A solution of 6.0 g. of cellulose acetate tosylate in 60 ml. of pyridine was heated on a steam-bath for twenty-four hours, at the end of which time, it was diluted with 40 ml. of acetone and treated with just enough water to obtain a homogeneous solution. This was slowly poured into excess acetone with good stirring. The precipitated product was filtered, washed with acetone and twice reprecipitated from hot alcohol by ether. The yield of the purified product (II) was 5.5 g.

In a similar manner, 3.0 g. of the tosyl ester gave 2.8 g. of a 3-picolinium salt (III) and 8.0 g. yielded 7.8 g. of an isoquinolinium salt (IV).

(1) Malm, Tanghe and Laird, THIS JOURNAL, **70**, 2740 (1948).
(2) Cary, Vitcha and Shriner, *J. Org. Chem.*, **1**, 280 (1936).
(3) King, Dodson and Subluskey, THIS JOURNAL, **70**, 1176 (1948).
(4) Kindly supplied for this project by Eastman Kodak Company, with analysis: 1.35 acetyls per glucose unit.

DEPARTMENT OF CHEMISTRY
ILLINOIS INSTITUTE OF TECHNOLOGY
CHICAGO, ILLINOIS RECEIVED JUNE 30, 1949

Separation of Hafnium and Zirconium by a Fractional Distillational Procedure

BY D. M. GRUEN AND J. J. KATZ

In view of current interest in hafnium–zirconium separations,[1] we have investigated a separation method first reported by van Arkel and De Boer.[2] This method involves fractional distillation at atmospheric pressure of the volatile complex compounds formed by reaction of zirconium and hafnium tetrachlorides with either phosphorus pentachloride or phosphorus oxychloride. Although van Arkel and De Boer showed that distillation resulted in considerable separation of hafnium and zirconium they gave no quantitative data on the relative volatilities of the zirconium and hafnium compounds.

The present work is concerned chiefly with the phosphorus oxychloride complexes, since these have lower boiling points and greater thermal stability than the corresponding phosphorus

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