

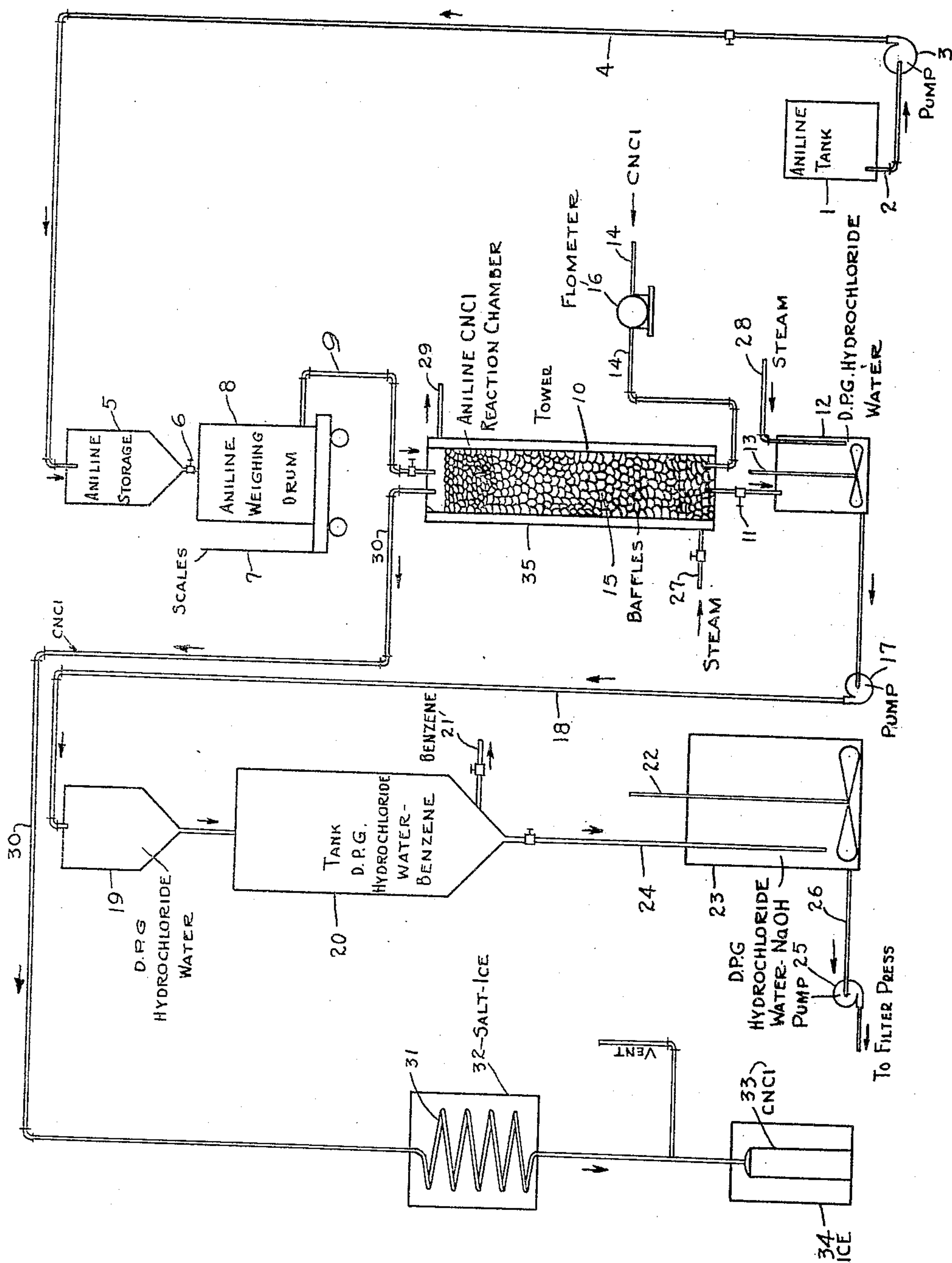
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PROCESS FOR THE CONTINUOUS PREPARATION OF A DI-SUBSTITUTED GUANIDINE

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PROCESS FOR THE CONTINUOUS PREPARATION OF A DI-SUBSTITUTED GUANIDINE

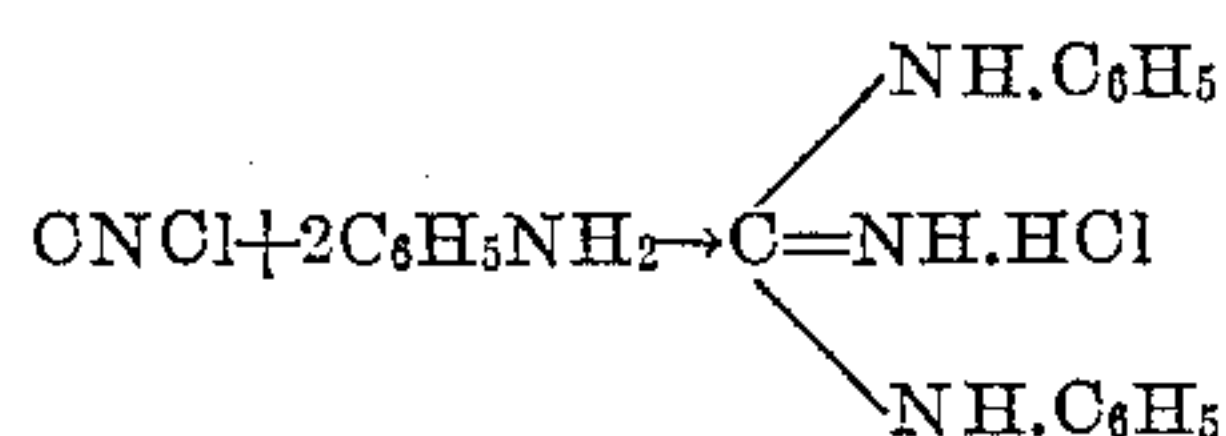
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The present invention relates to the manufacture of di-substituted guanidines, particularly the diaryl-guanidines, such as diphenyl-guanidine, the ditolyl-guanidines and the like, by a substantially continuous process, whereby high yields of a pure product are realized.

The process will be readily understood from the following description with reference to the accompanying drawing which represents in sectional view a preferred arrangement of the apparatus necessary for practicing the invention.

Referring to the drawing, aniline or other aromatic amine is placed in tank 1, from which it is removed through pipe 2, by means of pump 3, and is flowed through pipe 4 into a storage tank 5. From the tank 5 the aniline is flowed through a line controlled by a valve 6 into a measuring drum 8.

The drum 8 is placed upon an ordinary platform scale 7, whereby the rate of flow of the aniline from the drum is readily ascertained by the loss in weight indicated. The aniline flows from the drum 8 through the pipe 9 downward into the tower 10. At the same time, cyanogen chloride gas from any suitable source containing approximately 1.7% moisture is admitted to the tower 10 through the pipe 14 and passed upward in the tower counter-current to the aniline or other primary aromatic amine, where the two reagents are heated preferably between approximately 90 and 115° C. The following reaction takes place if aniline is employed as the primary aromatic amine:



The cyanogen chloride gas admitted by pipe 14 passes through flow-meter 16, whereby the rate of flow of cyanogen chloride may be determined. The tower 10 is filled with beads 15, as shown, or with any other substance such as pumice and the like, non-reactive to the materials supplied to the tower, and offering an obstructing or baffling means to the flow of liquid and gases therethrough. The tower 10 is surrounded by a heating ele-

ment 35, for example a jacket, for the admission of a heating medium, for example steam, by means of valve 27. The steam is allowed to escape from the jacket by pipe 29. A line 11, positioned at the bottom of the tower 10, permits the diphenyl or other diaryl guanidine hydro-chloride formed by the reaction of the amine and cyanogen chloride, to be dropped into tank 12. This tank is equipped with stirring means 13, where the said diphenyl-guanidine hydrochloride is dissolved in sufficient water, heated by steam from pipe 28, to form approximately a 15% solution thereof. This solution is withdrawn by pump 17 and forced through pipe 18 into chamber 19 and thence into a washing tank 20. Benzol, gasoline or other suitable solvent is supplied to the reaction product run into the tank 20 as described, in order to remove any uncombined aniline or other amine which may be present. When the washing operation is completed, the contents of the tank 20 is allowed to settle, whereupon the insoluble material, comprising the hydrochloride of diphenyl-guanidine, is withdrawn from the tank 20 and flowed through the pipe 24 into tank 23, while the benzol or other solvent employed in the washing step is removed from the tank 20 by means of pipe 21. The tank 23 is previously charged with a solution of alkaline material, for example a 15% solution of caustic soda, into which the diphenyl-guanidine hydrochloride is run. The acid salt of the aryl substituted guanidine is thereby neutralized and the free base is obtained. The slurry is agitated by means of stirrer 22 and the suspension is then removed from tank 23 through the line 26 by means of pump 25, and is run to a filter press, filtered, washed, dried and ground.

Tower 10, hereinbefore mentioned, is provided with an exit pipe to allow the removal of unreacted cyanogen chloride, which is passed by means of pipe 30 to coil 31, cooled by salt and ice in container 32. From coil 31, the condensed cyanogen chloride flows by gravity to storage, for example a cylinder 33, cooled for example by packing in ice in container 34, or may be connected by a pipe to the line 14 leading to the tower 10.

Other aryl substituted guanidines may be readily manufactured in a continuous manner by employing in place of the aniline mentioned in the example, other aromatic primary amines, such as ortho-toluidine, the m- and p-toluidines, the xylidenes and the like. By operating in the manner as described, it has been found that high yields of an aryl substituted guanidine may be readily obtained from an aromatic primary amine. Moreover, the product is of an exceptionally high degree of purity.

What is claimed is:

1. In the process of manufacturing a diaryl substituted guanidine by a continuous process, the steps in combination comprising continuously flowing vapors of cyanogen chloride containing not more than 1.7% of water counter-current to a stream of an aromatic primary amine, removing any unreacted amine from the diaryl guanidine hydrochloride thus produced by washing with a neutral organic solvent and precipitating the diaryl guanidine by the addition of an alkali of constant strength.

2. In the process of manufacturing a diaryl substituted guanidine by a continuous process the steps in combination comprising continuously flowing vapors of cyanogen chloride containing not more than 1.7% of water counter-current to a stream of an aromatic primary amine in a tower maintained at a temperature of approximately 90 to 115° C., removing any unreacted amine from the diaryl guanidine hydrochloride thus produced by washing with a neutral organic solvent and precipitating the diaryl guanidine by the addition of an alkali of constant strength.

3. In the process of manufacturing diphenyl-guanidine by a continuous process, the steps in combination comprising continuously passing vapors of cyanogen chloride containing not more than 1.7% of water counter-current to a stream of aniline in a tower at a temperature of approximately from 90 to 115° C., removing any unreacted aniline from the diphenyl guanidine hydrochloride thus produced by washing with a neutral organic solvent, and precipitating the diphenyl guanidine by the addition of an alkali of constant strength.

4. In the process of manufacturing diphenyl-guanidine by a continuous process, the steps in combination comprising continuously passing vapors of cyanogen chloride containing not more than 1.7% of water upward counter-current to a stream of aniline in a tower at a temperature of approximately from 90 to 115° C., removing any unreacted aniline from the diphenyl guanidine hydrochloride thus produced by washing with a neutral organic solvent, and precipitating the diphenyl guanidine by the addition of an alkali of constant strength.

5. In the process of manufacturing diphenyl-guanidine by a continuous process, the steps in combination comprising continuously passing vapors of cyanogen chloride containing not more than 1.7% of water upward counter-current to a stream of aniline in a tower at a temperature of approximately from 90 to 115° C., removing any unreacted aniline from the diphenyl guanidine hydrochloride thus produced by washing with benzene, and precipitating the diphenyl guanidine by the addition of sodium hydroxide without the previous addition of a weaker alkali.

6. In the process of manufacturing a diaryl substituted guanidine by a continuous process, the steps in combination comprising continuously passing vapors of cyanogen chloride containing substantially 1.7% of water upward counter-current to a stream of an aromatic primary amine in a reaction zone maintained at a temperature of approximately 90 to 115° C., forming approximately a 15% aqueous solution of the diaryl substituted guanidine hydrochloride thus produced, removing any unreacted amine therefrom by washing with a neutral organic solvent and precipitating the diaryl substituted guanidine by the addition of an alkali of constant strength.

In testimony whereof I hereunto affix my signature.

WILLIAM P. TER HORST.