MAKING OXIDES OF NITROGEN IN A VORTEX GLOW DISCHARGE

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Parameter measurements are reported for a vortex glow discharge and the way they affect the synthesis of oxides of nitrogen from air.

The oxidation of atmospheric nitrogen in a nonequilibrium plasma is of a considerable interest because it may be possible to reduce substantially the energy consumption in making NO by comparison with equilibrium arc plasmas. In a nonequilibrium UHF discharge, the specific energy consumption in the synthesis of NO has been reduced to 12 kW·h/kg NO with a molar concentration of the product at the outlet of 4%, while a catalyst can improve on this by a factor 1.5 [1]. A nonequilibrium UHF plasma has been used with electron cyclotron resonance to attain 2.7 kW·h/kg NO, which is close to the theoretical limit [2].

This confirms that the energy can be minimized in nonequilibrium discharges in which plasmochemical reactions predominate, which are stimulated by vibrational excitation, in which respect they have a great advantage over equilibrium discharges [3, 4]. A low energy consumption EC has also been obtained in a plasma-beam discharge [5]. Unfortunately, experiments with nonequilibrium electrode discharge have not yet given such good results as have electrodeless ones. Nitrogen oxide synthesis in a high voltage discharge at atmospheric pressure has been used with a narrow water-cooled tube (0.003 m) with an energy consumption of 26 kW·h/kg NO [6]. A turbulent flow at atmospheric pressure has been used in a high-voltage plasmatron with a self-establishing long discharge [7], which gave a molar concentration of 1% with EC of 15.5 kW·h/kg NO, which is less than the consumption in thermal arc plasmotrons [8].

Here we examine using a vortex glow discharge, which has an advantage over other types of electrode discharge [9-12]. The NO was made in a vortex tube 0.03 m in diameter and with a length from 0.06 to 0.5 m, with a glow discharge struck at the axis (Fig. 1). The design has been described in [13]. The gas pressure at the axis was varied over the range 8-32 kPa. We used air flowing at from 2×10^{-5} to 2.3×10^{-3} kg/sec. In the vortex chamber, the gas was spun up to a speed higher by an order of magnitude than the translational speed along the tube, which provided for stable discharge. The current was 0.1-1 A, with the diffuse mode of burning retained. At high currents, the discharge contracted, but the field strength was not substantially reduced.

The local gas temperature was measured with thermocouples, which were inserted radially at three points in the vortex tube (Fig. 1). The temperatures were corrected for the radiation [14]. In the same sections, ceramic tubes were used to sample the gas, which passed to a flow cell in an IKS-31 infrared spectrometer (Fig. 1) to determine the concentrations of the oxides of nitrogen (see Table 1, data from [15, 16]). The proportions of NO_2 and N_2O_4 were determined from the $2NO_2 = N_2O_4$ equilibrium, which enabled us to reduce the number of measurements while giving satisfactory accuracy in determining the amount of combined nitrogen, the more so in that in our case at moderate pressures in the discharge and in the measurement system, the proportions of N_2O_4 and NO_2 did not exceed 10% of the total oxides of nitrogen. The results were spot checked by measuring the concentrations of the oxides of nitrogen by the peroxide method [17].

The axial temperature was determined from the relative intensities of the rotational lines in the (0, 0) and (0, 2) band in the second positive system of nitrogen [14, 18, 19] by the use of an SDMS spectrometer (Fig. 1).

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TABLE 1

Molecule	Band center, cm^{-1}	α, $cm^{-2} \cdot atm$	ν_1, cm^{-1}	ν_2, cm^{-1}
NO	1876	110	1700	2000
NO_2	1617	2060	1520	1680
NO_2	2908	58	2860	2940

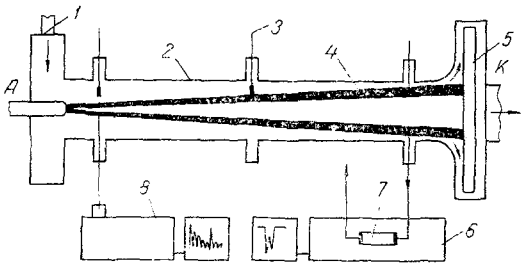

Fig. 1. The apparatus: 1) vortex chamber with anode; 2) pyrex or silica tube; 3) thermocouple; 4) reaction zone; 5) diffuser with cathode; 6) IKS-31 infrared spectrometer; 7) measurement cell; 8) SDMS spectrometer.

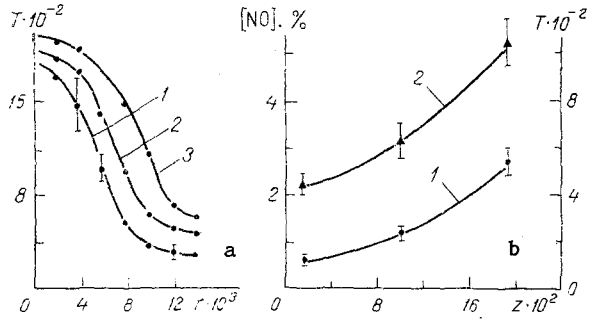

Fig. 2. Radial temperature profiles at various distances from anode (a): 1) 0.01; 2) 0.1; 3) 0.19 m; b) NO molar concentration profile (1) and mean mass translational temperature T in flow direction (z axis) (2). T in K and r and z in m.

Results and Discussion. The radial translational temperature distributions measured in three sections are shown in Fig. 2a. These gave the mean mass temperature, which can be related to the nitrogen oxide concentrations there (Fig. 2b). The downstream increase in that temperature is accompanied by an increase in NO concentration. The T distribution is highly inhomogeneous along the radius and along the axis. The NO synthesis zone, which has an optimum temperature range [3], expands downstream (Fig. 1).

We related the NO production to the pressure P and the bulk energy deposition W in $J \cdot cm^{-3} \cdot atm^{-1}$. We varied the pressure with a fixed flow and found that the maximum NO concentration occurred at 15.8 kPa (Fig. 3). A similar result has been reported previously [20]. It has been found [21] that for $P > P_0$, the efficiency in a nonequilibrium discharge falls, probably because of increased V-T relaxation and reduced disequilibrium. For $P < P_0$, the reduction in the volume yield is ascribed to reduction in the electron concentration,

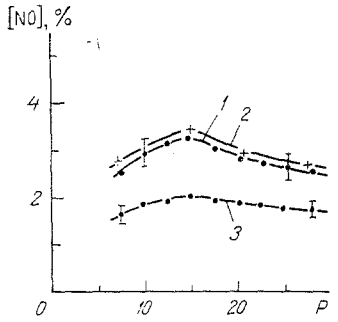

Fig. 3

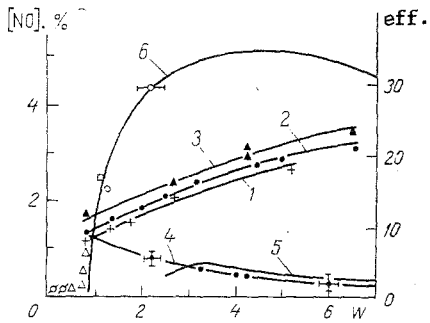

Fig. 4

Fig. 3. Effects of pressure and discharge current and length on NO yield (Q = 0.24 g/sec): 1) I = 0.2 A, z = 0.26 m; 2) 1.35 and 0.26; 3) 0.2 and 0.13, P in kPa.

Fig. 4. Dependence of yield of nitrogen oxides (curves 1-3) and reactor efficiency in % (curves 4-6) on specific energy consumption: 1) z = 0.06 m; 2) 0.13; 3 and 4) 0.26; 5) [5] data; 6) [2], W in $J \cdot cm^{-3} \cdot atm^{-1}$.

reduction in the translational temperature, and reduced oxygen dissociation [20]. Also, the reduced reactant concentrations lower the yield in the bimolecular chain reaction.

It has been found [22] that the residence time in the discharge affects the yield, since vibrational excitation and V-T relaxation involve the NO and atomic oxygen and are spatially separated. We measured the W dependence of the molar NO concentration for discharges with lengths z = 0.06; 0.13; 0.26 m (Fig. 4). An increase in length and correspondingly in residence time under otherwise equal conditions increases the NO concentration.

As the specific energy deposition increases, however, the efficiency H/EC decreases, in which H = 0.83 kW·h/kg NO is the minimum synthesis energy. In experiments with a nonequilibrium discharge at atmospheric pressure [6], the W dependence was similar to that found here for $W > 3$ $J \cdot cm^{-3} \cdot atm^{-1}$ (Fig. 4). In a review [2], an entirely different W dependence was found from theory and experiment on the efficiency, which was so for lower pressures, which may account for the experimental discrepancies.

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EFFECTS OF PLASMA FLOW STRUCTURE ON HEAT TRANSFER WITH POWDER PARTICLES

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Measurements have been made on the heat transfer to granular material in a plasma reactor having a multijet mixing chamber. The temperature distribution over the cross section has been measured by a spectral method and by calorimetry. The granular-material flow rate affects the heat flux to the walls. The measurements are fitted to an equation in dimensionless parameters.

The performance in processing a granular material in a plasma device is dependent on the thermophysical parameters, the relation between the mass flow rates, and the organization of the mixing, being ultimately governed by the transfer; the data on this are scanty and conflicting.

To accelerate the transfer, various reactor designs have been tested, one of the most promising being a multijet mixing chamber (plasma module). Even in the simplest style, a three-jet chamber can produce a plasma flow with fairly uniform temperature and velocity profiles. One can also use any method of injecting the granular material and can raise the reactor power by increasing the total number of plasmotrons or the unit power of each and by combining the multijet chamber with one or more modular reactors. This improves the performance, increases the total power, and extends the applications.

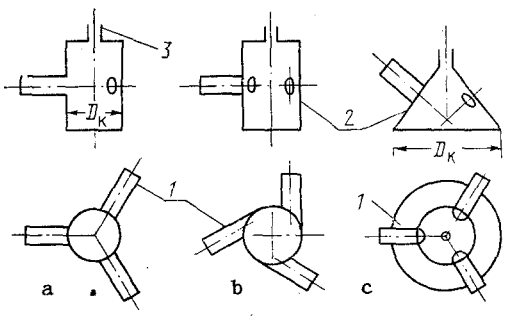

Fig. 1. Mixing chambers: a) cylindrical; b) tangential; c) canonical: 1) plasmotron; 2) mixing chamber; 3) inlet.

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