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Abstract

Results of measurements of the ratio of transverse (D_T/μ) and longitudinal (D_L/μ) diffusion coefficients to mobility and drift velocity (W) as function of reduced electrical field (E/N) for electrons in nitrous oxide are presented. The coefficients D_T/μ and D_L/μ have been determined by applying the Townsend-Huxley method. The drift velocities were obtained by using the Bradbury-Nielsen technique. A set of total and partial cross sections has been used to calculate numerically the D_T/μ and W .

Keywords: electron transport coefficients, nitrous oxide

1. Introduction

Electronegative gases such as nitrous oxide have numerous applications. N₂O are important for chemistry, medicine and technology. Electron interaction with N₂O is important for atmospheric chemistry, being a greenhouse gas with approximately 200 years of permanence in stratosphere [1].

Swarm measurements in N₂O were only sporadic and solely at low reduced fields. The drift velocity and ratio of diffusion to mobility in N₂O was measured in the early part of the last century [2]. Other swarm studies was concentrated on measurements of the electron attachment coefficient to concentration (η/N) [3] or electron growth constant to concentration (λ/N) [4]. In this work, measurements of drift velocity and the ratio of transverse and longitudinal diffusion coefficients to the mobility (D/μ and D_L/μ) for electrons in N₂O are extended to intermediate reduced electrical field values - the range E/N from 15 Td to 200 Td. In this energy range numerous inelastic processes are possible and the analysis of swarm data is somewhat more complex task compared to ultralow energies. Present modelling shows how the diffusion coefficients in this E/N range are sensitive to the choice of inelastic cross sections.

2. Cross sections

We note that between existing data of electron scattering cross sections (CS) in N₂O are some discrepancies. Early measurements of total CS by Zecca et al. [5] are in agreement with measurements of CS for elastic scattering and vibrational excitation by Azria et al. [6]. These data agree also with semiempirical momentum transfer CS used for swarm-data modelling by Hayashi (private information). From other side, measurements of total CS by Brüche [7] agree with those by Szymtkowski et al. [8] and Kwan et al. [9]. The two data sets [5,8] differ by almost a factor of 2 at the resonant maximum of total (momentum transfer) CS at about 2.5 eV. However, the most recent elastic and vibrational CS [10] would indicate that the total CS of Szymtkowski et al. [8] and Kwan et al. [9] are overestimated by 20%. All data indicate rise of the CS below 1 eV due to the rotational excitation enhanced by the permanent dipole moment (0.28D) of N₂O molecule.

Comparison of CS in N₂O and CO₂ may be interesting (see fig. 1). Both gases have a linear configuration but N₂O is asymmetric – for this reason N₂O has a slight dipole moment. Total cross section for CO₂ [11] exhibits a resonant structure around 3.8 eV and another, wide maximum at 25 eV; for N₂O TCS is similar: two resonance for about 2.3

eV and 20 eV [12], see fig. 1. Note however, that in spite of the non-polar character of CO₂, its cross sections also rises in the low-energy limit.

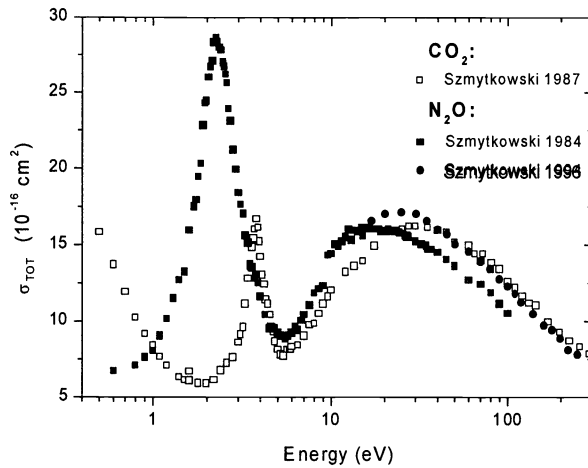


Fig. 1. Total cross sections for electron scattering in CO₂ and N₂O.

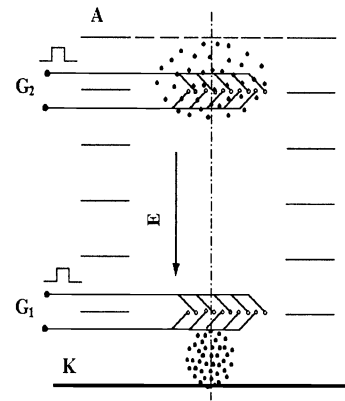


Fig.2. Schematic drawing of swarm experiments.

3. Experimental

Idea of swarm experiments is shown in fig. 2. Electrons emitted from by the UV radiation, pass through a source hole, drift under action of a uniform electric field and diffuse laterally to produce a widening radial spread of the electron swarm to be collected by the anode, consisting of five concentric rings. Grids G₁ and G₂ are used to pulse the swarm. The experimental Townsend-Huxley technique and the corresponding numerical procedure described previously [13] have been applied to determine both the ratio of transverse diffusion coefficient to mobility (D_T/μ) and the ratio of longitudinal diffusion coefficient to mobility (D_L/μ). Fractions of the total current falling on a selected part of the anode is a function of D_T/μ and D_L/μ . Formulae presented in detail in ref.[13] were used to derive the diffusion coefficients from measured currents. The length of the drift space is 9.88 cm and gas pressure ranged from 50-420 Pa.

The double grid Bradbury-Nielsen system was used for the drift velocity W measurements [13]. Some dependence of w on the gas density was observed; this is due to influence of diffusion effects. Values given are extrapolated to the infinite density limit. Overall uncertainty was 1.5% at the lowest E/N and 3% at the highest E/N .

3. Results and discussion

3.1. D_T/μ and D_L/μ

Present results for D_T/μ are shown in fig. 2. Measurements of D_T/μ agree well with those of Bailey and Rudd [2] up to $50 \cdot 10^{-21} \text{ Vm}^2$. At $100 \cdot 10^{-21} \text{ Vm}^2$ the results of Bailey and Rudd [2] are higher than present, we hypothesise gas impurity problems in the experiment of Bailey and Rudd. Present D_T/μ values agree reasonably well with semiempirical values of Hayashi [14] denoted on fig. 1. by "Hayashi II".

According to our knowledge, measurements of longitudinal diffusion coefficient to mobility in nitrous oxide are made the first time. Results for longitudinal drift coefficient to mobility in comparison to values in CO₂ [15] are shown on fig. 3. In this case D_L/μ for CO₂ is lower by 10-30% for all values of E/N . We note that in the contrast for CO₂ D_L/μ for N₂O increases slower in 40-70 Td range than for other values. Because of lack of other data for D_L/μ in N₂O we compare present data with those from the same laboratory for carbon dioxide.

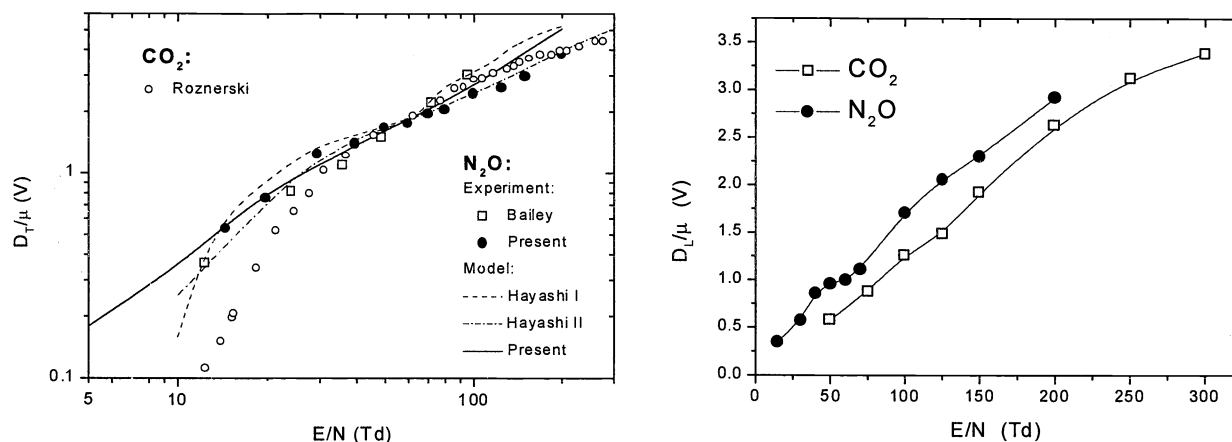


Fig. 2. D_T/μ and D_L/μ values for N_2O compare to other results [2,14] and for CO_2 data [15].

3.2. Model of cross sections

In this paper we tested recommended cross sections [16] based on experimental data described in the introduction, see fig. 3. Because experimental values of electronic excitation CS was measured sporadically and these data are rather incomplete we adopted semiempirical values of Hayashi. To calculation of D_T/μ from CS we used program BOLSIG. This program was designed to generate electron and transport data in pure gases or gas mixtures over a wide range of values of E/N by the numerical solution of the Boltzmann equation. The solution technique used in BOLSIG is based in part on the technique described in Pitchford et al. [17].

Comparison between results of semiempirical calculations and experimental swarm data are shown on fig. 2. Our semiempirical data obtained from recommended cross sections are in good agreement with experimental up to about 100 Td range of reduced electric field and only for 30 Td there can be noted a difference. For higher E/N agreement is quite reasonable - as the present model still does not contain higher electronic excitation states, for which we lack experimental data (see ref. [18]). Improvement of the model is under way.

We have also done calculations using "shareware" cross sections of Hayashi ("Hayashi I" in fig 2). Data deduced from semiempirical CS of Hayashi ("Hayashi I") and showed on fig.2 are in good agreement with experimental of Bailey in 50-100 Td but this curve tends quickly to the zero value below 15 Td. On the other hand for 30-70 Td it corresponds to our experimental results but for other values of E/N we can notice relatively higher differences. Agreement of Hayashi's model II (Hayashi, private information) in multiterm Boltzmann analysis with experimental values is better than of the model "Hayashi I", - the model II falls in-between present and Bailey's values. Unfortunately we do not know details of the cross sections used by Hayashi for that calculation.

3.3. Drift velocity

Good agreement of D_T/μ with Hayashi is not the case of the drift velocity, see fig. 4. In the calculation of the drift velocity from semiempirical cross sections any differences in input data influences directly the evaluation of the W value. Present W values are conformable to data of Phelps and Voshall [3], Pack [19] and Nielsen [20] but at intermediate reduced fields almost 50% lower than those calculated by Hayashi. This is a clear indication that the cross sections used by Hayashi should be verified. The present measurements indicates that the set of Kwan et al. [9] and Szymkowski et al. [8] is more appropriate for modelling diffusion coefficients at intermediate E/N values. Further modelling of diffusion coefficients from different sets of cross sections is planned.

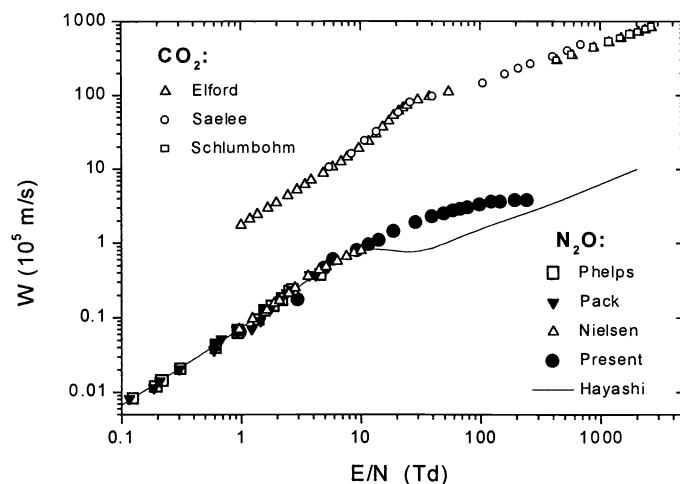


Fig. 3. Comparison between drift velocity data for CO₂ and N₂O.

4. CONCLUSION

In this work we present experimental results of the transverse diffusion coefficient to mobility (D_T/μ), the ratio of longitudinal diffusion coefficient to mobility (D_L/μ) and the drift velocities for electrons in nitrous oxide, in particular measurements of D_L/μ are shown the first time. Values of all presented electron transport parameters are generally in good agreement with data obtained by other authors. Moreover we produce the model of integral cross sections for electron scattering in N₂O for different kind of excitations (momentum transfer, vibrational, attachment, ionisation and electronic excitation). This model is used to reproduce of the transverse diffusion coefficient to mobility by numeric solution of Boltzmann equation. Results of calculations correspond to other models and experimental data.

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