Photochemical Removal of N₂O Diluted in N₂, N₂/O₂, or N₂/O₂/NO mixture by 193 nm Excimer Laser at Atmospheric Pressure

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Nitrous oxide (N₂O) in the Earth’s atmosphere, 47.5% of which arises from combustion processes in 1999, is a major contributor to the greenhouse effect due to its long residence time of about 150 years and its relatively large energy absorption capacity per molecule. Removal of N₂O has recently been studied using catalysts and also by catalyst free pulsed corona discharge and microwave discharge methods. In our previous study using microwave discharge, N₂O could be efficiently decomposed into N₂ and O₂ in N₂ at atmospheric pressure. However, when N₂O diluted in an N₂/O₂ mixture was decomposed by microwave discharge, a large amount of NO was emitted due to discharge reactions of N₂ and O₂. This NO emission is a general disadvantage in the application of discharge technique to N₂O removal in air. Thus, a higher selectivity is required in order to remove N₂O in the N₂O/N₂/O₂ mixtures.

We have recently proposed a photochemical process as an ideal method for a high selective removal of N₂O [1]. It should be noted that the absorption cross sections of N₂O (8.95 × 10⁻²⁰ cm² molecule⁻¹) is about 300 times larger than that of O₂ (3.2 × 10⁻²² cm² molecule⁻¹) at 193 nm. N₂O is selectively decomposed into N₂ + O(¹D) at 193 nm without NO emission. There is no absorption for N₂ at 193 nm. On the basis of above findings, an efficient and selective removal of N₂O is expected in air by using 193 nm ArF excimer laser.

In this work, we have studied photochemical N₂O removal in N₂, air, and N₂/NO mixture at atmospheric pressure by using 193 nm ArF excimer laser at a room temperature without using any catalysts. The N₂O conversion and formation ratios of N₂, O₂, and NO were measured as a function of laser irradiation time, laser power, or partial pressure of N₂O. The maximum conversion of N₂O in 1:20 mixture of N₂O/N₂ and 1:15:5 mixture of N₂O/N₂/O₂ was 98% and 95%, and the formation ratios of N₂O:NO in N₂O/N₂ and N₂O/N₂/O₂ mixtures at an irradiation time of 30 min were 94:46:6.7% and 93:46:3.8%, respectively, at a laser power of 100 mJ, a repetition frequency of 5 Hz, and an irradiation time of 30 min. The conversion of N₂O in N₂O/N₂/NO mixtures decreased with increasing the NO partial pressure and a small amount of NO₂ was observed as a product at high NO pressures. A reasonable agreement was found between experimental data and calculated ones using known photochemical and gas kinetic data, suggesting the validity of model calculation.

In summary, photochemical N₂O removal was studied in N₂O/N₂, N₂O/N₂/O₂, and N₂O/N₂/NO mixtures at atmospheric pressure and room temperature using an ArF excimer laser. The conversion of N₂O in N₂ and air was >95% at a laser energy of 100 mJ. We found that N₂O is efficiently decomposed by one photon decomposition at 193 nm at relatively high N₂O concentrations above 0.48-5.1% (v/v).

References