

Cross Sections of Charge Transfer by Slow Singly-Charged Carbon Ions from Various Hydrocarbon Molecules

KUSAKABE Toshio, IMOU Kazumichi, TOMITA Noriaki, HOSOMI Kenji,
NAKANISHI Hiroshi, SATOH Sinichi, NAKAI Yohta¹, TAWARA Hiroyuki²
and KIMURA Mineo³

Department of Science, Kinki University, Higashi-osaka 577-8502, Japan

¹ *Suzuka University for Medical Science, Suzuka 510-0293, Japan*

² *Max Planck Institute for Nuclear Physics, Saupfercheckweg 1, Heidelberg D-69117, Germany*

³ *Graduate School of Science, Kyushu University, Fukuoka 812-8581, Japan*

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Abstract

Single charge transfer cross sections for the dominantly ground state C^+ (2P) ions have been measured in collisions with CH_4 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_6 , $(CH_2)_3$ and C_3H_8 molecules in the energy range of 0.15 to 4.5 keV. These are compared with calculations based on a formula developed by Olson [Phys. Rev. A **6**, 1822 (1972)]. Most of the present data, except for those of CH_4 target, are found to show weak energy dependence over the collision energy range observed, and only slightly decrease with increasing the collision energy. This feature indicates that vibrational excited states of the target molecular ions produced after charge transfer might have resulted in creation of near or accidental resonant reaction channels.

Keywords:

single charge transfer cross section, slow ground state C^+ (2P) ion, hydrocarbon target, vibrationally excited molecular ion, accidental resonant reaction

1. Introduction

Knowledge on charge transfer processes of carbon ions colliding with various hydrocarbon molecules at low energies is important in basic research and in a number of applications such as astrophysics, plasma science and material science where low temperature plasmas play a key role. In particular, the edge plasmas in the current large tokamak devices contain a significant amount of carbon ions and hydrocarbons released from the carbon-based plasma-facing materials in their inner walls [1,2].

Although many investigations have been performed on charge transfer of ions in collisions with various atoms and molecules, to our knowledge, only a few data sets are available for collisions of C^+ ions with hydrocarbons so far: The cross sections for CH_4 , C_2H_6 and C_3H_8 at energies from 6.3 to 16.3 keV by using the recoil ion source [3] and for CH_4 and C_2H_6 at energies from 0.1 to 1.5 MeV [4] have been reported. In the C^+ (2P) ions + CH_4 collision, production cross sections of the secondary CH_n^+ ($n = 1 \sim 4$) and $C_2H_m^+$ ($m = 1, 2, 3$) ions have been measured in the energy range of 2 to 200 eV [5]. Most recently the isomer effect in charge transfer cross sections has been observed in two

isomeric C_3H_4 molecules (allene and propyne) at energies from 0.2 to 4.5 keV and analyzed theoretically [6]. On the other hand, Olson derived a simple formula for evaluating the charge transfer cross sections for singly charged ions colliding with atoms based on a Demkov-type approximation [7]. Even for molecular targets, this formula has been known to provide gross features [8 - 10].

In this paper, we present the single charge transfer cross sections of the ground state C^+ (2P) ions in collisions with CH_4 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_6 , $(CH_2)_3$ and C_3H_8 molecules in the energy range of 0.15 to 4.5 keV. The contribution of the metastable state ions is minimized by controlling the ionizing electron energy in the ion source. The present experimental results are compared with calculated results using Olson formula.

2. Experimental

A detailed description of the present experimental apparatus and methods has been previously given [8-10] and so only some important features in the present work are mentioned here.

Table 1 Charge transfer cross sections (in units of 10^{-16} cm²/molecule) for C⁺ ions in collisions with CH₄, C₂H₂, C₂H₄, C₂H₆, C₃H₆, (CH₂)₃ and C₃H₈ molecules.

Energy (keV)	Target molecules						
	CH ₄	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₃ H ₆	(CH ₂) ₃	C ₃ H ₈
0.15	5.54 ± 0.98	—	—	—	—	—	—
0.2	4.51 ± 0.80	38.7 ± 6.5	34.0 ± 5.6	21.4 ± 3.5	68.2 ± 9.4	71.9 ± 10.0	58.4 ± 8.0
0.25	3.41 ± 0.61	—	—	—	—	—	—
0.3	2.68 ± 0.49	34.5 ± 5.6	33.3 ± 5.4	16.2 ± 2.7	53.7 ± 7.4	58.6 ± 8.1	44.7 ± 6.2
0.4	2.37 ± 0.42	—	—	13.4 ± 2.2	—	—	—
0.5	2.27 ± 0.41	29.8 ± 4.9	29.9 ± 4.9	—	42.2 ± 5.8	48.4 ± 6.7	33.9 ± 4.7
0.6	—	—	—	12.1 ± 2.0	—	—	—
0.7	2.03 ± 0.22	—	—	—	—	—	—
0.8	—	25.6 ± 4.2	29.1 ± 4.8	11.3 ± 1.9	36.4 ± 5.0	39.8 ± 5.5	28.4 ± 3.9
1.2	1.99 ± 0.22	20.8 ± 3.0	27.5 ± 3.8	12.2 ± 2.0	32.9 ± 3.5	35.7 ± 3.9	26.0 ± 2.8
2.0	2.40 ± 0.26	22.7 ± 3.2	27.8 ± 3.8	16.1 ± 2.3	32.8 ± 3.5	33.7 ± 3.7	27.5 ± 3.0
2.7	2.99 ± 0.32	—	—	—	—	—	—
3.0	—	24.1 ± 3.3	26.5 ± 3.7	18.6 ± 2.6	33.2 ± 3.6	32.3 ± 3.5	29.9 ± 3.2
3.5	3.95 ± 0.42	—	—	—	—	—	—
4.5	5.28 ± 0.56	23.0 ± 3.2	27.8 ± 3.8	20.4 ± 2.8	33.1 ± 3.6	32.9 ± 3.6	31.2 ± 3.3

An electron impact ion source was used for C⁺ ion production from CO gas. In order to get the ground state C⁺ (²P) ions, the effective electron energy was set to be 25.3 eV [8]. The mass-analyzed C⁺ ion beam was introduced into a 40 mm long collision cell. The target gases of high purity (> 99.5 %) were fed into the cell which pressure was measured with a calibrated Pirani gauge. After collisions, both the incident C⁺ ions and product neutral carbon atoms were charge-separated with the electrostatic parallel plates and were detected with a micro-channel plate position-sensitive detector. The charge transfer cross sections were derived based on the growth rate method. Namely, the cross sections were determined from the slope of the linear part in the curve of observed fraction for neutral carbon atoms formed in charge transfer collisions versus the target gas thickness.

Total experimental uncertainties of the absolute cross sections have been estimated to range from 10.6 % at high energies to 18.0 % at low energies from the quadratic sum of possible uncertainties including that of the target thickness.

3. Results and discussion

The present data for the cross sections of single charge transfer by C⁺ (²P) ions for various hydrocarbons are given in Table 1 together with the estimated uncertainties.

3.1 Paraffin hydrocarbons (CH₄, C₂H₆, C₃H₈)

The observed charge transfer cross sections for some saturated hydrocarbons, CH₄, C₂H₆, and C₃H₈, are shown in Fig. 1 together with the previous data of at higher energies [3, 4]. As the collision energy increases, the present cross sections decrease and show a minimum at around 1 keV. Then, at further high energies, they increase and are smoothly connected with the previous data of Itoh *et al.* [3]. Also the sum of the cross sections of all the secondary ions, CH_n⁺ ($n = 1 \sim 4$), measured by Wilson *et al.* [5] has been found to coincide with the present data at lowest energies. In Fig. 1, the broken curves show the calculated results using Olson formula for the charge transfer into the ground C (³P) state atom from ground state target hydrocarbons. This process is endothermic for CH₄ and C₂H₆, while exothermic for C₃H₈. It is generally known that the variation of the cross sections as a function of the collision energy becomes weak as the energy defect ΔE in the charge transfer process becomes small [11]. The exothermic reaction in C⁺ + C₃H₈ collision meets the accidental resonant condition [9,10,12] if product molecular ions are in some vibrational excited states. In Fig. 1 (c), the solid curve is represented the calculated results for such an accidental resonant case ($\Delta E = 0$).

3.2 Acetylene hydrocarbons (C₂H₂, C₃H₄ (propyne) and isomer (C₃H₄ (allene)))

The observed cross sections for C₂H₂, C₃H₄ (propyne) as another acetylene hydrocarbons and its

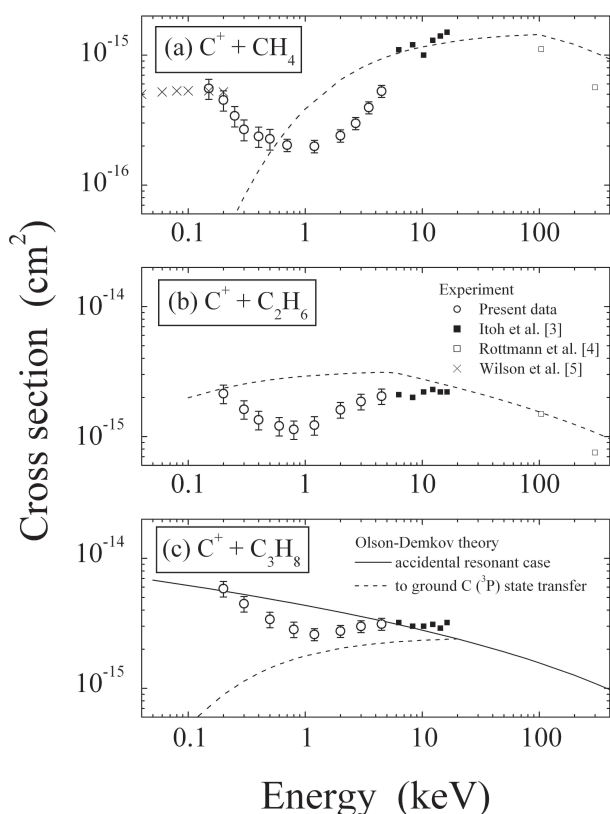


Fig. 1 Single-charge transfer cross sections for C^+ ions in collisions with paraffin hydrocarbons: (a) CH_4 , (b) C_2H_6 , (c) C_3H_8 .

isomer C_3H_4 (allene) [6] are shown in Fig. 2 (a), (b) and (c), respectively. The present data for these three hydrocarbons gradually decrease as the incident energy increases and level off above 1 keV. Although the data for two acetylene hydrocarbons, C_2H_2 and C_3H_4 (propyne), show similar energy dependence, those for C_2H_2 are roughly 0.63 times smaller than for C_3H_4 (propyne) over the energy range investigated in the present work. The calculated results for the endothermic reaction of charge transfer into the ground $C(^3P)$ state in $C^+ + C_2H_2$ collisions (broken curve in Fig. 2 (a)) seem to be slightly different from the present data. For both C_3H_4 hydrocarbons, similar reactions are fully exothermic, so that the charge transfer into the ground $C(^3P)$ state can be accidentally resonant, if some vibrational excited states of product molecular ions are taken into account. The calculated results for resonant reaction are shown in Fig. 2 (b) and (c) with the solid curves. It is noted that, at lower energies, the cross sections for allene are slightly larger than those for propyne. This is due to the fact that charge transfer into the excited $C(^1D)$ state (dot-dashed curve in Fig. 2) is exothermic for allene, while this channel of propyne is endothermic. This isomer effect in the charge transfer processes have previously been discussed in details together with more precise theoretical treatment using a

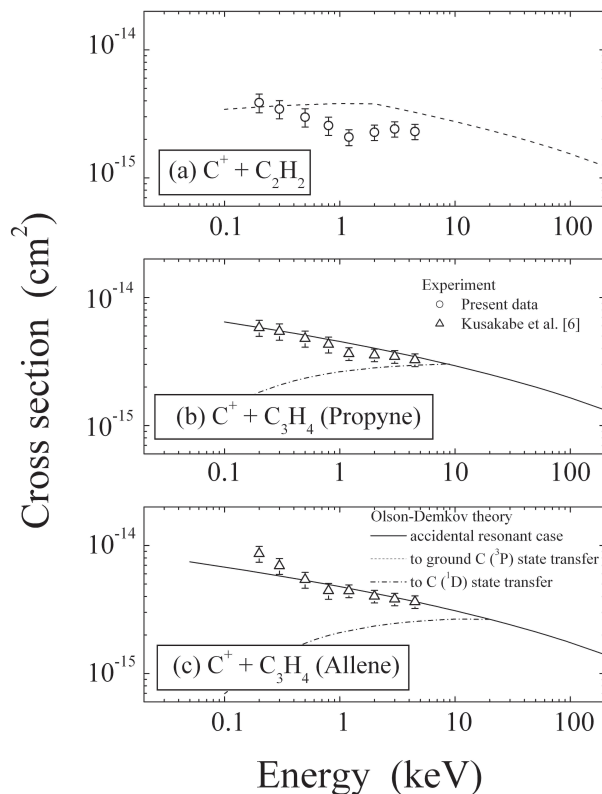


Fig. 2 Single-charge transfer cross sections for C^+ ions in collisions with acetylene hydrocarbons and isomer: (a) C_2H_2 , (b) C_3H_4 (propyne), (c) C_3H_4 (allene).

molecular orbital expansion method [6].

3.3 Olefin hydrocarbons (C_2H_4 , C_3H_6) and isomer ($(CH_2)_3$)

Figure 3 also compares the observed data for olefin hydrocarbons (C_2H_4 and C_3H_6) and C_3H_6 isomer $(CH_2)_3$. The cross sections are practically the same for both C_3H_6 and $(CH_2)_3$ molecules, though significant difference between two olefins, C_2H_4 and C_3H_6 , is observed at lower energies. Moreover, it is found that the energy dependence of the cross sections of allene (see 3.2) is very similar to that of C_3H_6 and $(CH_2)_3$. This is due to the fact that they have similar energy defect and are exothermic in charge transfer into the excited $C(^1D)$ state, while this channel of propyne is endothermic.

4. Summary

We have measured the single-charge transfer cross sections of the ground state $C^+(^2P)$ ions from various hydrocarbons in the collision energy range from 0.15 to 4.5 keV. The observed cross sections have been compared with the calculations using the Olson formula. In most collisions with the exothermic reaction channels, vibrational excited states of the target molecular ions after charge transfer result in near or accidental resonant processes. Generally the data

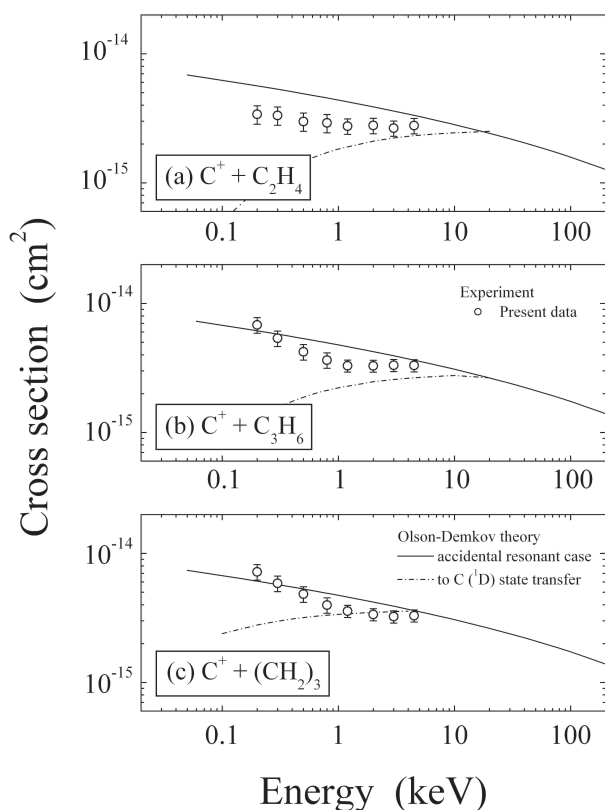


Fig. 3 Single-charge transfer cross sections for C^+ ions in collisions with olefin hydrocarbons and isomer: (a) C_2H_4 , (b) C_3H_6 , (c) $(CH_2)_3$.

decrease slowly as the collision energy increases. At the energy above 2 keV, the present data, except for CH_4 , are found to roughly proportional to $I_1^{-3.5}$ where I_1 is the first ionization potential of target hydrocarbons. However, it is difficult to make a simple empirical relation for the present data below 1 keV. Moreover, for some endothermic reactions for CH_4 , C_2H_2 and C_2H_6 , their energy dependence below 1 keV can not be reproduced qualitatively. These can be caused by the vibrational excitation of target hydrocarbons

prior to the actual charge transfer reactions occurring. Extensive theoretical analysis would provide clues for understanding such effect.

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