PLASMA ASSISTED N_2O OXIDATION AND REDUCTION

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Extended abstract

The reduction of atmospheric emission of N_2O is an important environmental problem because of N_2O input in the greenhouse effect and an eventual stratospheric ozone depletion problem. Among the identified anthropogenic sources, adipic acid production by nitric oxidation of cyclohexanol and/or cyclohexanon is one of the most important. Since N_2O concentration is relatively high in the off-gas from adipic acid production, the most effective way to reduce N_2O emission could be N_2O conversion back to NO_x, so nitrogen oxides in this case can be recycled in the process as nitric acid. This way can be more attractive compared to commercially demonstrated processes for N_2O destruction in the boilers or catalytic dissociation to N_2 and O_2. These studies were done under Rhone-Poulenc's initiatives and concern :

- experimental investigation of plasma assisted N_2O treatment in so-called GlidArc reactor,
- theoretical analysis of the physical and chemical mechanisms of this process under thermal non-equilibrium conditions,
- analysis of the opportunities to enhance the process efficiency.

Experiments were carried out in the GlidArc [1] reactor producing non-thermal plasma at atmospheric pressure. The bench-scale reactor consisted of a 2.5 l cylinder steel in which gliding electrical discharges were run between three pairs of knife-shaped steel electrodes placed around a gas inlet nozzle. The electrodes were supplied from an AC 3-phase 50 Hz power supply. Dry or wet $N_2O/N_2/O_2/NO_2$ mixtures with the N_2O contents in the range 31.6-50.6 mol. % and flow rates from 22 to 81.8 slm were treated. It was shown that N_2O can be effectively oxidized to NO_x in the GlidArc discharge. The efficiency of the

oxidation is close to 70 %. High value of the NO_x yield with respect to conventional processes for conversion of N_2O to recyclable NO_x (possible NO_x yield here as high as 33 moles per 100 moles of NO_x) makes the GlidArc results very attractive and promising.

The physico-chemical model of N_2O oxidation and reduction under plasma conditions has been proposed. This model permits to describe processes in both cold and in thermal plasmas. The experimental results were compared to the theoretical calculation of N_2O treatment for both thermal and non-equilibrium plasmas. Analysis has shown that N_2O oxidation efficiency in experimental conditions is significantly higher then in the case of cold non-equilibrium plasmas, and it exceeds the upper limit for thermal (both conventional and plasma) systems, too. The modeling has also shown that this result is related to a transient regime of the non-equilibrium plasma generation in the GlidArc discharge in which it is possible to combine the advantages of both thermal and non-equilibrium systems. The sensitivity analysis permits to determine chemical mechanisms of the process.

The effect of an additional increase (in twice) of the N_2O-to-NO_x conversion efficiency in a transient regime of plasma generation was found, too. This effect is related to an acceleration of O_2 dissociation in non equilibrium plasmas and a reaction of the vibrationally excited NO with oxygen molecule.

1. A. Czernichowski, Gliding arc. Applications to engineering and environment control, Pure and Applied Chemistry, vol. 66(6), 1301-1310 (1994).