unexplained reactions have also been referred to the directions of the valences.

The existence of "partial valence" is shown to follow from the electric charges of the atoms in a molecule.

Applications of the hypothesis to inorganic compounds have been taken from the work of A. A. Noyes and of Sir William Ramsay.

MASSACHUSETTS INSTITUTE OF TECHNOLOGY, BOSTON,
COLUMBIA UNIVERSITY, NEW YORK.

[CONTRIBUTION FROM THE HAVEMEYER LABORATORIES OF COLUMBIA UNIVERSITY. No. 183.]

RESEARCHES ON QUINAZOLINES (TWENTY-SIXTH PAPER). THE SYNTHESIS OF SOME STILBAZOLES, HYDRAZONES AND SCHIFF BASES IN THE 4-QUINAZOLONE GROUP.[1]

BY MARSTON TAYLOR BOGERT, GEORGE DENTON BEAL AND CARL GUSTAVE AMEND.

Received September 24, 1910.

Various papers from this laboratory[2] have shown that in the 4-quinazolone group it is a relatively simple matter to prepare derivatives carrying a methyl group in position 2 and amino groups on either or both the benzene and miazine portions of the nucleus. Such substances constitute interesting material for the study of the action of these various groups with aldehydes.

For, it is well known that methyl groups on a nuclear carbon adjacent to the nitrogen of a heterocycle, as, for example, in the α-picolines, the quinaldines, and the like, easily condense with aldehydes to compounds of the type R.CH : CH.R', in which R represents the heterocycle and R' the radical of the aldehyde used.[3] For compounds of this type where R' is a simple or substituted benzene nucleus, the name "stilbazole" has been introduced,[4] on account of the structural similarity of such substances to the stilbenes.

The condensation of aldehydes with *Bz*-amino compounds, or anilines, to compounds of the R.N:CH.R', or Schiff base type, is also an old story.

Bülow[5] and others[6] have shown that aldehydes condense to hydrazones with the *N*-amino groups of nitrogen heterocycles.

[1] Read at San Francisco meeting of the Society, July 15, 1910.

[2] Bogert *et al.*, THIS JOURNAL, **32**, 784 and 1297 (1910).

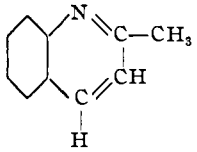
[3] Jacobsen and Reimer, *Ber.*, 16, 2006 (1883); Wallach and Wusten, *Ibid.*, 16, 2008 (1883); Döbner and Miller, *Ibid.*, 18, 1646 (1885); Ladenburg, *Ibid.*, 19, 439 (1886); Baurath, *Ibid.*, 20, 2719 (1887), and 21, 818 (1888); Bulach, *Ibid*, 20, 2047 (1887); Heyman and Königs, *Ibid.*, 21, 1424 and 2167 (1888); Eckhardt, *Ibid.*, 22, 279 (1889); Busch and Königs, *Ibid.*, 23, 2682 (1890); von Grabski, *Ibid.*, 35, 1956 (1902); Löw, *Ibid.*, 36, 1666 (1903); and others.

[4] Baurath, *Loc. cit.*

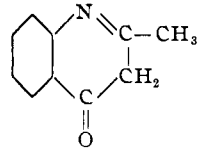
[5] *Ber.*, 40, 4749 (1907).

[6] Bogert and Gortner, THIS JOURNAL, 31, 947 (1909).

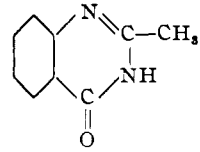
The first 4-quinazolone studied by us was the 2-methyl-4-quinazolone, whose structure resembles that of quinaldine or, still more closely, that of γ-quinaldone,



Quinaldine.

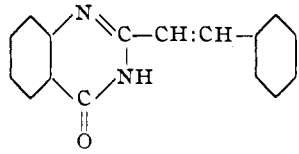


γ-Quinaldone.



2-Methyl-4-quinazolone.

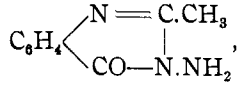
It condenses easily with aromatic aldehydes to the stilbazole; with benzaldehyde, for example, giving the simple styryl derivative,



Boiling with caustic alkali, or with strong hydrochloric acid, fails to hydrolyze these compounds to aldehyde and quinazolone again. By virtue of the —CO.NH— $\rightleftarrows$ —C(OH) : N —→ grouping in the quinazoline portion of the molecule, they dissolve in caustic alkalies and are reprecipitated from such solutions by carbon dioxide or by weak organic acids.

2,3-Dimethyl-4-quinazolone gives the corresponding 2-styryl-3-methyl-4-quinazolone with benzaldehyde. As the condensation product in this case cannot assume the enolic condition, it is insoluble in caustic alkali solutions.

Passing on to the 2-methyl-3-amino-4-quinazolone,



the opportunity was presented for the aldehyde to condense with either the 2-methyl or the 3-amino group, giving in the one case a stilbazole (*C*-benzal), in the other a hydrazone (*N*-benzal), or, finally, with excess of aldehyde, condensation might be effected with both groups simultaneously, yielding a compound which would be both stilbazole and hydrazone in structure. As will be seen from the experiments described beyond, representatives of all three of these classes have been obtained. In connection with these experiments, the following observations are of interest:

1. With benzaldehyde and the quinazolone in equimolecular proportion, the condensation occurs first with the *N*-amino group, giving the hydrazone.

2. With excess of aldehyde, the dimolecular condensation follows, *i. e.*, aldehyde condenses with both the methyl and the amino group

3. The hydrazone is readily hydrolyzed by boiling dilute hydrochloric acid, while the stilbazole is unaffected by this treatment. Hence, on boiling the dimolecular condensation product with 10 per cent. hydrochloric acid, the stilbazole, 2-styryl-3-amino-4-quinazolone, which could not be prepared by direct condensation, was easily obtained.

4. With cinnamic aldehyde, salicylic aldehyde, or vanillin, the aldehyde condenses only with the *N*-amino group. Heating either these hydrazones or the original 2-methyl-3-amino-4-quinazolone with excess of the aldehyde does not give the di-aldehyde condensation product, although, as has been mentioned already, these aldehydes condense freely with the 2-methyl in the absence of the 3-amino group. But, if these hydrazones be heated with benzaldehyde, then the 2-methyl is immediately changed to the styryl group. From which, it might be inferred either that this 2-methyl group is much more reactive towards benzaldehyde than towards the other aromatic aldehydes mentioned, or that if the failure of the other aldehydes to give di-aldehyde condensations is due to steric hindrance, the presence of another aldehyde group in position 3 does not prevent the entrance of the benzal group in position 2.

2-Methyl-7-amino-4-quinazolone presents a slightly different condition of affairs. Like the 2-methyl-3-amino-4-quinazolone, condensations are possible here with the methyl, the amino group, or with both. The amino group is, however, differently located in the molecule, being on the benzene and not on the miazine side of the nucleus, and in union with carbon instead of with nitrogen. Aldehydes condensing with this amino group should thus yield true Schiff bases instead of hydrazones.

In our first experiment with the 2-methyl-7-amino-4-quinazolone and benzaldehyde, a mono-benzal derivative was obtained which was apparently the Schiff base, since it could be hydrolyzed into aldehyde and quinazolone again by long boiling with dilute potassium hydroxide solution. Subsequent attempts to get this same product all failed. Irrespective of the amount of benzaldehyde used with the quinazolone, the same product resulted in all other experiments, but a product different from the one obtained in the first experiment. The fact that this product obtained in most of the experiments can be acetylated would argue the presence therein of an unchanged amino group. We do not feel satisfied with the results with this particular quinazolone, and if an opportunity presents will repeat the work. It is contrary to our experience in other cases that the 7-amino group should condense more readily with benzaldehyde than the 2-methyl, and the mono-benzal derivative isolated in the first experiment must be obtained again and further studied before we are convinced that in it the benzaldehyde has condensed with the 7-amino group.

No such complications appeared in the interaction of 2-methyl-7-

acetamino-4-quinazolone and benzaldehyde, which resulted in a smooth condensation between the aldehyde and the 2-methyl group.

In the case of 2,3-dimethyl-7-amino-4-quinazolone also, the mono-benzal condensation product obtained proved to be the styryl compound, *i. e.*, the stilbazole, from which it can be gathered that, under the conditions of our experiment at least, the 2-methyl reacts with benzaldehyde more rapidly than the 7-amino group.

Finally, in the 2-methyl-3,7-diamino-4-quinazolone, we find three groups capable of taking part in the reaction with aromatic aldehydes, yielding respectively stilbazoles, hydrazones, or Schiff bases, or compounds containing two or all of these functions simultaneously.

On heating this quinazolone with excess of benzaldehyde, all three groups reacted, the tri-benzal derivative being the chief product. At the same time there were formed two isomeric di-benzal derivatives, but in amount insufficient to identify.

When the 7-amino group was acetylated, the 2-methyl and 3-amino groups condensed smoothly with the benzaldehyde. The 6-acetamino isomer behaved similarly. When both 3- and 7-amino groups were acetylated, the 2-methyl group could be similarly condensed with the aldehyde.

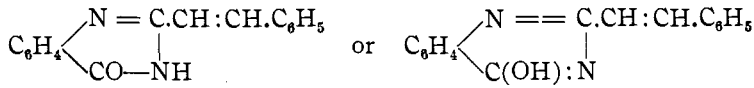
Summing up, then, our results would seem to show that, with reference to their activity towards benzaldehyde, and under the conditions of our experiments, the above groups can be arranged in the following order: first, the 3-(or *N*-) amino group; second, the 2-methyl group; and third, the 7-(or *Bz*-) amino group.

Experimental.

I. Condensations with 2-Methyl-4-quinazolone.

With the exception of the first, compounds carrying the —CO.NH— $\rightleftarrows$ —C(OH) : N— group are written in the keto form and called quinazolones.

2-Styryl-4-quinazolone (2-Styryl-4-hydroxyquinazoline),



2-Methyl-4-quinazolone was mixed with slightly more than the equimolecular amount of benzaldehyde and the mixture boiled for ten minutes over the naked flame. The melt, crystallized from alcohol, yielded colorless, silky needles, melting at 252–3° (cor.).

Found: N, 11.86. Calculated for $C_{16}H_{12}ON_2$: N, 12.0.

The compound is rather difficultly soluble in alcohol, chloroform or glacial acetic acid; very difficultly soluble in ether or carbon disulphide. The benzal group is not broken off by boiling with potassium hydroxide

solution or with hydrochloric acid. With bromine, in chloroform or glacial acetic acid solution, it yields a mono-bromo substitution product. No di-bromo addition was noted.[1] An effort was made to oxidize the styryl quinazolone to the 4-quinazolone-2-carboxylic acid by the method of Bamberger and Berle,[2] but the results were unsatisfactory.

In an attempt to get the intermediate alkine (*i. e.*, $C_9H_5ON_2.CH_2.CH(OH).C_6H_5$), equimolecular amounts of 2-methyl-4-quinazolone and benzaldehyde, mixed with some water, were heated in a sealed tube for 40 hours at 140°. At the close of the heating the tube was found to be filled with crystals, which when recrystallized from alcohol proved to be identical with the above 2-styryl-4-quinazolone.

2-o-Hydroxystyryl-4-quinazolone, $C_6H_4\begin{cases}N=C.CH:CH.C_6H_4.OH(o)\\ |\\ CO.NH\end{cases}$ — An equimolecular mixture of 2-methyl-4-quinazolone and salicylic aldehyde was boiled for ten minutes and the product crystallized from alcohol. Minute, pale yellow needles separated, melting with decomposition at 307°.

Found: N, 10.92. Calculated for $C_{16}H_{12}O_2N_2$: N, 10.60.

The substance is difficultly soluble even in hot alcohol. With hydrochloric acid or with potassium hydroxide, it yields bright canary-yellow salts.

2-m-Methoxy-p-hydroxystyryl-4-quinazolone, $C_6H_4\begin{cases}N=C.CH:CH.C_6H_3(OH)(p)(OCH_3)(m)\\ |\\ CO.NH\end{cases}$ —Equimolecular amounts of 2-methyl-4-quinazolone and vanillin were fused together over the flame until crackling ceased (ten minutes). The cold melt was crystallized repeatedly from alcohol. Pale yellow, minute needles resulted, which softened at about 275° (cor.) and at 280° (cor.) melted down to a brownish liquid.

Found: N, 9.63. Calculated for $C_{17}H_{14}O_3N_2$: N, 9.52.

The alkaline salts are a darker yellow, but possess no tinctorial power.

II. Condensation with 2,3-Dimethyl-4-quinazolone.

2-Styryl-3-methyl-4-quinazolone, $C_6H_4\begin{cases}N=C.CH:CH.C_6H_5\\ |\\ CO.N.CH_3\end{cases}$ — 2,3-Dimethyl-4-quinazolone and benzaldehyde were boiled together until the crackling ceased, and the cold melt was then crystallized from dilute alcohol. Fine, light yellow needles were obtained, melting at 170° (cor.).

Found: N, 10.72. Calculated for $C_{17}H_{14}ON_2$: N, 10.68.

To make certain that the 2-methyl group was the only one participa-

[1] Compare Wallach and Wusten, *Ber.*, 16, 2009 (1883); Dubke, *Ibid.*, 27, 79 (1894).

[2] *Ann.*, 273, 330.

ting in the above reaction, some 3-methyl-4-quinazolone was heated with benzaldehyde for 6–8 hours at 180°, but no condensation ensued.

III. Condensation with 2-Methyl-3-amino-4-quinazolone.

The 2-methyl-3-amino-4-quinazolone was prepared by the method of Bogert and Gortner,[1] from acetoanthranil and hydrazine hydrate.

In one case, however, another product was isolated, which we believe to be

Acetoanthranilic acetohydrazide, $CH_3CONH.C_6H_4.CONHNHCOCH_3$.—Acetoanthranil was stirred into an excess of aqueous hydrazine hydrate solution and the mixture warmed until complete solution resulted. Alcohol was then added and on dilution with water a white precipitate separated, melting at 180°, and thought at first to be impure acetoanthranilic acid (m. p. 186°). It was therefore boiled with acetic anhydride, to regenerate the acetoanthranil, but on cooling a product was obtained which crystallized from alcohol or from acetic anhydride in prisms, melting at 193° (cor.).

Found: N, 17.98. Calculated for $C_{11}H_{13}O_3N_3$: N, 17.94.

2-Methyl-3-benzalamino-4-quinazolone, $C_6H_4\begin{cases}N=C.CH_3\\ \quad\quad|\\ CO.N.N:CH.C_6H_5\end{cases}$—This has already been described by Bogert and Gortner,[2] who prepared it by boiling together for a few minutes 2-methyl-3-amino-4-quinazolone and benzaldehyde. It forms colorless prismatic needles. They give its melting point as 183° (cor.). By careful purification, we have succeeded in raising this to 187° (cor.). The condensation may be accomplished by boiling together the two constituents dry or, still better, by boiling them together in alcoholic solution.

Further heating with benzaldehyde, converts this mono-benzal derivative into the di-benzal compound described beyond.

Hydrochloride.—The powdered base was suspended in dry ether and a stream of dry hydrogen chloride passed in. When the reaction was complete, the precipitate was filtered out, washed with dry ether, and dried in vacuum over solid potassium hydroxide and concentrated sulphuric acid. It was then a nearly colorless amorphous powder, softening at 220° and decomposing without melting at about 300°.

Found: N, 14.15; Cl, 11.53. Calculated for $C_{16}H_{13}ON_3.HCl$: N, 14.01; Cl, 11.85.

2-Styryl-3-benzalamino-4-quinazolone, $C_6H_4\begin{cases}N=C.CH:CH.C_6H_5\\ \quad\quad|\\ CO.N.N:CH.C_6H_5\end{cases}$— 2-Methyl-3-amino-4-quinazolone and benzaldehyde were mixed, in the proportion of one molecule of the former to two of the latter, and the

[1] THIS JOURNAL, **31**, 947 (1909).

[2] *Loc. cit.*

mixture boiled for five or ten minutes. The solution was allowed to cool somewhat and was then poured into ten volumes of hot alcohol. On cooling, yellow crystals separated, which were purified by recrystallization from alcohol and then appeared in minute, nearly colorless stellate tufts, melting at 155° (cor.).

Found: N, 12.04. Calculated for $C_{23}H_{17}ON_3$: N, 11.96.

The benzal group attached to the nitrogen is easily broken out of this compound by hydrolysis with dilute mineral acid or with caustic alkali, while that attached to the CH is unaffected by such treatment. Even on boiling in simple alcoholic solution, the benzalamino group seems to suffer partial or slow hydrolytic cleavage, for the odor of benzaldehyde soon becomes quite pronounced in the recrystallization of the compound.

The same substance was obtained by heating either 2-styryl-3-amino-4-quinazolone or 2-methyl-3-benzalamino-4-quinazolone with benzaldehyde.

Hydrochloride.—Some of this was separated in the hydrolysis of the above substance with hydrochloric acid. It is a yellowish solid, not melting below 300°.

Found: N 10.52. Calculated for $C_{23}H_{17}ON_3.HCl$: N, 10.83.

2-Styryl-3-amino-4-quinazolone, $C_6H_6\begin{cases}N=C.CH:CH.C_6H_5\\ \quad\quad|\\ CO.N.NH_2\end{cases}$—2-Styryl-3-benzalamino-4-quinazolone was stirred into an excess of boiling dilute (10 per cent.) hydrochloric acid, and the benzaldehyde formed driven out with steam. The resultant clear solution was precipitated with sodium hydroxide and the yellow resinous precipitate purified by repeated crystallization from alcohol. Snowy plates, or broad needles, were thus obtained, melting at 164° (cor.).

Found: N, 16.03. Calculated for $C_{16}H_{13}ON_3$: N, 15.97.

Heated with benzaldehyde, the 2-styryl-3-benzalamino-4-quinazolone is reproduced.

2-Styryl-3-benzoylamino-4-quinazolone.—The above quinazolone was suspended in water and treated with benzoyl chloride and a little caustic alkali. The crude product crystallized from alcohol in colorless needles, melting at 195° (cor.).

Found: N, 11.59. Calculated for $C_{23}H_{17}O_2N_3$: N, 11.44.

2-Methyl-3-cinnamalamino-4-quinazolone,

$C_6H_4\begin{cases}N=C.CH_3\\ \quad\quad|\\ CO.N.N:CH.CH:CH.C_6H_5\end{cases}$.—An equimolecular mixture of 2-methyl-3-amino-4-quinazolone and cinnamic aldehyde was boiled for ten minutes. The crude product was crystallized from alcohol, treated with boneblack in alcoholic solution, and finally recrystallized from dilute

alcohol. It then appeared in broad, bright yellow needles, melting at 148–9° (cor.).

Found: N, 14.23. Calculated for $C_{18}H_{15}ON_3$: N, 14.53.

2-Methyl-3-salicalamino-4-quinazolone, $C_6H_4\begin{cases}N=C.CH_3\\ \quad\quad|\\ CO.N.N:CH.C_6H_4.OH(o)\end{cases}$

—A mixture of 2-methyl-3-amino-4-quinazolone and salicylic aldehyde, in the proportion of one molecule of the former to two of the latter, was boiled ten minutes, the solution cooled somewhat, and poured into five times its volume of hot alcohol. On cooling, short, pale yellow needles separated, melting at 171° (cor.).

Found: N, 14.91. Calculated for $C_{16}H_{13}O_2N_3$: N, 14.53.

On hydrolysis with hydrochloric acid or with potassium hydroxide solution, salicylic aldehyde is split out. Long boiling with excess of salicylic aldehyde fails to introduce another aldehyde group. It does not yield an acetamino derivative with acetic anhydride.

Potassium Salt.—Bright yellow.

Hydrochloride.—The quinazolone was suspended in dry ether and dry hydrogen chloride passed in. The hydrochloride is a bulky yellowish powder, melting with decomposition at about 250°.

Found: N, 13.50. Calculated for $C_{16}H_{13}O_2N_3.HCl$: N, 13.31.

2-Styryl-3-salicalamino-4-quinazolone, $C_6H_4\begin{cases}N=C.CH:CH.C_6H_5\\ \quad\quad|\\ CO.N.N:CH.C_6H_4.OH(o)\end{cases}$

—While the 2-methyl-3-salicalamino-4-quinazolone refused to condense with another molecule of salicylic aldehyde, it did condense with benzaldehyde when the two were boiled together for about ten minutes. The product crystallized from alcohol in yellowish needles, melting at 232–3° (cor.).

Found: N, 11.60. Calculated for $C_{23}H_{17}O_2N_3$: N, 11.44.

2-Methyl-3-vanillalamino-4-quinazolone, $C_6H_4\begin{cases}N=C.CH_3\\ \quad\quad|\\ CO.N.N:CH.C_7H_7O_2\end{cases}$ —

2-Methyl-3-amino-4-quinazolone was fused with twice the calculated amount of vanillin until water ceased to be evolved. On cooling, a pale yellow solid was obtained. This was pulverized and crystallized twice from alcohol, giving small yellowish prisms, or minute needles, melting at 215–6° (cor.).

Found: N, 13.77. Calculated for $C_{17}H_{15}O_3N_3$: N, 13.59.

With hydrochloric acid, or with potassium hydroxide, it forms deep yellow salts which show no tinctorial power.[1] Boiled with hydrochloric acid, the odor of vanillin appears.

[1] Compare *Ber.*, **39**, 2749 (1906).

IV. Condensations with 2-Methyl-7-amino-4-quinazolone.

2-Methyl-7-amino-4-quinazolone and Benzaldehyde.—In the first experiment, the aminoquinazolone was boiled for ten minutes with excess of benzaldehyde. The clear solution on cooling deposited rosettes of needles, which on recrystallization from alcohol appeared in short, silky needles of a pale cream color, melting at 324° (cor.).

Found: N, 16.1. Calculated for $C_{16}H_{13}ON_3$: N, 15.97.

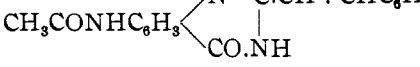
It is readily soluble in nitrobenzene, amyl alcohol or acetone; moderately soluble in boiling alcohol; difficultly soluble or insoluble in ether, chloroform, carbon tetrachloride or benzene. With concentrated hydrochloric acid, it turns a deep orange-yellow. Long boiling with dilute (10 per cent.) potassium hydroxide solution very slowly splits it into the aldehyde and quinazolone again.

Attempts to get this product a second time invariably failed. In all cases, apparently irrespective of the amount of benzaldehyde used, the product after careful purification by repeated crystallization from alcohol, melted at 324–5°. A mixture of this substance with that obtained in the first experiment showed a depression of 17° in the melting point and the analytical figures do not check either with those calculated for a mono- or a di-benzal derivative:

Found: C, 68.77 and 68.89; H, 5.38 and 4.91; N, 13.88, 13.94 and 13.72. Calculated for a mono-benzal derivative ($C_{16}H_{13}ON_3$): N, 15.97; for a di-benzal derivative ($C_{23}H_{17}ON_3$): N, 11.97.

The substance is not appreciably soluble in ether, chloroform, carbon tetrachloride or benzene. It dissolves in acetone, ethyl or amyl alcohol, or in nitrobenzene. With concentrated hydrochloric acid, it turns first yellow and then the color is discharged. With acetic anhydride, it gives an acetyl derivative which crystallizes from alcohol in colorless pearly plates, melting at 274–6° (cor.) and containing 12.57 per cent. of nitrogen.

2-Styryl-7-acetamino-4-quinazolone,



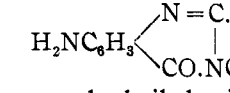
— 2-Methyl-7-acetamino-4-quinazolone was boiled with benzaldehyde and the solid product crystallized from alcohol. Short, colorless needles, melting at 323–4° (cor.).

Found: N, 13.65 and 13.94. Calculated for $C_{18}H_{15}O_2N_3$: N, 13.77.

The compound is not appreciably soluble in water or ether, but dissolves readily in acetone, ethyl or amyl alcohol, or in nitrobenzene. Treated with concentrated hydrochloric acid, it first turns intensely yellow, then the color is largely discharged. On boiling, it dissolves slowly in the acid.

V. Condensations with 2,3-Dimethyl-7-amino-4-quinazolone.

2-Styryl-3-methyl-7-amino-4-quinazolone,



—As before, the aminoquinazolone was merely boiled with benzaldehyde. On cooling, the crude product separated in yellow nodules, which were removed, washed with cold alcohol and then dissolved in hot alcohol. Water was added carefully to the hot alcoholic solution, and as the solution cooled yellow, twinned prisms crystallized out. On recrystallization, these melted at 229.5–230° (cor.). The mother liquor from these crystals showed a faint greenish fluorescence.

Found: N, 15.31. Calculated for $C_{17}H_{15}ON_3$: N, 15.16.

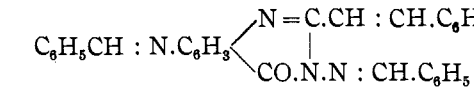
Heated with acetic anhydride, it yields the monoacetyl derivative described below.

2-Styryl-3-methyl-7-acetamino-4-quinazolone was prepared by boiling the above amino compound with acetic anhydride, as just stated, or by condensing the 2,3-dimethyl-7-acetamino-4-quinazolone with benzaldehyde. Purified by repeated crystallization from alcohol, it forms yellowish needles, melting at 272° (cor.).

Found: N, 13.32. Calculated for $C_{19}H_{17}O_2N_3$: N, 13.17.

VI. Condensations with 2-Methyl-3,7-diamino-4-quinazolone.

2-Styryl-3,7-dibenzalamino-4-quinazolone,



— 2-Methyl-3,7-diamino-4-quinazolone was boiled for a few minutes with excess of benzaldehyde. Upon cooling, a yellowish granular mass was obtained. This was extracted repeatedly with boiling alcohol, and the insoluble residue dried. This insoluble product melts at 238° (cor.) and is yellow in color.

Found: C, 79.1; H, 4.99; N, 12.29. Calculated for $C_{30}H_{22}ON_4$: C, 79.3; H, 4.85; N, 12.33.

It is readily soluble in chloroform, amyl alcohol or nitrobenzene; moderately soluble in acetone or benzene; but very difficultly soluble in ethyl alcohol or ether.

On concentrating the mother liquor from the above tri-benzal compound, a substance was isolated which on repeated crystallization from lcohol appeared in short, glistening, yellowish needles, melting fairly sharply at 196° (cor.).

Found: N, 15.47. Calculated for $C_{23}H_{18}ON_4$: N, 15.3.

This corresponds, therefore, to a di-benzal derivative. Its dilute alcoholic solution shows a greenish fluorescence.

In the mother liquor from this latter was found a third substance,

crystallizing in dark yellow glistening, plates, melting at 172° (cor.), and also showing a greenish fluorescence in alcoholic solution.

Found: N, 15.43. Calculated for $C_{23}H_{18}ON_4$: N, 15.3.

The results would indicate that the above two substances are isomeric di-benzal derivatives, but the amounts isolated were insufficient for further investigation.

2-Styryl-3-benzalamino-7-acetamino-4-quinazolone.—2-Methyl-3-amino-7-acetamino-4-quinazolone was boiled with excess of benzaldehyde for five or ten minutes. The quinazolone dissolved readily in the hot aldehyde and the water escaped with a strong crackling noise. On cooling, the crude product separated in clusters of dark-yellow needles. If necessary, the excess of aldehyde can be blown out by a current of steam. Recrystallized from alcohol, the product forms yellowish needles, melting at 261° (cor.).

Found: N, 13.96. Calculated for $C_{25}H_{20}O_2N_4$: N, 13.72.

It is practically insoluble in water, ether, or aqueous solutions of caustic alkalies, but dissolves easily in ethyl or amyl alcohol, acetone or nitrobenzene. Its alcoholic solution shows a greenish fluorescence. With cold concentrated hydrochloric acid, it turns a deep yellow, but does not dissolve to any great extent.

2-Styryl-3,7-diacetamino-4-quinazolone was prepared in a similar manner by condensing 2-methyl-3,7-diacetamino-4-quinazolone with benzaldehyde. Purified by crystallization from dilute alcohol, it melts at 283–4° (cor.), after preliminary softening a few degrees below this point.

Found: N, 15.56. Calculated for $C_{20}H_{18}O_3N_4$: N, 15.47.

It is soluble in water, ethyl or amyl alcohol, or chloroform, when hot, or in cold acetone, and dissolves with ease in hot acetone or nitrobenzene. With cold concentrated hydrochloric acid, it turns dark yellow and dissolves. It is somewhat tribo-electric.

VII. Condensation with 2-Methyl-3-amino-6-acetamino-4-quinazolone.

2-Styryl-3-benzalamino-6-acetamino-4-quinazolone was prepared like its 7-acetamino isomer by condensing the corresponding methylaminoacetaminoquinazolone with benzaldehyde. On boiling, the benzaldehyde solution changed from light yellow to dark reddish orange. When allowed to cool, a yellow crystallin cake resulted. The excess of benzaldehyde was blown out with steam, and the residual solid crystallized from alcohol. Short, yellow, silky needles were obtained, melting at 238–9° (cor.).

Found: N, 13.88. Calculated for $C_{25}H_{20}O_2N_4$: N, 13.72.

The compound is tribo-electric. It is darker yellow than its 7-acetamino isomer, more freely soluble in alcohol, and its alcoholic solutions show a stronger fluorescence.