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PRODUCTION OF HYDRAZINE FROM AMMONIA IN AN ATMOSPHERIC PRESSURE ELECTRON-BEAM CONTROLLED DISCHARGE


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DC, RF and microwave discharges in ammonia have been extensively studied for the synthesis of hydrazine [1, 2, 3, 4]. Chemical and electrical efficiencies approaching economic interest have been obtained in some cases. However, as these discharges are limited in operating pressure and in size, the achievement of the high flow rates required in industry appears to be very difficult.

The electron-beam controlled discharges which have been successfully used for pumping high power lasers [5] can be operated at atmospheric pressure with good uniformity over large volume thus permitting handling of large amounts of gas in fast flow systems. Furthermore the electron average energy can be adjusted (by varying the electric field) from very low values to a maximum of the order of 2 eV (limited by breakdown); this flexibility is of special interest for plasma chemistry. Very encouraging results have already been obtained for ozone production [6] with these discharges. Preliminary results for hydrazine are presented in this paper.

Experiment. The experimental arrangement is shown in Fig. 1. The electron gun has been described previously [7]. The discharge chamber is filled with pure ammonia after being evacuated to a pressure of 10^{-1} mbar. The electron beam (120 keV, 1.3 mA/cm²) enters the chamber through a 12 µm thick titanium foil of 15 x 5 cm². The electron-beam controlled discharge takes place between a grid which protects the electron window and the anode (aluminum alloy plate of 17 x 7 cm²). The cathode-anode spacing is 2 cm. A positive voltage is applied permanently to the anode. The discharge current flows when the gas is ionized by the electron beam (Fig. 2).

The other discharge parameters are:
- pulse duration : 100 µs
- repetition rate : 10 Hz
- discharge current : 2.3 A
- discharge voltage : 9 kV
- pressure : 100 mbar
- gas flow rate (standard conditions): 7.5 cm³ s^{-1}
- ratio electric field/gas density : 1.5x10^{-16} V cm²

The concentration ratio of hydrogen, nitrogen and ammonia are measured by gas chromatography downstream of the discharge and hydrazine is detected by a colorimetric method using p-dimethylamino-benzaldehyde [8].

Fig. 1 - Experimental arrangement.
1 - 120 keV electron-beam; 2 - Electron window (12 µm Ti foil); 3 - Electron gun vacuum chamber; 4 - discharge cathode (grid); 5 - Discharge zone; 6 - Gas flow (pure ammonia); 7 - Discharge chamber; 8 - Discharge anode; 9 - Insulated feed through; 10 - Gas flow (ammonia and products).

Fig. 2 - Time evolution of the electron-beam and discharge currents.
Upper trace : electron-beam current (arbitrary unit).
Lower trace : discharge current (A/cm)
Time scale : 20 µs/cm

Results. Approximately 1.1% of the ammonia flowing through the discharge chamber is decomposed and the hydrogen to nitrogen mole ratio is equal to 5. Assuming that hydrogen, nitrogen and hydrazine are the only products which can be formed in our experimental conditions, the hydrazine production can be calculated: the volume concentration is 0.37%, the efficiency is 7g/kW·hour and the
production rate is \(0.144\) g/hour.

After a two hours discharge run, the amount of hydrazine collected downstream the discharge by dissolving the gas output in water is one thousand times smaller than the yield calculated previously and the amount of hydrazine recovered by washing the chamber walls with water is only one hundred times smaller.

**Discussion.** The average residence time of the gas in the discharge chamber is 20 minutes and the surface of the aluminum alloy chamber walls is 3,000 cm\(^2\). The average deposit thickness of the hydrazine produced during a discharge run would be of the order of \(1\mu m\). Hydrazine condensation on the chamber walls and decomposition before it can be dissolved and analysed is the most probable explanation. Another possible explanation would be the formation of other nitrogen-rich compounds such as \(\text{HN}_3\) or \(\text{N}_2\text{H}_4\), but these compounds are so unstable that their presence in significant amounts seems unlikely.

The discharge chamber is now being modified in order to reduce the gas residence time and to avoid as much as possible its contact with metallic surfaces.

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**REFERENCES**