

## Ions produced from condensed CO targets under energetic ion impact

H. Tawara

*National Institute for Fusion Science, Nagoya 464-01, Japan*

T. Tonuma and H. Kumagai

*Institute for Physical and Chemical Research, Wako 351-01, Japan*

T. Matsuo

*Medical Research Institute, Tokyo Medical and Dental University, Yushima, Tokyo 113, Japan*

(Received 11 September 1992)

Various secondary ions have been produced under 1.5-MeV/amu  $\text{Ar}^{13+}$ -ion impact on a condensed CO target. Multiply charged  $\text{C}^{i+}$  and  $\text{O}^{i+}$  ions including H- and He-like ions have been observed to be unexpectedly large compared with condensed rare-gas targets and understood to be due to large initial kinetic energies, which are gained through Coulomb explosion of multiply charged molecular ions produced inside condensed targets during energetic collisions. The prominent positive cluster ions are  $[\text{CO}(\text{CO})_n]^+$  as expected, whereas the negative cluster ions are  $[\text{C}_n]^-$ .

PACS number(s): 34.50.Fa, 34.50.Gb, 79.20.-m

In energetic ( $\sim 1$  MeV/amu) highly-charged-heavy-ion collisions with rare-gas targets, secondary ions in very high charge states are known to be produced more efficiently [1], compared with those in electron impact [2]. Previous investigations [3–5] show that, in contrast with those from rare-gas atom targets [6], multiply charged secondary atomic ions from molecular targets have large kinetic energy, which is provided through Coulomb explosion of multiply charged molecular ions produced in collisions. The observed  $\text{I}^{17+}$  ions, which are likely to result from the dissociation of  $\text{I}_2^{34+}$  molecular ions produced under 1-MeV/amu Ar-ion impact on  $\text{I}_2$  molecules, are found to have their initial kinetic energy of more than 1000 eV [3].

In the present work, we investigate ions from condensed CO molecular-gas layers under energetic ion impact. Such collisions, involving condensed gas layers, are known to occur in outer space, where cosmic rays of high energy hit cold interstellar-planetary dusts [7]. Laboratory investigations involving condensed gas layers have been limited under kilo-electron-volt ion impact [8], and are few in high-energy collisions [9].

1.5-MeV/amu  $\text{Ar}^{13+}$  ions of a few picoamperes are sent into a target chamber whose background pressure is around  $(1-2) \times 10^{-9}$  Torr. CO gas of  $10^{-9}$ – $10^{-8}$  Torr is continuously introduced into the target chamber and condensed on a cooled 1- $\mu\text{m}$ -thick Al foil, whose temperature could be varied down to 15 K [10]. The secondary ions are detected with a channeltron after magnetic analysis. Relative ion intensities are found to be practically unchanged when the foil temperature is varied, as long as the target gas molecules form condensed layers thick enough to produce secondary ions.

A typical high-charge spectrum of ions produced from a condensed CO target is shown in Fig. 1(b), together with that from a gaseous CO target [11] [Fig. 1(a)]. In

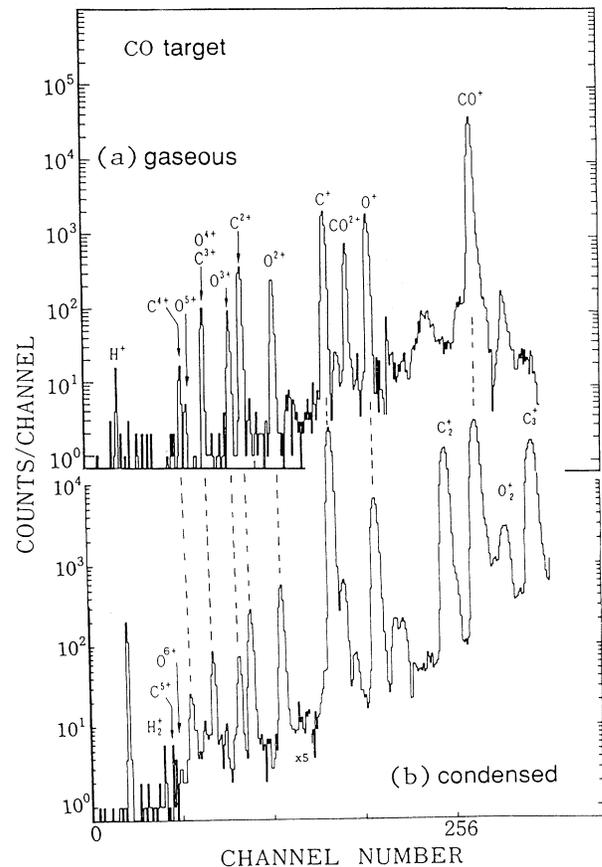


FIG. 1. Mass-to-charge-ratio spectra of multiply charged secondary ions: (a) gaseous CO target [11], and (b) condensed CO target.

these spectra we can see the following features:

(a) Both the parent  $\text{CO}^+$  and the dissociation-product  $\text{C}^+$  ions are of comparable intensities in the condensed target, while in the gaseous target,  $\text{CO}^+$  ions are stronger by roughly an order of magnitude over  $\text{C}^+$  ions.

(b) In the condensed target,  $\text{C}^+$  ions are stronger by roughly an order of magnitude than  $\text{O}^+$  ions. On the other hand,  $\text{C}^+$  and  $\text{O}^+$  ions are roughly equal to each other in the gaseous target.

(c) Doubly charged parent gas molecular  $\text{CO}^{2+}$  ions are very weak (only  $10^{-3}$  of  $\text{CO}^+$ ) in the condensed target, while the doubly charged ions have been found to be about 4% of  $\text{CO}^+$  ions in the gaseous target [11].

(d) Multiply charged  $\text{C}^{i+}$  (up to  $i=5$ ) and  $\text{O}^{i+}$  (up to  $i=6$ ) ions are observed in condensed (as well as gaseous) targets. It should also be noted that the ion intensity ratios of the neighboring charge ( $i+1/i$ ) for high  $i$  are roughly the same for both gaseous and condensed targets.

(e) In the condensed target  $\text{C}^+$ ,  $\text{C}_2^+$ , and  $\text{C}_3^+$  ions are found to have roughly comparable intensities [see Fig. 1(b)], whereas in gaseous targets no such cluster ion is observed. It should be also pointed out in condensed targets that  $\text{O}^+$  and  $\text{O}_2^+$  ions are much weaker, compared with  $\text{C}^+$  ions.

Most of the features observed above in condensed molecular-gas targets can be understood qualitatively, based on those in a gaseous molecular target [3]: a large initial kinetic energy of multiply charged atomic ions, gained from Coulomb explosion energy, which depends upon the charge state of ions at the moment of dissociation of multiply charged molecular ions.

Feature (a) clearly demonstrates that  $\text{CO}^+$  ions get only minor recoil energy (of a few hundred milli-electron-volts) [6] through direct ionization process, while  $\text{C}^+$  ions get more than 10 eV during the course of dissociation [11].

Feature (b) might suggest that  $\text{C}^+$  ions should get, on average, a kinetic energy higher than  $\text{O}^+$  ions, having higher escape probabilities from the surface. Experiments [12,13] indeed show that  $\text{C}^+$  ions have slightly higher energies than  $\text{O}^+$  ions, the difference being enhanced at heavier projectiles. This slight difference in the initial kinetic energy of the product atomic ions might result in a significant difference in  $\text{C}^+$  and  $\text{O}^+$  ion yields from condensed CO layers.

Feature (c) is understood from the fact that  $\text{CO}^{2+}$  ions also get a small recoil energy, as in  $\text{CO}^+$  ions, which result in low escape probabilities from surfaces (see discussion below on condensed rare-gas targets).

Feature (d) is in sharp contrast with those in condensed rare-gas targets, where very weak or practically no multiply charged ion ( $i \geq 3$ ) has been seen [10]. Indeed, measurements of rare-gas recoil ions under collision conditions similar to the present work show that their recoil energy is less than 1 eV for recoil ions of  $i=6-7$  [6], which is not sufficient to escape the surface binding before neutralization.

Feature (e) indicates that the dissociated carbon products stay on the condensed surfaces and form cluster ions through subsequent collisions. In fact, many measurements show some black residues (most likely the dissoci-

ated carbon atoms) on the surfaces of the condensed CO layers after bombardment. It is also noted that, in low-energy heavy ions [13],  $\text{C}_2^+$  to  $\text{C}^+$  ratios are only of the order of  $10^{-2}$ . On the other hand, weak  $\text{O}^+$  and  $\text{O}_2^+$  ions (smaller than  $\text{C}_2^+$  ions by an order of magnitude over  $\text{C}^+$  ions) suggest that the dissociated O atoms tend to leave the surface. The present observation, showing no  $\text{CO}_2$  ion and  $\text{CO}_2$ -based cluster-ion products, seems to be in contradiction to a speculation that formation of  $\text{CO}_2$  through chemical reaction of the parent CO with dissociated free O atoms leaves excess carbon residue on the surface [13].

Multiply charged  $\text{C}^{i+}$  ions were also observed in some other condensed targets, such as  $\text{C}_2\text{H}_2$  [14], but not in condensed  $\text{CH}_4$  [10] or  $\text{CO}_2$  [15] molecules. This difference can be understood through chemical structures, which determine the sharing of the Coulomb explosion energy [16] between multiply charged atomic ions during dissociation.

Presently, only little understanding has been obtained on collision mechanisms involving energetic ions in condensed gas targets. At the present collision energy, the collisions are in the so-called electronic collision regime. There the emission of ions from condensed layers under energetic particle collisions is caused through electronic excitation of targets, where a part of the energy of the incident particles is imparted to electrons of the target atoms or molecules and converted into their motion, and finally some of them leave the surfaces, if they get sufficient energy. As far as the production of multiply charged ions in collisions is concerned, the main mechanism responsible for ion productions should be the direct ionization of target species. As discussed already, the recoil energy of rare-gas ions under collisions with highly charged projectiles is much less than 1 eV. As the neutralization time of such ions in condensed gas phase targets is estimated to be of the order of  $10^{-13}$  s [17], a large part of the slow-product rare-gas ions is lost on the way to the surface, and should have very low probabilities of leaving the surface before being neutralized. On the other hand, as the Coulomb explosion times of multiply charged molecular ions produced in collisions are much shorter ( $\approx 10^{-15} - 10^{-14}$  s) [13] and the dissociated atomic ions get kinetic energy of the order of 10 eV, the atomic ions have high probabilities of escaping the surfaces, resulting in higher yields of multiply charged atomic ions from condensed molecular-gas targets, compared with those in condensed rare-gas targets.

It is necessary to briefly mention features in spectra of heavier positive cluster ions from a condensed CO target. As expected,  $[\text{CO}(\text{CO})_n]^+$  ions consisting of only the parent molecules are the most prominent. Their intensities up to  $n=6$  decrease smoothly as  $n^{-0.22}$ . At  $n=7$ , the yields are reduced drastically, and over  $n=7-11$  again decrease smoothly but less slowly as  $n^{-0.16}$ . This sudden change of cluster-ion intensities indicates that the stabilities of the structures of these cluster ions are varied between  $n=6$  and 7, suggesting that stable  $[\text{CO}(\text{CO})_6]^+$  cluster ions have a structure of a bipyramidal square with a  $\text{CO}^+$  core ion located at its center. Further increase of  $n (> 12)$  shows a slow reduction of intensities. Most of

these features in positive cluster-ion production are similar to those in kilo-electron-volt ion impact observed by Jonkman and Michl [13], who show that, in light-ion impact, larger cluster ions decrease rapidly as  $n$  increases, compared with those in heavy-ion impact.

In contrast, the observed prominent negative ions from condensed CO targets are  $[C_n]^-$ , which do not include any parent CO molecules but only dissociated carbons, suggesting that the dissociated carbon atoms stuck on the surface form negative cluster ions.

In conclusion, in the present work, unexpectedly intense multiply charged  $C^{i+}$  (up to  $i=5$ ) and  $O^{i+}$  (up to  $i=6$ ) ions from a condensed CO target have been ob-

served, compared with those from condensed rare gases, where very weak or practically no multiply charged ( $i \geq 3$ ) ion could be observed [10]. As expected, the most prominent positive cluster ions are found to be  $[\text{CO}(\text{CO})_n]^+$  based on the parent CO molecules, while the dominant negative ions  $[C_n]^-$  are composed only of the dissociated carbon atom products.

Information on the energy distributions of ions, as well as neutral atoms, which are not treated here, that are produced from condensed gases would provide more insight into collisions, the understanding of which is still primitive [18].

- 
- [1] C. L. Cocke, *Phys. Rev. A* **20**, 749 (1979); T. Tonuma, H. Shibata, S. H. Be, H. Kumagai, M. Kase, T. Kambara, I. Kohno, A. Ohsaki, and H. Tawara, *ibid.* **33**, 3047 (1986); C. L. Cocke and R. E. Olson, *Phys. Rep.* **205**, 153 (1991).
- [2] H. Tawara and T. Kato, *At. Data Nucl. Data Tables* **36**, 167 (1987).
- [3] H. Tawara, T. Tonuma, K. Baba, T. Matsuo, M. Kase, T. Kambara, H. Kumagai, and I. Kohno, *Nucl. Instrum. Methods B* **23**, 203 (1987).
- [4] R. Mann, F. Folkmann, R. S. Peterson, Gy. Szabo, and K. O. Groenenveld, *J. Phys. B* **11**, 3045 (1978).
- [5] E. Krishnakumar, V. Krishnamurti, F. A. Rajgara, U. T. Raheja, and D. Marthur, *Phys. Rev. A* **44**, 4098 (1991).
- [6] R. E. Olson, J. Ullrich, and H. Schmidt-Böcking, *J. Phys. B* **20**, 809 (1987).
- [7] L. J. Lanzerotti, W. L. Brown, and R. E. Johnson, *Nucl. Instrum. Methods B* **14**, 373 (1986).
- [8] D. E. David, T. F. Magnera, R. Tian, D. Stulik, and J. Michl, *Nucl. Instrum. Methods B* **14**, 378 (1986).
- [9] E. F. Da Silveira, M. G. Blain, and E. A. Schweikert, *J. Phys. (Paris) Colloq.* **50**, C2-79 (1989); L. J. Lanzerotti, W. L. Brown, and K. J. Marcantonio, *Astrophys. J.* **33**, 910 (1987).
- [10] H. Tawara, T. Tonuma, H. Kumagai, and T. Matsuo, *J. Phys. B* **25**, L423 (1992).
- [11] T. Matsuo, T. Tonuma, M. Kase, T. Kambara, H. Kumagai, and H. Tawara, *Chem. Phys.* **121**, 93 (1988).
- [12] R. M. Wood, M. F. Steuer, and A. K. Edwards, *J. Chem. Phys.* **69**, 4487 (1979); G. Sampoll, R. L. Watson, O. Heber, V. Horvat, K. Wohrer, and M. Chabot, *Phys. Rev. A* **45**, 2903 (1992).
- [13] H. T. Jonkman and J. Michl, *J. Am. Chem. Soc.* **103**, 733 (1981); J. Michl, *Int. J. Mass Spectrom. Ion Phys.* **53**, 255 (1983).
- [14] T. Tonuma, H. Kumagai, T. Matsuo, H. Shibata, and H. Tawara, *Nucl. Instrum. Methods B* **67**, 544 (1992).
- [15] H. Tawara, T. Tonuma, H. Kumagai, and T. Matsuo, *J. Chem. Phys.* **94**, 2730 (1991).
- [16] H. Tawara, T. Tonuma, H. Shibata, S. H. Be, H. Kumagai, M. Kase, T. Kambara, and I. Kohno, *Z. Phys. D* **2**, 239 (1986).
- [17] F. Besenbacher, J. Böttiger, O. Graversen, J. L. Hansen, and H. Sorensen, *Nucl. Instrum. Methods* **191**, 221 (1981).
- [18] R. E. Johnson, B. U. R. Sundqvist, A. Hedin, and D. Fonyö, *Phys. Rev. B* **40**, 49 (1989).