Conversion of NO to N_2 in Continuous Microwave Discharge

Junwang Tang, Tao Zhang,* Dongbai Liang, Xiaoying Sun, and Liwu Lin
State Key Laboratory of Catalysis, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, PO Box 110, Dalian 116023, P. R. China

(Received May 1, 2000; CL-000419)

The direct conversion of NO to N_2 by a continuous microwave stable discharge at atmospheric pressure is reported, and the conversion of NO to N_2 is up to 60% or more in the presence of excess O_2 or O_2 and H_2O. The effect of the power of microwave on the conversion of NO is observed, and novel results are obtained.

NO_x pollution has become one of the most serious environmental problems, and many research groups are attracted to this topic. Although considerable progress has been achieved in the reduction of NO_x emissions, the release of NO_x into the atmosphere from the combustion of fossil fuels in both stationary and mobile sources continues to be a major environmental problem. Among the many abatement processes developed, one of them is the study on the removal of NO_x from combustion gases using a pulsed streamer corona discharge (PSCD), which utilizes a high-voltage electrical discharge.[1–4] Using the PSCD method, in the presence of excess O_2, NO was mainly converted to NO_2,[2,5] or NO_3,[4,6] then the NO_x was subsequently absorbed by NH_3 or other additives.[2] The problems existing in this process are high energy consumption and secondary pollution. In order to adapt to the higher requirement of environmental protection in the future, new methods of reducing NO have been explored. In the paper, it probably is the first time to report the new method, which is the direct conversion of NO to N_2 by a continuous microwave stable discharge (CMD) at atmospheric pressure without the need of any additives.

The microwave reaction system consisted of a microwave generator, a rectangular waveguide, a circulator, a resonant cavity and a plunger. The microwave energy was supplied by a 200 W, 2.45 GHz microwave generator and the effective microwave power for the experiments ranged from 5 to 60 W. For the experiments, a special reactor was used that enabled the CMD to be carried out at atmospheric pressure. A quartz tube of 8mm i. d. was aligned vertically at the center of the single mode resonant cavity, so that the discharge region seated in the microwave field of maximum intensity. In this paper, the residence time of the flowing gas in the CMD region was about 1 second, which is much shorter than that reported by other papers,[1] The concentration of NO was also much higher, up to 2000 ppm. The gas composition (NO, NO_2, N_2 and so on) was determined by an on-line NO_x-analyser (FGA 4000/4005) and Gas Chromatograph (GC-8800, 13X and PQ columns).

Table 1 shows the conversion of NO under different conditions, in which the gas flow-rate is invariable, namely, 60 mL/min. In group 1, where there were only NO (2000 ppm) and the balance gas (He), the conversion of NO to N_2 was up to 88.03%, and the total conversion of NO was 96.1%. In group 2, namely the dry gas, which consisted of O_2 (2%), NO (2000 ppm) and He, the conversion of NO to N_2 decreased, while that to NO_2 increased. The total conversion of NO also decreased, which can be attributed to the effect of excess O_2. In group 3, namely the wet gas, which consisted of NO (2000 ppm), O_2 (2%), H_2O (5%), and He, compared with group 2, the conversion of NO to N_2 decreased a little, but that to NO_2 increased remarkably. About 60% of NO was converted to N_2, and the total conversion of NO was higher than 80%. This suggested that the addition of H_2O promoted the conversion of NO to NO_2, but did not lower the production of N_2 in the CMD.

Table 1. Conversion of NO in continuous microwave discharge

Condition	Power of CMD/W	Conversion of NO/%	Conversion of NO to N_2/%	Conversion of NO to NO_2/%
Group 1[a]	30—40	95.72	86.2	9.52
	35—45	96.10	88.03	8.07
Group 2[b]	30—40	77.23	59.61	17.62
	35—45	77.54	62.29	15.25
Group 3[c]	30—40	82.04	54.28	27.76
	34—45	84.41	60.30	24.11

[a] 2000 ppm NO. [b] 2000 ppm NO, 2% O_2. [c] 2000 ppm NO, 2% O_2, 5% H_2O. Balance gas He. Total flow-rate, 60 ml/min.

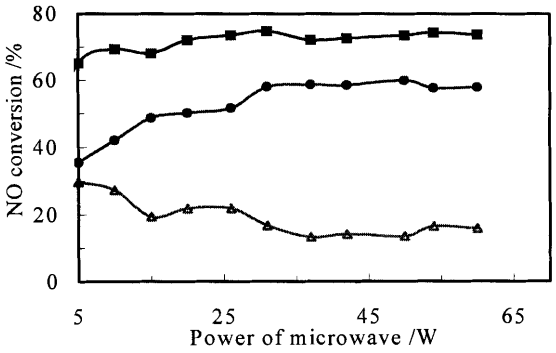

Figure 1. The effect of microwave power on NO conversion in continuous microwave discharge, NO total conversion (squares), NO conversion to N_2 (circles) and to NO_2 (triangles).

In order to study the effect of the power of the CMD on the conversion of NO, another experiment was made under the condition of group 2 at atmospheric pressure, and the results are shown in Figure 1. The total conversion of NO increased slightly with the increasing of the power, while the conversion

of NO to N_2 increased greatly, and that to NO_2 decreased remarkably. When the power was increased further, all conversions were leveled to the constant values. This suggests that increasing the power of the CMD is beneficial for the conversion of NO to N_2.

Although the reaction mechanism in the CMD is still not clear, it is speculated as follows. The difference of energy between the electrons and the heavy ions is large, and the average temperature of the reacting gas is comparatively low in the CMD.[7] At the same time, the life time of the electrons are long in the CMD.[7] By these electrons with high energy and long life time, it is prompted the decomposing of NO to a number of active N radicals. The formation of the N radicals can induce, and sometimes accelerate the reaction between N and N, or N and NO, which is easier in lower average temperature of the reacting gas,[1,3,8] so the conversion of NO to N_2 can be very high in the CMD, especially when there is no O_2 present. It is interesting to note that the decomposition of NO to N_2 is not weakened with the presence of H_2O. On the other hand, NO_2 formation is strengthened by the existence of H_2O.

So far as we know, this work represents the first observation of the conversion of NO in the CMD at atmospheric pressure. The present finding also reveals the difference between the CMD and other discharge methods.[4–6]

The authors thank Prof. Can Li, director of State Key Laboratory of Catalysis, Dalian Institute of Chemical Physics, for helpful discussions.

References

1 G. Sathiamoorthy, S. Kalyana, W. C. Finney, and B. R. Locke, *Ind. Eng. Chem. Res.*, **38**, 1844 (1999).
2 Y. S. Mok and I. S. Nam, *J. Chem. Eng. Jpn.*, **31**, 391 (1998).
3 A. Harano, Y. Imaizumi, and M. Sadakata, *J. Chem. Eng. Jpn.*, **31**, 694 (1998).
4 K. Takaki, M. A. Jani, and T. Fujiwara, *IEEE Trans. Plasma Sci.*, **27**, 1137 (1999).
5 H. Miessner, R. Rudolph, and K. P. Francke, *Chem. Commun.*, **1998**, 2725.
6 K. Urashima, J. S. Chang, J. Y. Park, D. C. Lee, A. Chakrabarti, and T. Ito, *IEEE Trans. Ind. Appl.*, **34**, 934 (1998).
7 H. Zhao, in "Plasma Chemistry and Processing," ed. by H. Zhao, USTC (University of Science and Technology of China) Press, Hefei (1993), pp. 181–186.
8 H. Namba, Y. Aoki, O. Tokunaga, R. Suzuki, and S. Aoki, *Chem. Lett.*, **1988**,1465.