PAPER

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IOP Publishing Plasma Sources Science and Technology

Plasma Sources Sci. Technol. **26** (2017) 044004 (4pp) https://doi.org/10.1088/1361-6595/aa61db

Reduced mobility of He^+ in CF_4

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Received 14 September 2016, revised 30 January 2017
Accepted for publication 21 February 2017
Published 23 March 2017

Abstract

This paper is devoted to a presentation of a cross section set for the scattering of He^+ ions in CF_4, which is assessed by using available experimental data for exothermic charge transfer cross sections that produce $CF_3{}^+$ and $CF_2{}^+$ ions and endothermic charge transfer cross sections that produce CF^+, C^+ and F^+ ions. Due to the significant particle losses, experimental transport coefficients have not been measured. Transport properties of He^+ ions in CF_4 needed for modeling discharges containing mentioned ions are calculated by the Monte Carlo method at a temperature of $T = 300$ K. Significant differences between flux and bulk transport coefficients are noticed, which is important for fluid models that exploit flux transport coefficients as input data.

Keywords: He^+, CF_4, Monte Carlo simulation, cross sections, transport coefficients

(Some figures may appear in colour only in the online journal)

1. Introduction

He-CF_4 mixtures are used in gas electron multipliers for various imaging purposes (x-rays, charged particles, thermal neutrons and dark matter detection) [1]. Bursts of electron multiplication affect the production of various ions that may affect time distribution of detected particles [2]. Experimental transport coefficients needed as input data for models for the transport of He^+ ions in CF_4 gas are missing. Although some experimental points for scattering cross sections of He^+ ions in CF_4 are obtained by Fisher *et al* [3], a cross section set that can be used in modelling is not established yet. Quantum-mechanical calculation of a particular cross section is a demanding task requiring knowledge of ion-molecule potential energy surface, which has to be constructed from the structure of the reactants. Less calculation-intensive methods such as Denpoh–Nanbu theory [4–6] require knowledge about thermodynamic formation data and are applicable for a range of molecules. Although in this case thermodynamic formation data are known, such an approach is hard to apply, since reaction does not proceed via the excited ($HeCF_4{}^{+*}$) complex but via excited states of $CF_4{}^+$ ($CF_4{}^{+*}$). It is also more appropriate to select threshold energies for reaction products from threshold energies of $CF_4{}^+$ states [7] than from enthalpies of formation.

Charge transfer reactions of ions with molecules are important elementary processes in modeling kinetics in all kinds of plasmas. In many cases, cross sections for these reactions are known to represent the most significant part of the cross section set. It can be concluded from observation of the line spectra of excited F atoms obtained in spectrometric measurements [7] in CF_4 that the charge transfer reaction is a dominant process in collisions with inert gas ions. That argument appears sufficient in neglecting other possible reactions.

The aim of the present paper is to report on a topic important both for fundamental studies and for applications. We assessed the cross section set for He^+ in CF_4 by using existing experimental data [3] for charge transfer collisions producing radical ions of CF_4. In the following section we will discuss compilation of existing data and establish one possible cross section set. Next we describe the calculation of transport parameters and at the end discuss our results. Flux and bulk reduced mobilities, calculated from flux and bulk drift velocities by Monte Carlo simulation are significantly different in the region of moderate E/N.

2. Cross section set

Our aim in this section will be to establish the cross section set since only the cross section set contains relevant information to calculate transport properties of selected ion in particular gas. In our selected case the general knowledge

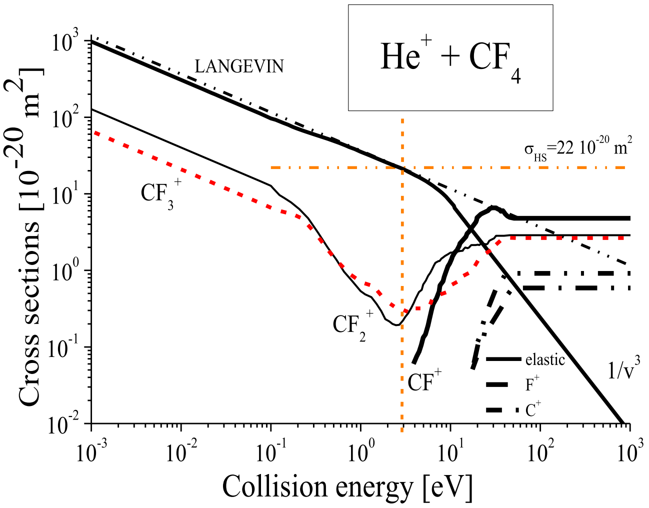

Figure 1. Cross section set for $He^+ + CF_4$.

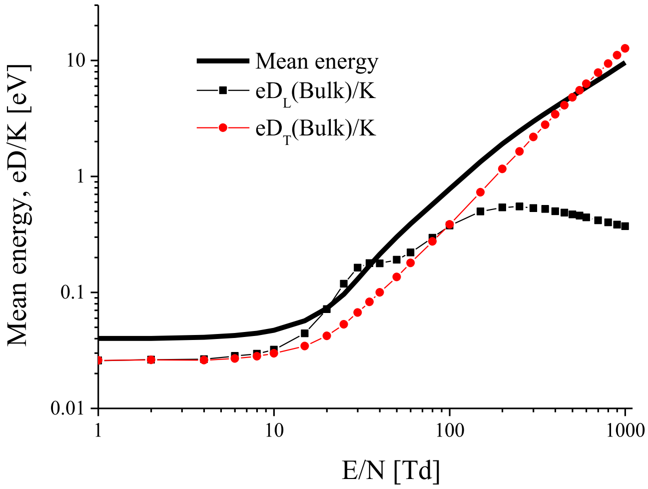

Figure 2. Mean energy and characteristic energy for He^+ in CF_4 as a function of E/N.

about the total cross section indicates that at low energies it would be affected by long-range attractive forces while at high energies by repulsive forces.

For small energies, when interaction potential is very close to induced dipole potential, one may assume that the total momentum transfer cross section is $\sigma_{mt} = 1.105\ \sigma_L$, where σ_L is the Langevin's cross section [8]. The Langevin's cross section was determined by using the average polarizability of the gas. The average polarizability of CF_4 is poorly determined [3] and may produce discrepancies in the calculated mobilities of ions in CF_4 [9, 10]. As a consequence, this would affect plasma parameters prediction in modeling. We adopted the value of 3.86 $10^{-30}\ m^3$ used by Stojanović *et al* [9] who found excellent agreement between experimental and calculated reduced mobility of ${CF_3}^+$ ions in CF_4.

From the exothermic cross section measurements of Fisher *et al* [3] for ${CF_2}^+$ and ${CF_3}^+$ production from $He^+ + CF_4$, one may conclude that scattering is appropriate to describe induced polarization potential up to 0.2 eV, thus assuming that charge transfer reactions are the dominant interaction one may obtain for the elastic momentum transfer cross section by deducing experimental reactive cross sections [3] from assumed total momentum transfer cross section.

When the collision energies are larger than the crossing point between the 1.105 σ_L curve and the hard sphere (HS) cross section (represents purely the repulsive part of the potential), repulsive interaction is beginning to dominate [3]. At the crossing point ($\sim$3 eV) the elastic momentum transfer cross section is smoothly connected to the $1/v^3$ trend [11, 12], where v is the center-of-mass velocity (see figure 1). This trend assumes that repulsive interaction is with anisotropic (forward) scattering probability.

Finally, in the cross section set all exothermic and endothermic cross sections of Fisher *et al* [3] are included. Reactive cross sections were approximated by constant values at all ion kinetic energies above 50 eV by using the data for production ratio between observed ions as suggested in [13].

Although the cross sections for ${CF_2}^+$ and ${CF_3}^+$ production are almost an order of magnitude lower than the total momentum transfer cross section their magnitude below 0.3 eV is significant and has to be known if ion transport at lower temperatures has to be modelled. From the calculations of Krstić and Shultz [11] assuming validity of capture theories down to about 0.1 meV extrapolation of the experimental measurements [3] towards low energies is safely done by $1/v$ trend. Below 0.1 meV we used extrapolation with the constant cross section.

3. Transport coefficients

Transport properties needed for modeling CF_4 discharges containing He^+ ions are calculated by the Monte Carlo method. A code that properly takes into account thermal collisions was used [14]. It has passed all relevant benchmarks [6] and was tested in our work on several types of charged particles [6, 15]. Swarm parameters of He^+ in CF_4 for a temperature of $T = 300$ K are presented.

The calculated transport parameters are the mean energy, characteristic energy, drift velocity, diffusion coefficients and rate coefficients for ions [16]. Note that these transport parameters are the only information present in the literature up to now and there are no published experimental data for the transport coefficients of He^+ in CF_4.

In figure 2 we present the mean energy, obtained as an average of particle ensemble [14, 16] and compare it to characteristic energies (diffusion coefficient normalized to mobility eD/K in units of eV, where $K = v_d/E$ and v_d is the drift velocity of the ion) determined in the direction of the field and transversal to the electric field. In figure 2 values that are taking into account only bulk values of transport coefficients that can be experimentally determined [17] are shown. They can be compared with values obtained when experimental measurements become available.

Characteristic energy curves above 200 eV show that the energy in the longitudinal direction is significantly affected by

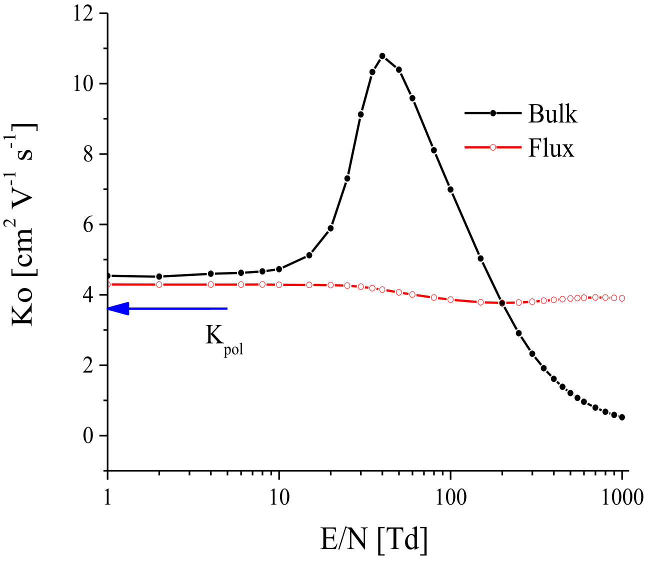

Figure 3. The bulk and flux reduced mobility for He^+ in CF_4 as a function of E/N.

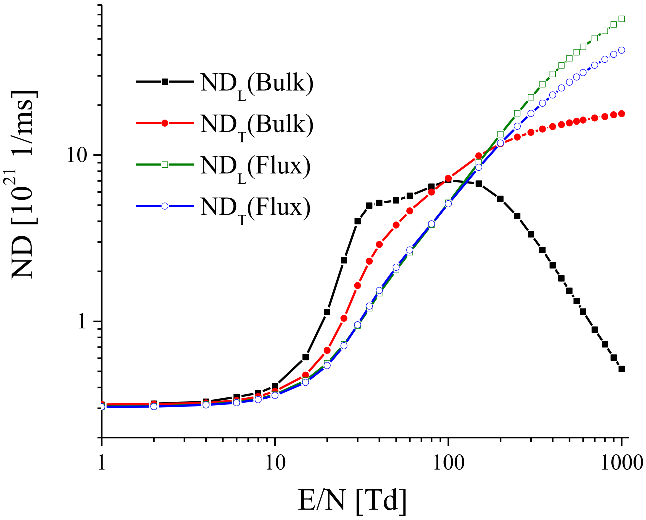

Figure 4. The transversal and longitudinal diffusion coefficients for He^+ in CF_4 as a function of E/N.

non-conservative collisions. Note that the mean energy cannot be directly measured in experiments. Mapping of mean energy versus E/N may be used directly to provide the data in fluid models in situations when local field approximation fails. As visible on the figure the energy increases from 10 Td and before that a thermal value of 0.04 eV.

The mobility K of an ion is a quantity defined as the velocity attained by an ion moving through a gas under the unit electric field. One often exploits the reduced or standard mobility defined as:

$$K_0 = \frac{v_d}{N_0 E} N, \tag{1}$$

where N is the gas density at elevated temperature T, $N_0 = 2.69 \cdot 10^{25}\,\mathrm{m}^{-3}$ and E is the electric field. Let us remind the reader that the bulk drift velocity ($W = \mathrm{d}\langle x \rangle/\mathrm{d}t$) is a reaction corrected flux drift velocity ($w = \langle v \rangle$): $W = w + S$, where S is the term representing a measure of the effect of reactions on the drift velocity. For flux mobility are responsible elastic collisions. The difference between bulk and flux reduced mobilities is a consequence of energy-dependent reactions.

Generally both flux and bulk reduced mobilities converge, when $E \to 0, T \to 0$, to a polarization limit value, when interaction between ion and molecule can be described with induced polarization potential (if one neglects reactions). The polarization limit value for our studied case is $K_{\mathrm{pol}} = 3.608\,\mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$ (see figure 3). If exothermic reactions take place, as in the our case, then reduced mobility acquires larger values since the elastic cross section is smaller at the expense of exothermic cross sections [5].

Flux and bulk values of reduced mobility for He^+ ions in CF_4 as a function of E/N (E-electric field strength, N-gas number density) are shown in figure 3.

Due to the exothermic processes producing ${CF_2}^+$ and ${CF_3}^+$ reduced mobility at lowest E/N is larger than polarization limit value. Flux reduced mobility is nearly flat at these E/Ns since collision frequency for elastic scattering is nearly constant [5]. When collision frequency for elastic collisions begins to decrease one would expect the increase of the flux reduced mobility. This is not happening since endothermic reactions remove high-energy ions from the swarm front. This shifts the center of mass of ions backward.

At the low E/N tail of thermal energy distribution function (EDF), decreasing part of the collision frequency for reactions allows the increase of ions energy at the front of the swarm. This affects the center of mass of the ions moving ahead (source term S is positive) and bulk reduced mobility is increasing.

For the ion energies larger than thermal, by increasing reduced electric field, EDF in a wider range senses the drop of high energy ions in the front of the swarm due to the exothermic reactions and so bulk reduced mobility is increasing steeply.

For the energies for which collision frequency for reactions begins to increase, fast ions are removed from the swarm front and bulk reduced mobility decreases with E/N.

Thus, the significant peak in the bulk reduced mobility at about 35 Td is obtained as a result of difference in energy dependence of elastic and exothermic cross sections [18].

Transversal and longitudinal diffusion coefficients are given in figure 4. At very low energies due to the similarity of the cross sections for elastic and exothermic scattering, flux and bulk diffusion coefficients are similar. Very large non-conservative effects, almost a reminder of the positron transport [19], are noticed at higher E/Ns. Similar to the results for drift velocity, flux diffusion coefficients are significantly larger than the bulk values at largest E/N's, due to the reactive collisions.

In figure 5 we show rate coefficients for elastic momentum transfer and for all reactive processes as a function of E/N. The rate coefficients as the final output of our calculations are needed as input in fluid equations for the description of ion transport in CF_4 gas.

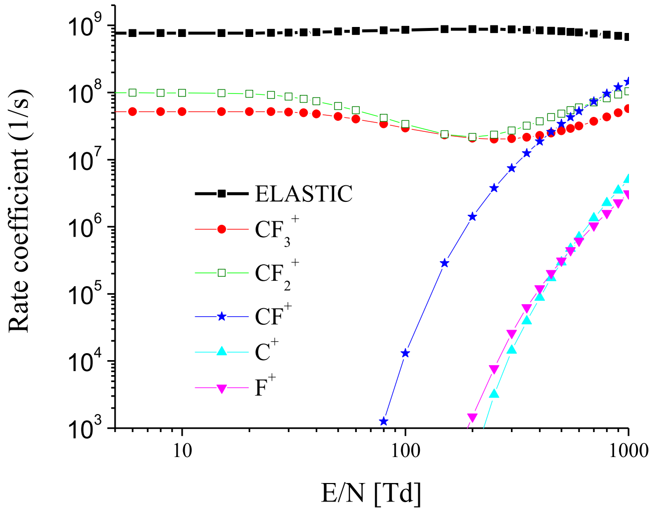

Figure 5. Rate coefficients for momentum transfer and for production reactions as a function of E/N.

Calculated rate coefficients are valid for swarm conditions. The precision of the calculated rate coefficients depends on the precision of the measured cross section, which is stated to be 50%–60% [3]. At higher energies, where cross sections for reactions are extrapolated, precision of the calculated rate coefficients is lower. This precision is fortunately increasing due to the measured cross sections ratio at 861 eV [13]. Note that exothermic rate coefficients obtained from capture theories are constant and are significantly different from the rate coefficients obtained in this work.

4. Conclusion

In this work we determined elastic momentum transport cross sections as a function of energy for He^+ scattering on CF_4 that can be used in modelling transport of He^+ in CF_4 gas. We exploited data for a simple theoretical total momentum transfer cross section and obtained an elastic momentum transfer cross section by deducing all experimentally obtained charge transfer cross sections. In that we assumed that measured charge transfer cross sections are the collisions with the highest probability. Thus, in this paper we have assessed the cross section set for He^+ ions in CF_4 that can be used as an independent input in the modelling transport of He^+ ions. This assessment was performed by using measured charge transfer cross sections.

Since to the best of our knowledge no direct information exists in the literature on how the mobility of high recombination energy ions such as He^+ ions behaves in CF_4, we calculated transport parameters by using the Monte Carlo simulation method [5, 18].

In this paper we have obtained and discussed both flux and bulk reduced mobility data. Data for swarm coefficients for positive and negative ions are needed for hybrid and fluid codes and the current focus on liquids or liquids in the mixtures with rare gases dictates the need to produce data compatible with those models. In view of the present interest in models of liquids and/or liquids in mixtures with rare gases, data for swarm coefficients for positive and negative ions are needed for hybrid and fluid codes.

Acknowledgments

This work was performed in the Laboratory of Gaseous Electronics Institute of Physics University of Belgrade financed by the Ministry of Education, Science and Technology of Serbia, Projects No. 171037 and 41001.

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