As a result of the work here reported, the following conclusions have been reached:

(1) The action of the mercaptides of lead and sodium on the quinones used consists of substitution and of reduction, and no addition-products have as yet been isolated.

(2) The substance described as an addition-product in the paper referred to, under the name tetrathioethylquinonedibenzoyldithiobenzoylacetal, does not possess the formula there assigned to it, but is really the dibenzoate of tetrathioethylhydroquinone, a reduction-product of tetrathioethylquinone.

(3) It was shown that the solvent used determines or largely modifies the course of several reactions between the substances under examination.

(4) By the selection of a suitable solvent and in other ways, the method of making tetrathioethylquinone, was improved so that 95 per cent. of the theoretical yield can now be obtained instead of 10 per cent. or less by the older methods.

(5) The preparation, solubilities and analysis of the dibenzoate of tetrathioethylhydroquinone (m. p. 131°), of the lead double salt of acetic acid and tetrathioethylhydroquinone and of the dithioethyldiethoxyhydroquinone dibenzoate (m. p. 184-184.5°) are here described.

In conclusion, I wish to express my thanks to Professor H. S. Grindley, of the Department of General Chemistry, for permission to undertake this work, in a field of research previously entered by himself.

UNIVERSITY OF ILLINOIS,
URBANA, ILLINOIS.

[CONTRIBUTIONS FROM THE HAVEMEYER LABORATORIES OF COLUMBIA UNIVERSITY, No. 107.]

THE CONDENSATION OF SUCCINYLOSUCCINIC ACID DIETHYL ESTER WITH GUANIDINE. A DERIVATIVE OF 1,3,5,7-NAPHTTETRAZINE, A NEW HETEROCYCLE.[1]

BY MARSTON TAYLOR BOGERT AND ARTHUR WAYLAND DOX.

Received July 21, 1905.

IN extending the work upon the quinazoline compounds, which has been going on in this laboratory for several years,[2]

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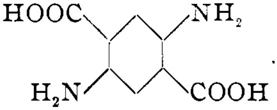
[1] Read at the meeting of the New York Section of the American Chemical Society, April 7, 1905.

[2] This Journal, **22**, 129, 522 (1900); **23**, 611 (1901); **24**, 1031 (1902); **25**, 372, 935 (1903); **27**, 649 (1905).

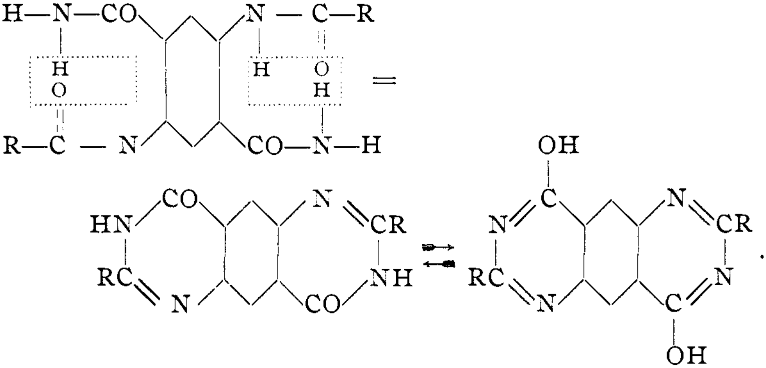
it was decided to conduct some experiments with, *p*-diaminoterephthalic acid, in the hope of obtaining naphttetrazines therefrom by the usual quinazoline condensations.

If *p*-diaminoterephthalic acid has the structure generally assigned to it, it contains the anthranilic acid grouping twice, as a glance at its constitutional formula will show:

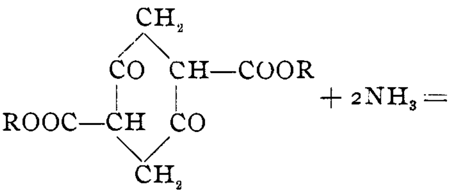


It might, therefore, be expected to give the usual anthranilic acid reactions and condensations.

Most of the quinazoline syntheses from anthranilic acids depend upon the intermediate formation of acylated anthranilamides, which then pass into the quinazolines by loss of water and ring closure. *p*-Diaminoterephthalic acid should give a naphttetrazine by such a condensation.[1]



p-Diaminoterephthalic acid was prepared from succinylosuccinic ester by converting the ester into the diimine, oxidizing the latter in concentrated sulphuric acid solution with bromine, and saponifying the resultant diaminoterephthalic ester. The reactions are as follows:



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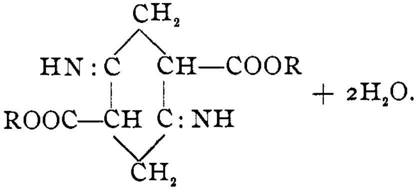
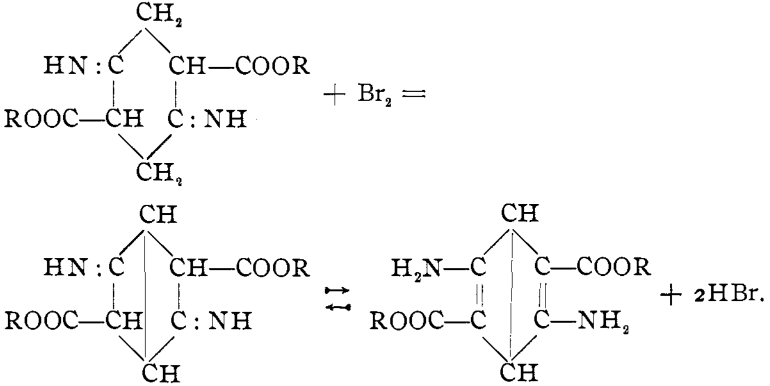
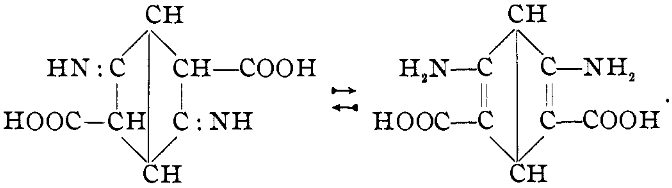
[1] Compare above references; also *J. prakt. Chem.* [2], **36**, 143 (1887); *Ibid.*, **40**, 1 (1889); *Ibid.*, **51**, 564 (1895); *Ber.*, **26**, 1349 (1893); *Ibid.*, **35**, 3480 (1902); etc.





Saponification of the ester then gives the free acid:



(I.) (II.)

This is the method of Baeyer[1] and Böninger.[2] Geuther[3] was of the opinion that the succinylosuccinic ester diimine possessed the tautomeric structure and was a true diaminodihydroterephthalic ester. Hantzsch and Hermann[4] assigned to the orange-red diaminoterephthalic ester the quinoid structure (I), because of its color, and to the salts with mineral acids, all of which are colorless, the diamino structure, the presence of the mineral acid inducing the change of the imino to the more basic amino group. Böninger,[5] for similar reasons, assigned the quinoid structure to the yellowish green, free diaminoterephthalic acid, and the

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[1] *Ber.*, **19**, 430 (1886).

[2] *Ibid.*, **21**, 1765 (1888).

[3] *Ann. Chem.* (Liebig), **244**, 213.

[4] *Ber.*, **21**, 1755 (1888).

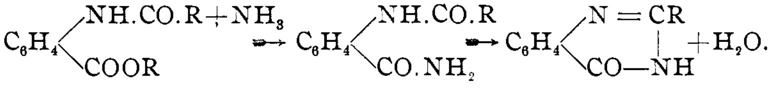
[5] *Loc. cit.*

diamino structure to its colorless salts with strong mineral acids. In giving the structure of the diamino tautomer (II), I have retained the para bond. This formula illustrates most simply the migration of hydrogen from carbon to nitrogen, and involves no readjustment of the binding. Further, it is not impossible that this may represent the true structure of the compound, and be the cause of some of its peculiar reactions.

In the preparation of the diimine from succinylosuccinic ester and ammonium acetate, Baeyer[1] recommends using 10 parts of acetate to 1 of ester, and working on small amounts (1 gram at a time). We have found that half as much acetate gives fully as good results, in fact we have occasionally used 2 parts of acetate to 1 of ester without noting any great difference in the yield or purity of the product; and further, 100 grams of ester can be converted into diimine just as readily as 1 gram. By repeated crystallization of the diimine from various solvents, we could not raise its melting-point above 177-178° (corr.). Baeyer gives it as 181°. If the fusion with ammonium acetate is properly conducted, the first product is practically pure and can be converted directly into the diaminoterephthalic ester.

The oxidation of succinylosuccinic ester diimine to *p*-diaminoterephthalic ester by bromine, in concentrated sulphuric acid solution, proceeds most satisfactorily at 40-50°, and is complete in about half an hour. The diamino ester is precipitated as sulphate, from which the ester is liberated by the action of sodium acetate.

Weddige[2] has prepared quinazolines by heating acylanthranilic esters with alcoholic ammonia at 150-160°:



$$C_6H_4\left\langle\begin{matrix}NH.CO.R+NH_3\\COOR\end{matrix}\right. \rightarrow C_6H_4\left\langle\begin{matrix}NH.CO.R\\CO.NH_2\end{matrix}\right. \rightarrow C_6H_4\left\langle\begin{matrix}N=CR\\ \quad|\\CO—NH\end{matrix}\right. +H_2O.$$

Attempts to prepare acyl derivatives of diaminoterephthalic ester were unsuccessful, so that the above condensation could not be realized.

The diamino ester reacts with phenyl isocyanate and with phenyl isothiocyanate, but the character of the products is still undetermined. The reaction should proceed as follows:[3]

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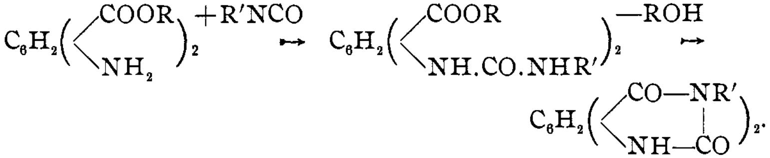
[1] *Loc. cit.*

[2] *J. prakt. Chem.* [2], **36**, 155 (1887).

[3] Compare Söderbaum and Widman: *Ber.*, **22**, 1669 (1889); Stewart: *J. prakt. Chem.* [2], **44**, 415 (1891); Wielandt: *Ibid.* [2], **49**, 319 (1894); etc.



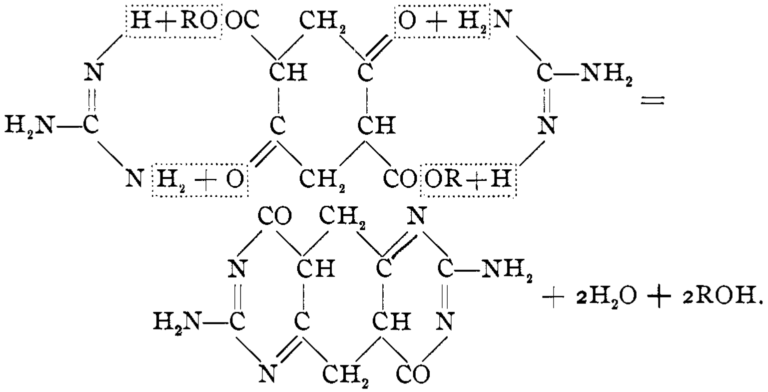
$$C_6H_2\left(\begin{matrix}COOR\\NH_2\end{matrix}\right)_2 \xrightarrow{+R'NCO} C_6H_2\left(\begin{matrix}COOR\\NH.CO.NHR'\end{matrix}\right)_2 \xrightarrow{-ROH} C_6H_2\left(\begin{matrix}CO-NR'\\ |\\ NH-CO\end{matrix}\right)_2.$$

p-Diaminoterephthalic acid may be prepared in any desired amount, with ease and rapidity, by the process outlined. It is exceedingly inert and insoluble. All attempts to obtain ortho condensations from it were futile. Acid anhydrides, formamide,[1] urea,[2] thiourea,[3] and phenylisothiocyanate had no action upon it.

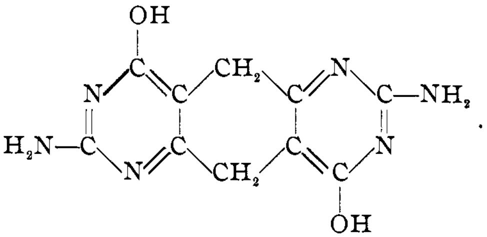
The study of *p*-diaminoterephthalic acid and its derivatives is being continued and the results will be communicated later.

The experiments with *p*-diaminoterephthalic acid having resulted so unsatisfactorily, we sought by another path to reach the same goal, and, by condensing guanidine with succinylosuccinic ester, achieved our purpose.

The condensation may be expressed thus:



The sodium salt may be a derivative of the tautomeric enol form:



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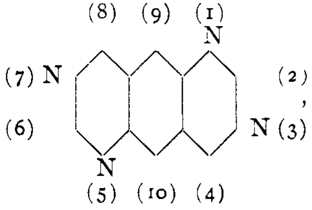
[1] Niementowski: *J. prakt. Chem.* [2], **51**, 564 (1895).

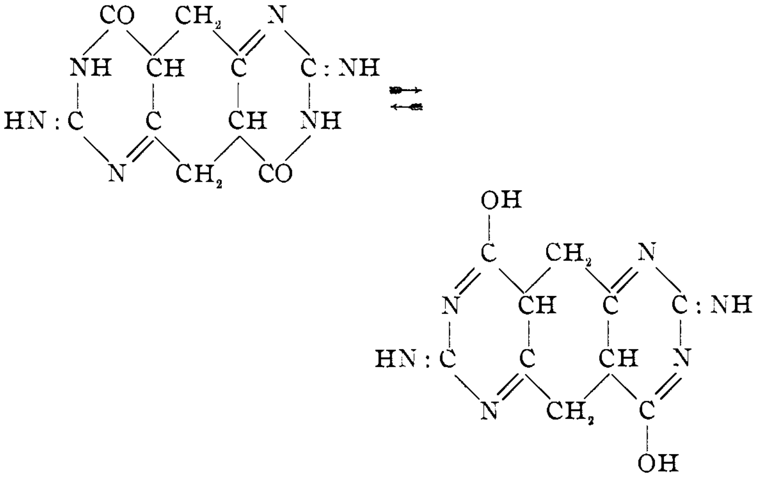
[2] Niementowski: *Ibid.* [2], **40**, 1 (1889).

[3] Stewart: *Loc. cit.*

The latter would be a mesodihydro derivative of the 1,3,5,7-naphttetrazine heterocycle,



and might be designated 2,6-diamino-4,8-dihydroxy-9,10-dihydro-1,3,5,7-naphttetrazine. It is possible, on the other hand, that the guanidine reacts with both its primary amino groups, leaving the imino group unaffected. The configuration would then be:



Other possible tautomers would arise by a different wandering of the hydrogens in the two —C(:NH)—NH—CO—CH: groupings. Which of these formulas correctly represents the structure of the condensation-product cannot yet be stated positively. Further experiments in this direction are under way.

The compound is an interesting one, as no similar heterocycles have ever been prepared so far as we can ascertain. Perhaps the nearest approach to it is the benzodipyrazolon obtained by condensing succinylosuccinic ester with hydrazine hydrate.[1]

We also endeavored to prepare the naphttetrazine from a pseudothiourea and succinylosuccinic ester diimine, thus,

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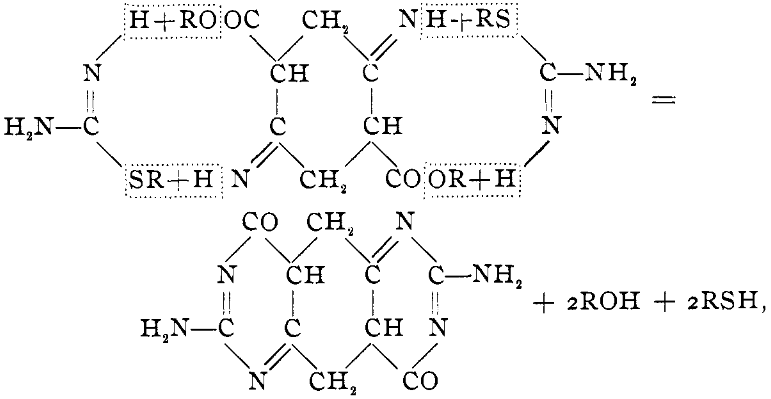
[1] *J. prakt. Chem.* [2], **51**, 64 (1895).

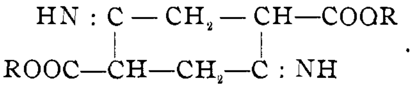


but the attempt was unsuccessful, although mercaptan was evolved in the reaction.

The naphttetrazine is characterized by great insolubility, infusibility, and inertness. It forms a yellow crystalline sodium salt, which possesses a beautiful green fluorescence.

EXPERIMENTAL PART.

Succinylosuccinic Acid Diethyl Ester Diimine,



$$\begin{array}{ccccc} HN:C & - & CH_2 & - & CH-COOR \\ | & & & & | \\ ROOC-CH & - & CH_2 & - & C:NH \end{array}.$$

—This was prepared, by a modification of the Baeyer method,[1] from succinylosuccinic ester and ammonium acetate. The reaction is an interesting one to watch.

One part of the ester is mixed with 5 parts of dry ammonium acetate, in a strong round-bottomed flask, and the mixture melted down carefully over the naked flame. The melted ester appears as clear, yellow, oily drops floating about in the colorless, water-clear ammonium acetate. After heating the melt for a few moments it begins to "crackle" (presumably from separation of water). This crackling soon ceases, and in a minute or two the liquid suddenly boils up vigorously (the flame should be removed for a few seconds, or the boiling may become too violent). Simultaneously, the yellow oily drops of ester solidify to pale yellow lumps of the diimine, which swim about in the liquid. The boiling should not be continued more than a minute or two longer to complete the reaction.

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[1] *Loc. cit.*

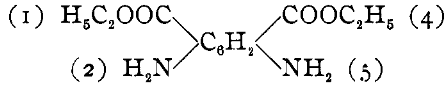
Baeyer heated the mixture of ester and acetate until a clear yellow solution resulted. This he says, took about a minute, when he used 1 gram of the ester to 10 of acetate. Our experience showed that it was inadvisable to continue heating until complete solution resulted, and that the best time to stop was a minute or two after the melted ester had changed to the solid diimine. It is quite true that if the mixture be boiled down rapidly over the free flame, all of the solid will gradually dissolve (using 25 grams of ester, it took fifteen minutes vigorous boiling before a clear solution resulted), but the color of the melt gradually changes from a light yellow to a dark olive-green or even to an olive-brown, and the product is darker-colored and less pure.

Upon the completion of the reaction, the melt is allowed to cool, with occasional shaking to prevent the formation of large crystals. A solid cake of ammonium acetate results, containing amorphous pale yellow lumps and light yellow crystals of the diimine, the crystals representing the diimine which was held in solution by the molten ammonium acetate. The cake is crushed up and washed thoroughly with cold water. The diimine remains undissolved as a pale yellow solid, practically pure, and in nearly quantitative yield. By crystallization from alcohol, it may be obtained in beautiful light yellow needles, but the crude product is sufficiently pure for direct conversion into *p*-diaminoterephthalic ester.

In spite of repeated crystallizations from various solvents, the melting-point of the diimine could not be raised above 177-178° (corr.). Baeyer gives it as 181°. It is apparently insoluble in cold water, very difficultly soluble on boiling; very difficultly soluble in dry ether, carbon disulphide, or naphtha; very difficultly soluble in cold carbon tetrachloride, difficultly soluble on boiling; difficultly soluble in cold glacial acetic acid, easily soluble on boiling; moderately soluble in cold absolute alcohol, but dissolves readily on boiling; difficultly soluble in cold ethyl nitrate, easily soluble on boiling; easily soluble in chloroform, acetone, benzene, nitrobenzene, aniline, or ethyl acetate. It crystallizes well from absolute alcohol, ethyl nitrate, or from a mixture of chloroform and naphtha. It is not readily decomposed by boiling with dilute caustic potash, ammonia being evolved very slowly. It is moderately soluble in cold concentrated sulphuric acid, and on diluting this solution a white precipitate separates (probably the sulphate). We have also endeavored to prepare the diimine by passing dry ammonia gas into various solutions of succinylosuccinic ester, but the results were generally unsatisfactory.

An attempt was made to prepare a naphtetrazine by the action of a pseudothiourea upon this diimine. One molecule of the diimine and two of pseudomethylthiourea hydriodide were dissolved in warm 95 per cent. alcohol, two molecules of sodium ethylate added, and the mixture boiled seven hours under a return condenser. The odor of mercaptan was evident, but no precipitate separated, and, on cooling, the diimine crystallized out unchanged. The boiling was then continued for seven hours longer, but no naphtetrazine could be detected in the product.

p-Diaminoterephthalic Acid Diethyl Ester,



$$\begin{matrix} (1)\ H_5C_2OOC \\ (2)\ H_2N \end{matrix} \Big> C_6H_2 \Big< \begin{matrix} COOC_2H_5\ (4) \\ NH_2\ (5) \end{matrix}$$

—Succinylosuccinic ester diimine was oxidized to diaminoterephthalic ester by dissolving the diimine in concentrated sulphuric acid and adding bromine, as recommended by Baeyer.[1] The oxidation proceeds very satisfactorily at 40-50°, but the "clouding" of the solution, which Baeyer gives as the indication of the completion of the reaction, was not observed. Although we continued the oxidation much longer than he, the solution remained quite clear, with excess of bromine undissolved. The oxidation is apparently complete in half an hour. The solution is then decanted from excess of bromine, the bromine in the solution blown out by a stream of dry air, and the liquid poured upon cracked ice wet with sulphurous acid. The difficultly soluble sulphate of the diamino ester separates as a white precipitate. The mixture is left in an ice pack for an hour or two, the precipitate filtered out, washed well with ice water, suspended in a small amount of water and treated with saturated sodium acetate solution, whereby the free ester is separated as an orange-yellow flocculent precipitate. When dry, it forms an orange-yellow powder. On adding this dry ester to boiling absolute alcohol, it immediately turns a brilliant red and dissolves, crystallizing from the solution on cooling in orange-red prisms, about the color of potassium dichromate. Frequently,

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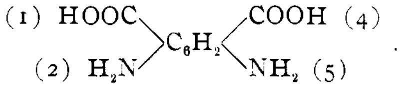
[1] *Loc. cit.* In the oxidation of succinylosuccinic ester itself to dihydroxyterephthalic acid, concentrated sulphuric acid appears to be just as good a solvent as carbon disulphide.

it separates in light yellow crystals of similar habit, which, on recrystallization; change to the orange-red variety. This dimorphism was first reported by Häussermann and Martz.[1] The ester is apparently insoluble in water or naphtha; difficultly soluble in ether, carbon tetrachloride, carbon disulphide, cold absolute alcohol, cold benzene, or cold ethyl acetate; moderately soluble in the last three solvents at their boiling-points, in cold acetone, cold glacial acetic acid, cold nitrobenzene, cold aniline, or cold ethyl acetate; easily soluble in the five latter solvents at their boiling-points, or in chloroform. Its solutions are generally golden brown with a yellow fluorescence.

Some of the ester was treated with excess of acetic anhydride. Heat was evolved and the ester gradually dissolved, but no acetyl derivative could be isolated from the product. Fusion of the ester with urea or with thiourea likewise gave negative results.

The ester reacts with phenyl isocyanate and with phenyl isothiocyanate, and the general behavior of the products is much like that of the naphtetrazine described beyond. Further work upon the subject is requisite, before their nature can be positively stated. The product obtained with phenyl isothiocyanate is yellow, very difficultly soluble in water, absolute alcohol, amyl alcohol, amyl acetate or valerate, ethyl nitrate, naphtha, ether, chloroform, carbon tetrachloride, carbon disulphide, acetone, xylene, or nitrobenzene, but dissolves in phenol, aniline, pyridine, or quinoline. The solutions all show a strong yellowish green fluorescence. It dissolves in caustic or carbonated alkali or in ammonium hydroxide, and is reprecipitated from these solutions by acids.

p-Diaminoterephthalic Acid,



$$\begin{matrix}(1)\ HOOC & & COOH\ (4)\\ & C_6H_2 & \\ (2)\ H_2N & & NH_2\ (5)\end{matrix}.$$

was prepared by saponifying the above ester.[2] It is not necessary to use the free ester for this purpose; the sulphate of the ester obtained in the above oxidation of succinylosuccinic ester diimine will do just as well. By boiling this sulphate with excess of aqueous alkali, and precipitating with acetic acid, the diamino acid is obtained as a yellowish green micro-crystalline

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[1] *Ber.*, **26**, 2984 (1893).

[2] Böninger: *Loc. cit.*

solid. It may be purified by dissolving in alkali and reprecipitating with acetic acid. It is insoluble in all ordinary solvents except glycerol and caustic alkalies. It is infusible and exceedingly inert. Boiling acetic anhydride had no appreciable effect upon it, nor could an acetyl derivative be obtained through its silver salt.[1] The silver salt was suspended in dry ether and treated with acetyl chloride; it was also heated with acetyl chloride for six hours in a sealed tube at 150°, but in neither case did any reaction occur. The acid was not attacked by boiling phenyl isothiocyanate. Fusions with formamide,[2] with urea,[3] or with thiourea[4] were also fruitless. At high temperature, the acid decomposes with formation of a small amount of substance which gives the reactions of *p*-phenylenediamine.

REACTIONS WITH SUCCINYLOSUCCINIC ESTER.

Behavior of Succinylosuccinic Ester with Urea, and with Phenyl Isocyanate.—Succinylosuccinic ester and urea were boiled together in absolute alcohol solution, but no reaction ensued. Fused zinc chloride was then added to the solution, but its only effect was to form a yellow, insoluble zinc compound with the ester, which was decomposed by concentrated hydrochloric acid into zinc chloride and the ester again. Fusion of the ester with urea also failed to induce any condensation.

Succinylosuccinic ester dissolves freely in hot phenyl isocyanate, and crystallizes out unchanged on cooling.

Succinylosuccinic Ester and Guanidine Salts.—One molecule of succinylosuccinic ester and two of guanidine thiocyanate were dissolved in absolute alcohol, and the solution boiled six hours under a return condenser. On cooling, practically all the ester crystallized out unchanged. The mixture was again heated and two molecules of sodium ethylate added. The surface of the ethylate instantly changed to a bright red, the solution turned yellow, and a yellowish white precipitate separated. The ethylate rapidly dissolved and the precipitate gradually increased in amount. After boiling for three hours, the solution was filtered hot, and the precipitate washed with alcohol. It is sufficiently soluble in alcohol to give a red color with ferric chloride solution, but this color is not soluble in ether. The precipitate, when dry,

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[1] Wheeler: *Am. Chem. J.*, **20**, 222.

[2] Niementowski: *J. prakt. Chem.* [2], **51**, 564 (1895).

[3] Niementowski: *Ibid.*, **40**, 1 (1889).

[4] Stewart: *Loc. cit.*

appeared as a yellow powder. It was purified through its sodium salt. The purified salt was dissolved in hot caustic soda, and excess of hydrochloric acid added, to precipitate the white hydrochloride. This was treated, without filtering, with excess of ammonium hydroxide. The free base then appeared as a yellow flocculent precipitate. On careful heating, a slight yellow sublimate was obtained, but most of it charred and gave off a strong odor of ammonia. It is apparently insoluble in water, glycerol, ether, ethyl acetate, amyl valerate, acetone, carbon disulphide, chloroform, carbon tetrachloride, benzene, nitrobenzene, aniline, phenol, pyridine, or quinoline. In alcohol, its solubility is only sufficient to give the color reaction with ferric chloride. It dissolves freely in dilute caustic alkalies, but is reprecipitated by the addition of concentrated alkali. It is soluble in concentrated nitric or sulphuric acids, but not in concentrated hydrochloric acid. Its solution in concentrated sulphuric acid is reprecipitated by sodium acetate. It dissolves also in glacial acetic acid, and is reprecipitated by the addition of ammonium hydroxide but is redissolved by excess of the precipitant.

The purified free base was analyzed with the following results: Found: C, 48.56, 48.68; H, 4.16, 4.12; N, 32.54, 32.16, 32.30, 32.63. Calculated for $C_{10}H_{10}O_2N_6$; C, 48.78; H, 4.06; N, 34.15.

It will be noted that the figures for nitrogen are nearly 2 per cent. too low. The reason for this we do not know. The nitrogen was determined by various methods, but the results were invariably too low. We found some comfort, however, upon examining the literature, to see that other experimenters, dealing with similar heavy molecules very rich in nitrogen, have had similar troubles. That our product is really a naphttetrazine seems fairly well established by the other analytical results, both for the free base and for its sodium salt, and, further, by the fact that we have recently prepared a similar condensation product from succinylosuccinic ester and acetamidine, the analysis of which resulted much more satisfactorily[1].

Hydrochloride.—This is colorless, insoluble in hydrochloric acid, and less stable than the sulphate, being dissociated by water.

Sulphate.—The free base was dissolved in concentrated sulphuric acid, the solution diluted carefully and allowed to stand some time.

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[1] A paper will shortly appear on this condensation-product.

The sulphate gradually separated as a colorless crystalline precipitate, composed of beautiful, microscopic, glassy rhombohedra.

Sodium Salt.—The free base dissolves very easily in dilute caustic soda, but the sodium salt thus formed is insoluble in the strong alkali. The base is, therefore, suspended in water, brought into solution by the addition of a little concentrated caustic soda, and the solution heated to boiling. Concentrated caustic soda is run in until the sodium salt begins to separate in the boiling solution. The heating is then stopped, and, on cooling, all the sodium salt crystallizes out in yellow needles or prisms. The alkaline solution of the crude base turns red in the air, but this is apparently due to a small amount of impurity, as after several recrystallizations from caustic soda the alkaline solution no longer shows this behavior. The pure salt forms light yellow crystals with a beautiful green fluorescence. Its aqueous solution shows the same fluorescence, but the addition of strong caustic soda precipitates it so completely that not a trace of fluorescence remains in the mother-liquor, while the separated crystals are strongly fluorescent.

The crystals contain six molecules of water of crystallization. They are quite stable in the air, and lose water when washed with absolute alcohol. In aqueous solution it evidently dissociates. It is decomposed by acids. It is very difficult to purify, the best agent for washing the crystals free from the alkaline mother-liquor being 95 per cent. alcohol, but even this appears to cause some dissociation.

The water of crystallization was determined with the following result: Water found, 27.70, 27.71. Calculated for $C_{10}H_8O_2N_6Na_2.6H_2O$, 27.14.

The dried substance was then analyzed for sodium: Na found, 15.10, 15.14. Calculated for $C_{10}H_8O_2N_6Na_2$, 15.86.

The alkaline solution of the sodium salt gives no precipitates with alkaline solutions of lead, aluminum, or zinc salts, but is precipitated by carbon dioxide or acetic acid, the precipitate redissolving in excess of acetic acid.

Potassium Salt.—This was prepared in the same way as the sodium salt. Crystallized from caustic potash, it forms needles of a paler yellow than the sodium salt. It is much more soluble in strong alkali than the latter, and hence still more difficult to purify.

Guanidine acetate, used instead of the thiocyanate, gave the same condensation-product.

An attempt was made to prepare the naphttetrazine by direct fusion of succinylosuccinic ester and guanidine acetate, in the same way that the diimine was made from succinylosuccinic ester and ammonium acetate, but no condensation occurred.

Finally, a solution of free guanidine was prepared from the carbonate and condensed with succinylosuccinic ester, but this variation showed no advantage over the use of a guanidine salt and sodium ethylate either in the yield or purity of the product.

HAVEMEYER LABORATORIES, COLUMBIA UNIVERSITY,
April, 1905.

[CONTRIBUTION FROM THE JOHN HARRISON LABORATORY OF CHEMISTRY, NO. 90.]

DOUBLE FLUORIDES OF TANTALUM.[1]

BY CLARENCE W. BALKE.

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THE results set forth in this article constitute a chapter in the reinvestigation of the compounds of columbium and tantalum now in progress in this laboratory. While partly confirmatory in nature, they mark a distinct advance in certain directions, and, it is hoped, will materially aid in a continuation of the investigation.

Berzelius[2] first prepared and analyzed the potassium salt, $2KF.TaF_5$. In addition he mentioned a calcium, a magnesium, and a lead salt, and further, a potassium salt containing as much as 63 per cent. of tantalic acid, and an ammonium salt, but these were not analyzed, nor were any formulas assigned to them.

H. Rose[3] prepared and analyzed the potassium salt and also the 3 : 1 sodium salt, $3NaF.TaF_5$.

Marignac,[4] in 1866, published the most complete investigation of these salts which has been made up to the present time. He studied the ammonium, potassium, sodium, zinc and copper salts.

A. Joly[5] described an ammonium oxyfluoride of tantalum of the

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[1] From author's thesis presented to the University of Pennsylvania for the Ph.D. degree.

[2] *Pogg. Ann.*, **4**, 6 (1825).

[3] *Ibid.*, **99**, 481 (1856).

[4] *Bibl. Univ. Arch.*, **26**, 89; *Ann. ch. Phys.* [4], **9**, 247 (1866).

[5] *C. R.*, **81**, 1266 (1875).