Spectroscopic Temperature Measurement in Dielectric Barrier Discharges

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Abstract - We have performed spectroscopic temperature measurements in dielectric barrier discharges in nitrogen at atmospheric pressure. The rotational temperature T_{rot} was determined from the transition $C\,^3\Pi_u \rightarrow B\,^3\Pi_g$ of N_2. For very low frequency excitation (50 Hz to 4 kHz) no significant rise of T_{rot} above the gas inlet temperature was observed. For excitation in the range 30 to 60 kHz a pronounced increase of T_{rot} was found. It tends to scale with the residence time in the reactor. Thus in barrier-discharge modeling at higher frequencies a possible heavy-particle temperature rise must be regarded.

Introduction

Dielectric barrier discharges are now widely used to promote non-equilibrium plasma-chemical reactions at atmospheric pressure e.g. for exhaust gas cleaning [1]. They are highly effective in creating electrons with mean energies up to several eV which then produce ions and radicals. In the modeling of such processes it is generally assumed that the translational temperature of the heavy particles remains closely to the gas inlet temperature. However, the energy of the electrons is not fully coupled into dissociation and ionization of molecules but also into their electronic, vibrational and rotational excitation. A part of this excitation energy leads to a heating of the heavy particles via radiationless dexcitation processes. Another direct heating mechanism of heavy particles is heating of ions. The gas temperature influences both the efficiency of discharge excitation and also the heavy particle chemical reactions in the gas phase [2]. In order to investigate the influence of the discharge parameters on heavy particle temperature we have measured spectroscopically the rotational temperature in the second positive emission band system of nitrogen. We first give a short description of the experimental set-up and the evaluation procedure and then discuss the results.

Experiment and evaluation procedure

The measurements were carried out by observing end-on the light emitted from a barrier discharge reactor. The outer electrode and dielectric of the reactor were

formed by an alumina tube of 47 mm inner diameter which was metal-coated on its outside on a length of 200 mm. The inner electrode consisted of a concentric steel cylinder. Synthetic air at atmospheric pressure was fed axially through the gap between tube and cylinder with a controlled standard flow rate. Gap widths of 0.75 and 1.5 mm were used. In order to guarantee well defined temperature conditions in the reactor, the gas was heated in a line section before entering the reactor. The reactor was heated to the same temperature by heater windings.
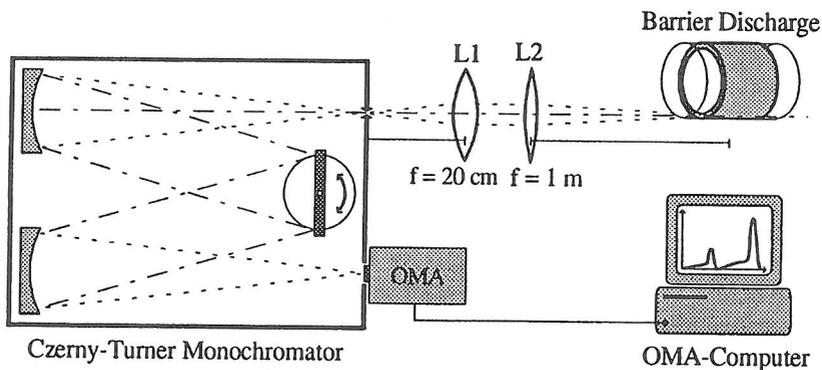

Fig. 1: Schematic representation of the optical set-up used in the experiments.

The barrier discharges were excited by sinusoidal or pulsed high voltage signals applied to the electrodes by either a high voltage transformer at mains frequency (50 Hz), a pulse generator delivering bipolar high-voltage pulses with rise times of 5 µs and repetition rates between 0.5 and 5 kc/s, or a power amplifier with an impedance matching network at frequencies between 30 and 100 kHz. The discharge power was measured with the help of the charge-voltage method a detailed description of which is given in [3].

The light emitted from the discharge was focused onto the entrance slit of a Czerny-Turner spectrograph of 1m focal length. The light was detected by a diode array camera and stored by an optical multichannel analyzer. A schematic view of the optical arrangement is shown in Fig. 1. The focus of the imaging system was located in the axial center of the discharge gap. There will, however, be also some light from different axial positions reaching the entrance slit. Therefore the measured temperature will not represent exactly the value prevailing at the axial center of the reactor but will be some intermediate value between entrance and exit temperature.

The temperature was determined from unresolved rotational spectra of the $v'=0 \rightarrow v''=2$ line of the $C\,^3\Pi_u \rightarrow B\,^3\Pi_g$ second positive system of N_2. For this purpose the spectra were measured with a reduced resolution of 0.185 nm and the intensities at

certain distances from the band-head were compared to those of calculated spectra for the same resolution [4]. In Fig. 2 examples of calculated spectra for two different resolutions are given. The determination of the rotational temperature T_{rot} by this method was accurate to about ± 30 K. The individual rotational lines in the spectra were not resolved.
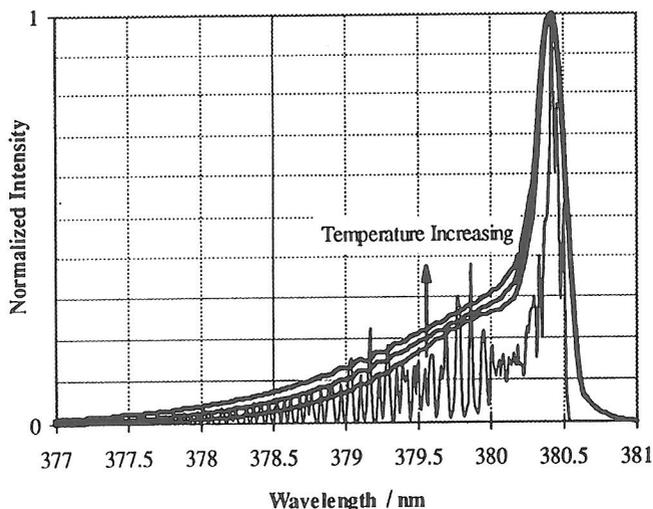

Fig. 2: Calculated rotational spectrum of the transition v'=0 → v''=2 of the second positive system of N_2.(C $^3\Pi_u$ → B $^3\Pi_g$) Thin curve: T = 373 K, convoluted with Gaussian of FWHM 0.02 nm; Bold curves: T = 273, 373, 473 K, convoluted with apparatus profile,FWHM 0.19 nm.

Experimental results and discussion

We assume that the rotational temperature represents to good accuracy the translational temperature of the measured molecules [5, 6]. We further assume that the distribution over the individual rotational states is not changed during the fast electronic excitation. The energy input per unit volume of gas passing through the reactor should vary as the number of discharge filaments crossing through this volume during its passage, i.e. proportional to A L f / S, where A is the reactor cross section and L its length, which was fixed in the experiments. S and f are the gas flow and the excitation frequency.

In Fig. 3 the measured rotational temperatures are plotted against the respective gas inlet temperatures for two different gap widths. For the larger gap width the flow rate and power were both increased in order to keep the residence time and the energy volume density constant. The rotational temperature follow the gas inlet temperature within the described accuracy range.
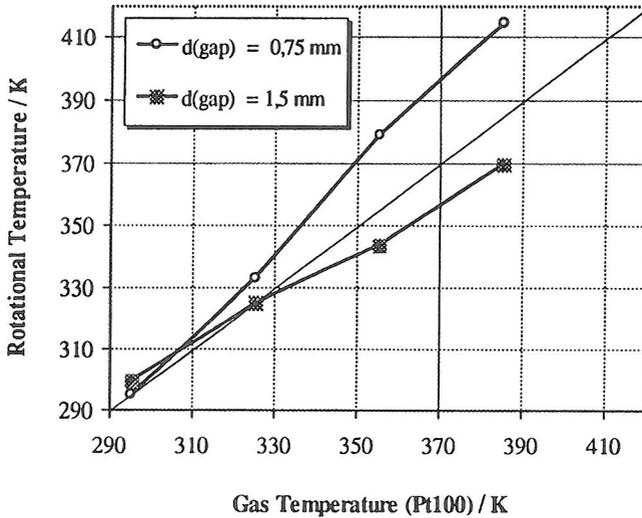
Fig. 3: Variation of T_{rot} with gas inlet temperature at 50 Hz excitation and constant residence time. Parameter: 10 l./min, 2 W at 0.75 mm; 20 l./min, 4 W at 1.5 mm.
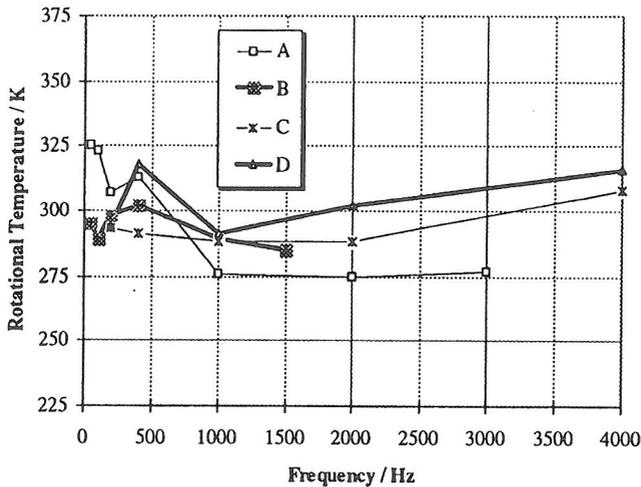

Fig. 4: Dependence of T_{rot} on excitation frequency in synthetic air with an inlet temperature of 295 K. A: 0.8 W/(l./min), power/frequency constant, $d_{gap} = 0.75$ mm; B: as A, but $d_{gap} = 1.5$ mm; C: 8 W, 1.25 l./min, $d_{gap} = 0.75$ mm l./min $d_{gap} = 1.5$ mm

In Fig. 4 the rotational temperatures are plotted over a wide frequency range and for higher power levels than in Fig. 3. For frequencies up to 400 Hz the discharge was excited by sinusoidal voltage signals. For frequencies above 1 kHz pulsed voltage was applied to the discharge. Within the measuring accuracy the observed values for T_{rot} are equal to the inlet temperatures. From this observation we conclude that the energy input and appearance of the discharges is relatively homogeneous. In this range of discharge parameters which may be used for exhaust gas remediation the rise of the gas temperature induced by the discharge action can be neglected.
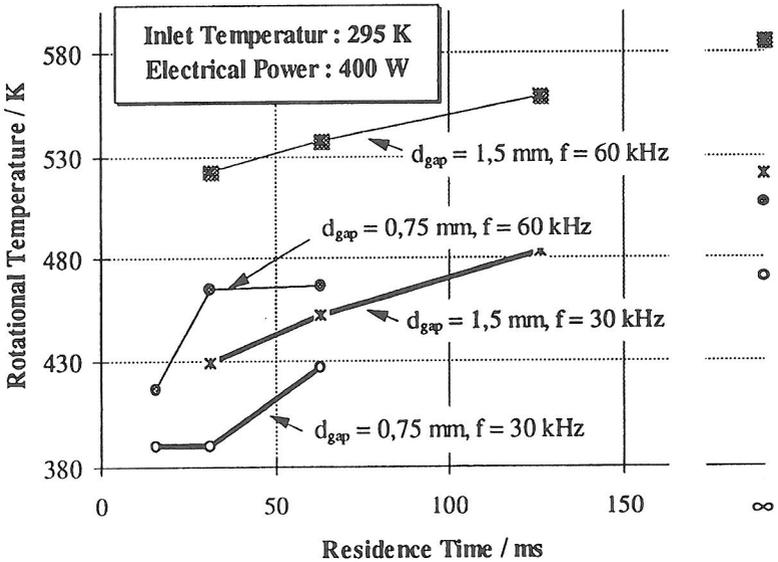

Fig. 5: Measured rotational temperatures as a function of the residence time for synthetic air and an inlet temperature of 295 K.

If higher frequencies and the associated higher plasma power levels are employed the gas temperatures rise markedly above the inlet temperatures as would be expected. In the cases of 30 and 60 kHz a drastic rise of T_{rot} up to 520 and 580 K respectively is observed for zero flow. These temperatures fall with increasing gas flow or - equivalently - reduced residence time. In **Fig. 5** these results for T_{rot} are displayed as a function of the residence time for constant electrical parameters. A variation of the discharge gap length from 0.75 to 1.5 mm doubles the cross section A. Consequently very similar temperature rises are observed for 1.5 mm / 30 kHz and 0.75 mm / 60 kHz. The results thus confirm the above proportionality.

Conclusions

We have shown by our measurements that the dielectric barrier discharge will heat the heavy particles translationally if moderate to high power levels are coupled into the discharge. It is therefore not realistic to keep the heavy particle temperatures at close to room temperature in modeling calculations. For low power levels in the very low frequency regime the temperature rise may be neglected.

Acknowledgments

References

[1] B.M. Penetrante, S.E. Schultheis (eds.), "Non-Thermal Plasma Techniques for Pollution Control", NATO-ASI Series G, Vol. 34 Part B (Springer, Berlin, 1993).

[2] M. Klein, G. Lins, M. Römheld and R.J. Seeböck, this conference; M. Klein, „Barrierenentladung zur Entstickung motorischer Abgase", Thesis, University of Karlsruhe, Karlsruhe, FRG, May 1995 (unpublished).

[3] U. Kogelschatz in "Process Technologies for Water Treatment", ed. by S. Stucki, (Plenum, New York, 1988), pp. 87-120.

[4] K. Behringer, private communication.

[5] K. Behringer, Plasma Phys. Contr. Fusion 33, 997 (1991).

[6] A. Chelouah, E. Marode, G. Hartmann and S. Achat, J. Phys. D: Appl. Phys. 27, 940 (1994).