Patented July 29, 1941 2,251,245

UNITED STATES PATENT OFFICE

2,251,245

PRODUCTION OF HYDROXY ALKYL AMINES

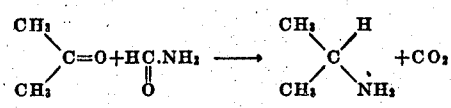
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No Drawing. Application October 14, 1939, Serial No. 299,522. In Germany October 1, 1938

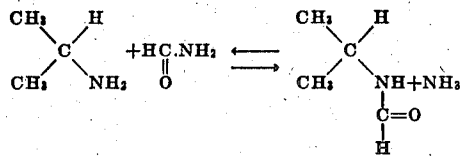
3 Claims. (Cl. 260—570.8)

The present invention relates to the production of hydroxy alkyl amines.

It is known that amines can be prepared by causing ketones to react with an excess of formamide, for instance, according to the formula:



However, by the reaction of the excess formamide with the amines so formed, the reaction product always contains a certain amount of the corresponding formylamines:



This necessitates a special saponifying treatment of the reaction mixture in order to obtain the pure amines.

It was very surprising, therefore, when I found that by causing hydroxy alkyl formylamines to react with a carbonyl compound selected from the group consisting of aldehydes and ketones, the corresponding hydroxy alkyl amines are directly obtained without the formation, in any appreciable amount, of byproducts. Especially, the reaction mixture does not contain the corresponding formyl compounds; the mixture may be employed therefore, as such, for further reactions or other uses.

Among the hydroxy alkyl amines to be used for my present invention, the hydroxy ethyl amine has proved to be most suitable; it is not necessary to use a large excess thereof, because the reaction proceeds almost quantitatively. Suitable carbonyl compounds are, for instance, benzaldehyde, acetophenone, cyclohexanone, stearylmethylketone, saturated, unsaturated, branched-chain and other ketones obtained from fatty acids, etc. The term "aliphatic ketones" as used in this specification and in the appended claims includes cyclohexanone.

In a special modification of the process of my invention, the hydroxy alkyl formylamines are prepared in the reaction mixture itself. For this purpose, formamide is heated with the hydroxy alkyl amine and the aldehyde and/or ketone which are to be brought into reaction. The formamide reacts with the hydroxy alkyl amine to form the hydroxy alkyl formylamine which latter, on heating to higher temperatures, reacts with the aldehyde and/or ketone.

The products obtainable according to my invention are suitable, for example, as intermediates for the production of textile assistants and similar products.

My invention is furthermore illustrated by the following examples, without being limited thereto. The parts are by weight:

Example 1

196 parts cyclohexanone, 183 parts hydroxy ethyl amine and 132 parts formamide are refluxed at 115° C. for half an hour. The temperature is then slowly raised to 170° C. and, after some time, to 215° C. and left at this value for 50–60 hours. Any unreacted hydroxy ethyl formylamine may be removed by washing. In this manner, 185 parts N-hydroxy ethyl cyclohexylamine boiling at 125–127° C. under 16 mm. Hg-pressure are obtained.

Example 2

440 parts of the ketones obtainable from the fatty acids prepared by oxidation of paraffines and having a boiling range of from 132–140° C. under 12 mm. Hg-pressure, 200 parts hydroxy ethyl amine and 133 parts formamide are slowly heated, while stirring, to 120° C. and, after about 3 quarters of an hour, to about 200° C.; the mixture is kept at this temperature for 50–60 hours. On working up as usual, 473 parts are obtained of an amine mixture which boils between 180 and 230° C. at 16 mm. Hg-pressure and which is clearly soluble in the calculated amount of dilute acetic acid.

Example 3

500 parts of the ketone mixture obtainable from the fatty acids prepared by the oxidation of paraffines and having a boiling range of from 140–149° C. under 12 mm. Hg-pressure, and 450 parts hydroxy ethyl formylamine are heated for 80 hours to about 200° C. On working up as usual, there are obtained 535 parts of an amine mixture boiling between 198 and 260° C. under 14 mm. Hg-pressure; this corresponds to a yield of more than 92%.

Example 4

6 parts benzaldehyde are heated to 160–220° C. for 30–50 hours with a mixture of 100 parts formamide and 138 parts hydroxy ethyl amine or, preferably, with the corresponding quantity of hydroxy ethyl formylamine which has been separately prepared. There are obtained 95 parts

hydroxy ethyl benzylamine boiling at 148–149° C. under 12–13 mm. Hg-pressure.

Example 5

180 parts acetophenone are heated with 300 parts hydroxy ethyl formylamine for about 50 hours to 170–220° C. There are obtained 146 parts hydroxy ethyl-α-phenyl ethylamine boiling at 146° C. under 12–13 mm. Hg-pressure.

Example 6

40 parts acetophenone, 60 parts 1.3-hydroxy-propylamine and 35 parts formamide are heated for about 50 hours to 160–210° C. There are obtained about 25 parts γ-hydroxy propyl phenyl ethylamine boiling at 156–158° C. at 16 mm. Hg-pressure.

Example 7

Into 214 parts hydroxy ethylamine there are dropped 213 parts formic acid methyl ester. Thereby the hydroxy ethyl formylamine is formed in the known manner. The mixture is then heated to 80° C. for half an hour and the volatile constituents are removed by heating to the same temperature under reduced pressure. The crude hydroxy ethyl formylamine obtained in this way is heated, while stirring, for 60 hours in an oil bath of 203–204° C. with 562 parts octadecyl methyl ketone (boiling point (12 mm. Hg):174–200° C.). The N-hydroxy ethyl octadecyl methyl amine obtained in this way may be purified by distallation under reduced pressure. Or any unreacted hydroxy ethyl formylamine as well as any hydroxy ethylamine which might have been formed are washed out with water; the resulting amine is sufficiently pure for many purposes. By distillation, 545 parts of the amine boiling between 206 and 245° C. (at 12 mm. Hg) are obtained.

Example 8

420 parts hydroxy propyl formylamine (obtainable from hydroxy propyl amine and formic acid methyl ester similarly as described in Example 7) are refluxed at 189° C. for about 85 hours, while stirring, with 625 parts of a ketone mixture boiling between 130 and 220° C., obtainable from the fatty acids prepared by oxidation of paraffines and having a boiling range of from 135–147° C. under 12 mm. Hg-pressure. After 10 hours from the beginning of the reaction, a small quantity of formic acid (1–3 ccms.) which acts catalytically is preferably added. On working up as usually, 675 parts of an amine mixture boiling between 190 and 256° C. (under 12 mm. Hg) and having an average molecular weight of 328 are obtained.

In order to remove the last traces of ketones from the reaction product, the amines may be transformed into their sulfates and the amines be recovered therefrom by extracting the aqueous solutions of the sulfates with suitable solvents, for instance, with benzine. In most cases, however, it is sufficient to purify the amine by fractional distillation, since it contains only small amounts of ketones (less than 5%).

I claim:

1. The process which comprises, to form hydroxy lower alkyl amines, heating to a temperature of from about 160 to about 220° C. a hydroxy lower alkyl formylamine with a carbonyl compound selected from the group consisting of aliphatic ketones, aryl aldehydes and lower alkyl-aryl ketones to split off carbon dioxide.

2. The process which comprises, to form hydroxy lower alkyl amines, heating to a temperature of from about 160 to about 220° C. a hydroxy lower alkyl amine and formamide with a carbonyl compound selected from the group consisting of aliphatic ketones, aryl aldehydes and lower alkyl-aryl ketones to split off carbon dioxide.

3. The process for preparing hydroxy lower alkyl amines which comprises heating to a temperature between about 80 and about 170° C., a mixture of a hydroxy lower alkyl amine and formamide with a carbonyl compound selected from the group consisting of aliphatic ketones, aryl aldehydes and lower alkyl-aryl ketones, to form a hydroxy alkyl formylamine in the reaction mixture and thereupon heating the mixture to a temperature between about 150 and about 260° C. to split off carbon dioxide and to form hydroxy lower alkyl amines from the hydroxy alkyl formylamine and the carbonyl compound.

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