

## PRODUCTION OF GASEOUS CHLORAMINE

### BACKGROUND OF THE INVENTION

This application claims a benefit of priority from  
5 U.S. Provisional Application No. 60/426,104 the entire  
disclosure of which is herein incorporated by reference.

### Field of the Invention

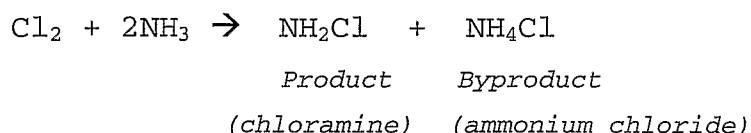
The present invention relates to a process and  
10 apparatus for producing an inorganic compound. More  
particularly, the present invention relates to a gas-  
phase process and apparatus for producing an inorganic  
compound, for example, chloramine gas, from commercially  
available gases.

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### Description of the Prior Art

An efficient process for forming chloramine is the  
reaction of chlorine gas with ammonia, as set forth in  
the reaction below:

20



25 Examples of this basic reaction are disclosed in  
U.S. Patent No. 2,837,409 to Sisler et al. and U.S.  
Patent No. 3,488,164 to Grushkin et al.

The chlorine/ammonia reaction is especially  
30 effective if it is carried out by introducing gaseous  
chlorine into a large excess of gaseous ammonia,

immediately mixing the reactants and withdrawing them from the reaction zone.

Although a highly desirable reaction, there are many  
5 drawbacks associated with the reaction of gaseous  
chlorine and gaseous ammonia. One notable drawback is  
the creation of the by-product ammonium chloride. At  
temperatures below 350°C, ammonium chloride condenses and  
solids precipitate from the product gas. The solids may  
10 foul the reactor if not properly controlled. To prevent  
or minimize this formation of solid ammonium chloride,  
the reaction must take place at a temperature above  
350°C.

15 U.S. Patent No. 4,038,372 to Colli discloses a  
process for manufacturing chloramine. The chloramine is  
formed from a gaseous reaction of chlorine and ammonia at  
about 360°C. The resulting product gas discharges to a  
discharge zone, which is heated to avoid ammonium  
20 chloride from precipitating out of the product gas. The  
product gas stream is then entrained in a high velocity  
jet of entraining gas. This gas cools the product gas  
stream and carries the gas stream to a filter system  
where the ammonium chloride solids are separated from the  
25 chloramine gas.

Great Britain Patent No. 1,149,836 discloses a  
process for the production of chloramine. The process  
includes the reaction of chlorine and ammonia in the  
30 presence of an inert diluent gas. The reaction takes  
place at a temperature of at least 250°C. The gaseous  
reaction products are maintained at a temperature of  
about 50°C to about 250°C until at least a portion of the

ammonium chloride is solidified. The ammonium chloride is collected on a glass wool filter, and thereafter, the gaseous chloramine is recovered, preferably in a solvent.

5           The present invention overcomes the burdensome problem of the formation of solids by providing a novel reactor with a solids collection and removal system downstream of the reactor. This novel removal system allows for a simple reactor design, which is easy to  
10 clean and continuously operates over longer periods of time.

#### **SUMMARY OF THE INVENTION**

          It is an object of the present invention to provide  
15 a reactor for the continuous production of inorganic compounds.

          It is another object of the present invention to provide one or more means for removing solids from the  
20 product gas stream to avoid fouling the reactor.

          It is a further object of the present invention to provide such a reactor that has a simple streamlined design.  
25

          It is yet a further object of the present invention to provide such a reactor that is easy to clean.

          It is still a further object of the present  
30 invention to provide such a reactor that has an increased continuous operation time.

These and other objects of the present invention are achieved by a gas-phase reaction of commercially available feed gases in the presence of an inert carrier gas in a novel reactor to form process gas compounds.

5 The term "feed gas" or "feed gases" is meant, for purposes of this application, to include reactive gas(es) used in the processes of the present invention. The reactor has a streamlined, compact configuration and a solids collection and removal system downstream of the  
10 reactor, where solids are efficiently removed from the product gas stream, leaving high purity product gas. This novel removal system allows for a simple reactor design, which is easy to clean and operates continuously over longer periods of time. In a preferred embodiment,  
15 the novel reactor is used to form chloramine product gas.

#### **BRIEF DESCRIPTION OF THE DRAWINGS**

Fig. 1 is a plan view of a reactor according to the present invention;

20

Fig. 2 is a plan view of a solids collection and removal system according to the present invention; and

Fig. 3 is a plan view of a reactor with a split  
25 discharge line according to an embodiment of the present invention;

Fig. 4 is a plan view of a solids collection and removal system with two collection units according to an  
30 embodiment of the present invention;

Fig. 5 is a plan view of another embodiment of a solids collection system according to the present

invention; and

Fig. 6 is a graph illustrating the abundance of chloramine with relation to ammonia and chlorine in a product gas formed according to the present invention.

#### **DETAILED DESCRIPTION OF THE INVENTION**

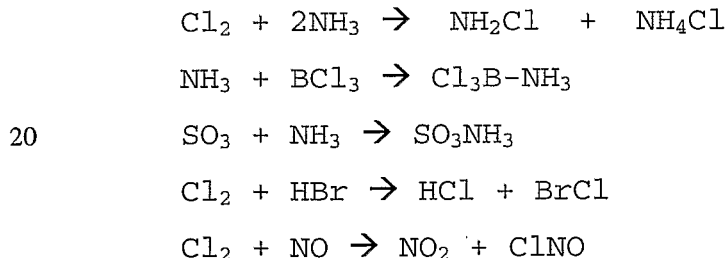
Referring to Fig. 1, a gas-phase reactor according to the present invention is represented generally by reference numeral 10. Reactor 10 has an outer shell tube 12 and an inner tube 16, both of which feed reaction chamber 18. At least one first commercially available feed gas is fed to outer tube 12 via first inlet 13. One or more inert gases is fed to outer tube 12 via second inlet 14. The one or more inert gases act as a diluent and as a carrier for the at least one first commercially available gas. At least one additional or second commercially available feed gas is concentrically fed (i.e., one or more feed tubes positioned within one or more additional feed tubes) to reaction chamber 18 via inner shell tube 16. By concentrically feeding the feed gas to reaction chamber 18, the reaction occurs immediately upon discharge of the feed gas to the reaction chamber, thus providing a more efficient reaction. As a result, a greater conversion of feed gas to product gas results, thus avoiding the undesirable formation of solids in reaction chamber 18.

Suitable commercially available feed gases for use in the reactor of the present invention include, without limitation, acetylene, ammonia, boron trichloride, boron trifluoride, butadiene, butane, butene, carbon dioxide, carbon monoxide, chlorine, cis-2-butene, deuterium,

dimethylamine, dimethyl ether, ethane, ethylene, ethyl  
chloride, hydrogen, hydrogen bromide, hydrogen chloride,  
hydrogen sulfide, isobutane, isobutylene, methane, methyl  
bromide, methyl chloride, methyl mercaptan, methylamine,  
5 nitric oxide, nitrogen dioxide, nitrous oxide, oxygen,  
perfluoropropane, propane, propylene, sulfur dioxide,  
sulfur hexafluoride, trans-2-butene, trimethylamine, or  
any combinations thereof.

10        Suitable inert gases for use in the present  
invention include, but are not limited to, nitrogen,  
argon, helium, neon, or any combinations thereof.

By way of example, reactions using the novel reactor  
15 of the present invention may include, but are not limited  
to, the following:



In one embodiment of the present invention,  
25 chloramine gas is produced using the novel reactor of the  
present invention.

Chlorine or chlorine containing gas is fed to the  
reactor via first inlet 13 and outer tube 12 at a flow  
30 rate about 0.001 ft<sup>3</sup>/min to about 0.1 ft<sup>3</sup>/min.  
Preferably, the chlorine gas is fed to the reactor at a  
flow rate about 0.01 ft<sup>3</sup>/min to about 0.05 ft<sup>3</sup>/min, and

more preferably about 0.0125 ft<sup>3</sup>/min to about 0.015 ft<sup>3</sup>/min.

The inert gas is fed to the reactor via second inlet 14 and outer tube 12 at a flow rate about 0.1 ft<sup>3</sup>/min to about 1 ft<sup>3</sup>/min. Preferably, the inert gas is fed to the reactor at a flow rate about 0.12 ft<sup>3</sup>/min to about 0.36 ft<sup>3</sup>/min, and more preferably 0.15 ft<sup>3</sup>/min to about 0.18 ft<sup>3</sup>/min.

10 Gaseous ammonia is fed to the reactor via inner shell tube 16 at a flow rate of about 0.002 ft<sup>3</sup>/min to about 0.2 ft<sup>3</sup>/min. Preferably, the gaseous ammonia is fed to the reactor at a flow rate about 0.032 ft<sup>3</sup>/min to about 0.096 ft<sup>3</sup>/min, and more preferably about 0.04 ft<sup>3</sup>/min to  
15 about 0.048 ft<sup>3</sup>/min.

A critical aspect of the present invention, when forming chloramine gas, is the pre-mixing of the chlorine gas and inert gas prior to preheating the gases.

20 Chlorine gas by itself is highly corrosive at higher temperatures. It has been found that by mixing the chlorine gas and inert gas prior to heating reduces and/or eliminates the corrosiveness of the chlorine gas. As a result, materials that are less expensive and easier  
25 to machine can be used for making reactor 10 of the present invention.

Suitable materials for constructing reactor 10 of the present invention include, but are not limited to,  
30 hastelloy C, stainless steel, brass, borosilicate glass, silicate, sodium silicate, potassium silicate, silica, or any combinations thereof. Preferably, the materials used

to construct reactor 10 include hastelloy C, stainless steel, or a combination thereof.

Another important aspect of the present invention is  
5 the preheating of all of the gases prior to their introduction to reaction chamber 18. As a result of preheating the feed gases, a smaller, more compact reactor can be used without the problem of the reactor fouling with solids, such as ammonium chloride in the  
10 case of chloramine. In addition, the higher temperatures provide higher conversion rates and/or selectivity. This provides a key advantage to continuously and efficiently producing product gas.

15 The device or element for heating the feed gases include, for example, heat tape, high resistivity wire, steam, furnace, or any combinations thereof. Preferably, heat tape is used to heat outer shell tube 12, which in turn heats both chlorine gas and inert gas flowing  
20 through outer tube 12 and the gaseous ammonia flowing through inner tube 16.

In the case of chloramine gas formation, outer shell tube 12 and inner tube 16 discharge into reaction chamber  
25 18. The chlorine gas reacts with the ammonia gas at reaction zone 20 in reaction chamber 18. Reaction chamber 18 is heated to a temperature in excess of about 350°C by one or more heating elements 22 and measured by one or more temperature sensors associated with heating  
30 elements 22. It is critical to the invention that the reaction occur at a temperature in excess of about 350°C to prevent the condensation and precipitation of ammonium

chloride, a by-product of the gas-phase reaction occurring in reaction chamber 18.

A device or element for heating reaction zone 20 include, for example, heat tape, high resistivity wire, steam, furnace, and any combinations thereof. Preferably, heat tape is used.

The product gas stream exits reaction chamber 18 via discharge tube 26 at a temperature still in excess of about 350°C.

Referring to Fig. 2, the product gas stream, via discharge tube 26, enters a solids collection system according to the present invention, represented generally by reference numeral 30. Solids collection system 30 has a trap 32 with one or more baffles 34, 36. Baffles 34, 36 help collect solids that may have precipitated out of the product gas. Following trap 32, solids collection system 30 has one or more filters 38, 40. Filters 38, 40 further collect any precipitated solids that may be in the product gas. The product gas discharges from solids collection system 30 via discharge line 42.

Any suitable filters, compatible with the desired product gas, may be used with solids collection system 30. Suitable filters for use in solids collection system 30 of the present invention include, but are not limited to, one or more cartridge filters, bag filters, granular bed filters, or any combinations thereof. Preferably, one or more cartridge filters are used. In a preferred embodiment of the present invention, one or more

cartridge filters sold under the tradenames CT-101A<sup>®</sup> and Micro-Klean III<sup>®</sup> by CUNO may be used.

Referring to Fig. 3, another embodiment of a reactor  
5 according to the present invention is represented  
generally by reference numeral 50. Reactor 50 has the  
same attributes as those described with respect to  
reactor 10 set forth above, however, reactor 50 has  
discharge tube 26 that feeds product gas to at least two  
10 solid collection system feed tubes 52, 54.

Referring to Fig. 4, a solids collection system for  
use with the reactor depicted in Fig. 3 is represented  
generally by reference numeral 60. Solids collection  
15 system 60 has the same attributes as those set forth  
above for solids collection system 30 depicted in Fig. 2,  
however, system 60 has two collection units 62, 64.

Collection unit 62 receives product gas via  
20 collection system feed tube 52. Collection unit 64  
receives product gas via collection system feed tube 54.  
Both collection units 62, 64 remove solids from the  
product gas by the same mechanisms described above for  
collection system 30 depicted in Fig. 2.

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A benefit of having more than one solids collection  
system according to the present invention is that it  
provides an end user of the reactor with various  
operating configurations to optimize the continuous  
30 production of the desired product gas. For example, the  
reactor with two or more collection system tubes, and  
corresponding collection systems, can be operated  
simultaneously on a continuous basis.

In another embodiment, the reactor with two or more collection system tubes and corresponding solids collection systems can be run in parallel, but not simultaneously. Therefore, when one or more collection  
5 systems require maintenance, those collection systems can be taken off-line, while one or more remaining collection systems either remain on-line or are put into service to replace the systems taken off-line. As a result, the continuous process never requires down time due to  
10 maintenance of the two or more solids collection systems.

It should be understood that while Fig. 3 depicts a reactor with two collection system feed tubes and Fig. 4 depicts two associated solid collection systems, one  
15 skilled in the art would appreciate that the present invention can be configured with any number of collection system feed tubes and associated solids collection systems to ensure continuous operation and production of product gas.

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Referring to Fig. 5, another embodiment of a solids collection system according to the present invention is represented generally by reference numeral 70. Solids collection system 70 has cyclone 72, to which product gas  
25 is fed via reactor discharge tube 26. Cyclone 72 is effective at removing any solids that may have precipitated out of the product gas. Any solids removed by cyclone 72 will collect in collection drum 76. Product gas exits cyclone 72 via cyclone discharge line  
30 78, which in turn feeds filter 80. Filter 80 further collects any remaining solids that may have precipitated out of the product gas. The product gas discharges from filter 80 via filter discharge line 82.

It should be understood that while Fig. 5 depicts a reactor with one solids collection system, one skilled in the art would appreciate that the present invention can be configured with any number of collection systems,  
5 similar to those set forth above with respect to Figs. 3 and 4, to ensure continuous operation and production of product gas. In addition, any combination of the solids collection systems depicted in Figs. 2, 4 and 5 may be configured, as will be appreciated by one skilled in the  
10 art.

The present invention is further illustrated by the following example.

**EXAMPLE 1**

Cl<sub>2</sub> was diluted in 12 parts of N<sub>2</sub>. Ammonia gas was charged at a stoichiometric amount with a slight excess.

5 The first run flowed 0.125 L/min Cl<sub>2</sub> mixed with 1.5 L/min N<sub>2</sub>, which was reacted with 0.3 L/min NH<sub>3</sub>. The two gases were reacted at temperatures between 350 °C to 400+ °C. The total reaction time to convert 10 kilos was 170 hours. The throughput was then increased four times by

10 increasing the flow rates by four times. However, the N<sub>2</sub> ratio was decreased three times to 2 L/min in order to increase the overall throughput while minimizing the increase in the overall flowrate.

15 In order to monitor the progress of the reaction, a GC/MS was placed in-line with the reaction. By splitting the stream exiting the second filter, one of the streams was sent directly into the GC/MS. This apparatus was able to quantify the ratio between the amounts of

20 chloramine, ammonia, and chlorine exiting the reactor by comparing the size of the peaks of elements with certain molecular weights. A sample of this data can be seen in Fig. 6.

25 It can be seen from Fig. 6 that the stream consists almost entirely of chloramine at a ratio greater than 10:1 with respect to ammonia. Ammonia is expected because there is a slight excess being fed into the reactor. It can also be seen that chlorine does not

30 appear because it is almost entirely consumed in the reaction.

It is to be understood that what has been described is merely illustrative of the principles of the invention and that numerous arrangements in accordance with this invention may be devised by one skilled in the art  
5 without departing from the spirit and scope thereof.

**WHAT IS CLAIMED IS:**

1. A process for the continuous production of inorganic gas, comprising the steps of:

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- (a) providing a reactor with a reaction zone;
- (b) feeding at least two feed gases concentrically to the reaction zone;
- (c) reacting the at least two feed gases in the reaction zone, thereby forming a product gas;
- 10 (d) providing at least one solids collection system; and
- (e) flowing the product gas to the at least one solids collection system,

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wherein at least one of said at least two feed gases is heated prior to being fed to the reaction zone.

2. The process of claim 1, wherein said reactor further comprises at least two reactor feed tubes for concentrically feeding the at least two feed gases to the reaction zone.

3. The process of claim 1, wherein at least one of said at least two feed gases is heated to a temperature greater than 350°C.

4. The process of claim 1, wherein said reactor is constructed from a material selected from the group consisting of hastelloy C, stainless steel, brass, borosilicate glass, silicate, sodium silicate, potassium silicate silica, and any combinations thereof.

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5. The process of claim 1, wherein said at least two feed gases are selected from the group consisting of acetylene, ammonia, boron trichloride, boron trifluoride, butadiene, butane, butene, carbon dioxide, carbon  
5 monoxide, chlorine, cis-2-butene, deuterium, dimethylamine, dimethyl ether, ethane, ethylene, ethyl chloride, hydrogen, hydrogen bromide, hydrogen chloride, hydrogen sulfide, isobutane, isobutylene, methane, methyl bromide, methyl chloride, methyl mercaptan, methylamine,  
10 nitric oxide, nitrogen dioxide, nitrous oxide, oxygen, perfluoropropane, propane, propylene, sulfur dioxide, sulfur hexafluoride, trans-2-butene, trimethylamine, and any combinations thereof.

15 6. The process of claim 1, further comprising feeding at least one inert gas concurrently with at least one of said at least two feed gases.

7. The process of claim 6, wherein said at least  
20 one inert gas is selected from the group consisting of nitrogen, argon, helium, neon, and any combinations thereof.

8. The process of claim 1, wherein said at least  
25 one solids collection system is selected from the group consisting of trap, filter, cyclone, and any combinations thereof.

9. The process of claim 8, wherein said filter is  
30 selected from the group consisting of cartridge, bag, granular bed, and any combinations thereof.

10. An apparatus for the continuous production of gaseous compounds, the apparatus comprising:

- 5 (a) at least one reactor with at least two reactor feed tubes for concentrically feeding feed gas to the reaction zone; and
- (b) at least one solids collection system.

10 11. The apparatus of claim 10, wherein said at least one reactor includes a reaction zone.

12. The process of claim 10, wherein said at least one reactor is constructed from a material selected from  
15 the group consisting of hastelloy C, stainless steel, brass, borosilicate glass, silicate, sodium silicate, potassium silicate silica, and any combinations thereof.

13. The apparatus of claim 10, wherein said at  
20 least one solids collection system is selected from the group consisting of trap, filter, cyclone, and any combinations thereof.

14. The process of claim 13, wherein said filter is  
25 selected from the group consisting of cartridge, bag, granular bed, and any combinations thereof.

15. A process for the continuous production of chloramine gas comprising the steps of:

- 30 (a) providing a reactor having a reaction zone, and having a first feed gas tube and a second feed

gas tube that concentrically feed gas to the reaction zone;

- (b) feeding a chlorine-containing gas to the first feed gas tube;
- 5 (c) feeding ammonia to the second feed gas tube;
- (d) reacting the chlorine-containing gas with the ammonia in the reaction zone, thereby forming chloramine gas;
- (e) providing at least one solids collection
- 10 system; and
- (f) flowing the chloramine gas to the at least one solids collection system,

wherein said chlorine-containing gas, said ammonia,  
15 or both said chlorine-containing gas and said ammonia are heated prior to step (d).

16. The process of claim 15, wherein said chlorine-containing gas, said ammonia, or both are heated to a  
20 temperature greater than 350°C.

17. The process of claim 15, wherein said chlorine-containing gas is fed to the reaction zone at a flow rate about 0.001 ft<sup>3</sup>/min to about 0.1 ft<sup>3</sup>/min.

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18. The process of claim 15, wherein said ammonia is fed to the reaction zone at a flow rate about 0.002 ft<sup>3</sup>/min to about 0.2 ft<sup>3</sup>/min.

30 19. The process of claim 15, further comprising feeding inert gas to said reaction zone cocurrently with said chlorine-containing gas.

20. The process of claim 20, wherein said inert gas is selected from the group consisting of nitrogen, argon, helium, neon, and any combinations thereof.

5        21. The process of claim 20, wherein said inert gas is fed at a flow rate about 0.1 ft<sup>3</sup>/min to about 1 ft<sup>3</sup>/min.

10        22. The process of claim 15, wherein the reactor is constructed from a material selected from the group consisting of hastelloy C, stainless steel, brass, borosilicate glass, silicate, sodium silicate, potassium silicate silica, and any combinations thereof.

15        23. The process of claim 15, wherein said at least one solids collection system is selected from the group consisting of trap, filter, cyclone, and any combinations thereof.

20        24. The process of claim 23, wherein said filter is selected from the group consisting of cartridge, bag, granular bed, and any combinations thereof.