LYCEN 9431
Septembre 1994

Fragmentation of high-energy ionic hydrogen clusters by single collision with helium

S. Ouaskit*, B. Farizon, M. Farizon, M. J. Gaillard, and A. Chevarier, N. Chevarier, E. Gerlic, M. Stern.

Institut de Physique Nucléaire de Lyon, IN2P3 - CNRS et Université Claude Bernard, 43, boulevard du 11 Novembre 1918, 69622 Villeurbanne Cedex, France

Abstract

Fragmentation of mass-selected 60-keV/amu-H_n^+ induced by single collision with helium has been studied for various cluster sizes n (9, 13, 21, 25, and 31). The absolute cross sections of the charged fragments H_p^+ are measured from p equal 3 to n-2. The deduced mass distributions are strongly different from those obtained at lower collision energy (where molecular evaporation is mainly involved) due to a strong production of ionic fragments with a size of $p/n \leq 0.5$. Moreover, the distributions for $p/n \leq 0.5$ are found to have scaling properties and to follow a power law $A^{-\tau}$, where A is the normalized fragment mass (p/n) and τ an exponent close to 2.6.

key words : high-energy cluster collision, fragmentation phenomena, Coulombic explosion, ionic hydrogen clusters.

*Permanent address : Université Hassan II, Faculté des Sciences II, B.P. 6621 - sidi Othmane, Casablanca, Maroc

To be published in Int. J. Mass Spectrum Ion Processes

Fragmentation phenomena are of experimental interest in various fields of physics and are the object of a fast developing field of statistical mechanics which concerns many systems of the microscopic world including polymers, gels, atomic clusters or atomic nuclei(1). A particular significant outcome of this research is the realization that in any circumstances the fragment size distribution and related quantities are invariant. Fragmentation of atomic clusters has been studied experimentally by electron(2) and photon interaction(3) and low-energy atomic collision (few eV/amu)(4,5). Up to now, mainly atomic or molecular evaporation has been observed(2). Recently, fission processes of doubly charged clusters have been reported for ionized van der Waals(6,7) and metallic(8,9) clusters. Directly pertinent to the present work are also the studies of the dynamical fragmentation of C_{60}^+ (10) ions and the fragmentation of CH_4^+ (11) caused by fast-proton impact.

The availability of high-energy mass-selected ionic hydrogen clusters gives a new interest in collision-induced fragmentation. Indeed, in this velocity range (1.55 v_0, v_0 being the Bohr velocity), the relative velocity between the projectile and the target atom is around or greater than the velocity of the electrons in the cluster. Then, the typical time for a collision with a target atom is short enough, compared to the typical time of the motion of the protons in the cluster, so that during the collision the protons can be considered to be stationary in the projectile frame(12). Moreover, in this velocity range, the collision can induce large electronic excitation up to ionization. Thus, processes such as ionization of the incident cluster followed by the dissociation of the transient multicharged cluster can be involved

In this paper, we report on the experimental measurements of charged fragment production after fragmentation of 60-keV/amu H_n^+ clusters (n=9, 13, 21, 25, 31) induced by single collision with helium.

The experiment was performed at the high-energy cluster-beam facility of the Institut de Physique Nucléaire de Lyon (13). The experimental apparatus is described in details in ref 14. In short, neutral clusters are formed in a cryogenic source and then ionized by electron impact. Cluster ion beams of 0.54, 0.78, 1.26, 1.50 and 1.86 MeV energies are formed by the new radiofrequency (RF) quadrupole post accelerator associated to the Cockroft Walton accelerator. The beam is pulsed with a cycle combining the cluster source cycle and the RF power one. After momentum analysis by a magnetic field, the incident beam is defined by two collimating apertures which ensure an angular dispersion of ± 0.8 mrad. Before reaching the gas target, the beam goes through two parallel plates. A voltage which depends on the energy of the beam can be applied or set to zero between these two plates with a fast voltage amplifier. Thus, a cluster burst can either be deflected in the monitoring surface-barrier (SB) detector or reach the target. The intensity of the collimated incident cluster beam in a burst corresponds to about 1000 clusters per second. The target is a gas-jet system described in detail previously [15]. A differential pumping system is able to maintain a residual gas pressure of 5.10^{-7} Torr in the region close to the gas jet and along the beam line. The charged fragments are magnetically deflected in a movable SB detector located one meter after the target. The corresponding time of flight is 0.30 μs. Care was taken to ensure a complete collection of the fragments for a given q/m range. The detector is connected to a pulse height analyzer, the signal being proportionnal to the total energy of the detected fragments.

The mass spectrum of charged fragments H_p^+ from mass p=3 to 29 obtained by collision of H_{31}^+ clusters with helium atoms is displayed in fig. 1a. This spectrum clearly shows 15 separated peaks where the total energies are ranging from 0.18 to 1.86 MeV. The highest energy peak corresponds to the detection of the transmitted H_{31}^+ clusters. Mainly fragments of odd mass number are obtained as already observed in the spectrum of the cluster source. The width of the peaks increases with the size of the detected fragment. This effect connected to the response of the detector will be discussed in a forthcoming paper. This spectrum is obtained for a given number of incident clusters with a target thickness of $0.73 \pm 0.08\ 10^{14}$ atoms/cm^2. Fig. 1b displays the spectrum obtained without gas target for the same number of incident clusters. These spurious events correspond to about 3% of the incident clusters. The behaviour of this spectrum looks similar to the one obtained with the gas target except for the H_{29}^+ fragments whose production is relatively enhanced. The spurious events are likely due to collisions of incident clusters with the residual gas and to spontaneous evaporation of a H_2 molecule after the magnetic analysis. This beam analysis takes place 10 μs after the cluster ionization at the end of the accelerator and 1 μs before the detection.

From the spectra 1a and 1b, we can deduce that only 22% of

H_{31}^+ incident clusters are dissociated in the gas jet in agreement with the dissociation cross section measured previously ($\sigma^{diss} = (37.2 \pm 1.7)\ 10^{-16}\ cm^2$)[14]. Therefore, the fragment yields are measured under single-collision conditions. The absolute cross sections of the fragment production are deduced by dividing the number of fragments per incident cluster by the target thickness after substraction of spurious events. The thickness of the gas jet depends on two parameters, the inlet gas pressure measured with a precision better than 0.3%, and the position of the jet capillary with respect to the incident ion beam known with a precision better than 4%. The position of the jet capillary with respect to the incident cluster beam is kept the same for the various cluster sizes. The determination of the target thickness and tests of the absolute calibration are described in ref. 15. The errors in the absolute cross sections deduced from these measurements vary from ±10% to ±15%, taking into account the target thickness and the statistical uncertainties. Similar studies have been done for H_{25}^+, H_{21}^+, H_{13}^+ and H_9^+incident clusters.

The production cross sections of ionic fragments of mass p (p=3 to 23) resulting from the fragmentation of H_{25}^+ clusters colliding with helium at 60 keV/amu are presented in fig. 2a versus the fragment mass p. The main feature of the distribution is the prominent production of clusters of masses which are intermediate (p ≅3 to 13). Due to this intermediate-mass fragments production, the behaviour of the fragment production is clearly different from the one obtained in low-energy collision[5] (few eV/amu) of H_{25}^+ clusters on helium (Fig. 2b) where mainly (sequential) molecular evaporation results from the collision. This last process leads to a fragment distribution where the yield of each fragment increases with its size since a small energy transfer in the collision is more probable than a large one. In our case, the molecular evaporation process can be connected to the right side of the fragment mass distribution but cannot explain the entire distribution. Besides this general behaviour, one can remark some specific features in the p range from 15 to 23. Moreover, the H_9^+ fragment production is enhanced compared to H_7^+ and H_{11}^+ production as is clearly seen in Fig. 1 in the H_{31}^+ case. This can be related to the shell effect predicted in the structure of ionic hydrogen clusters[16].

In Fig. 3, the absolute production cross sections of fragments of mass p (p=3 to n-2) resulting from the fragmentation of H_n^+ clusters (n=9, 13, 21, 25, and 31) colliding with helium at 60 keV/amu are displayed versus the normalized fragment mass p/n. The mass distributions of the various fragment yields are found to be quite similar. For p/n values smaller than 0.5, the distribution of the production cross section (σ) for H_{21}^+, H_{25}^+, and H_{31}^+ can be parametrized by a common power law ($(p/n)^{-2.6}$) (see Fig. 4). Therefore, the mass distribution of intermediate mass fragments exhibits a finite size scaling of the form expected in many theories of critical phenomena[1]. The small discrepancy observed for n=9 and 13 is due to the choice of the scaling parameter which is not well suited when the number of p is small. Using data on proton induced reactions at high energies Campi[1a] has studied the production of intermediate mass fragments. The present cluster distributions look similar to the ones obtained for high-energy proton (a few GeV) induced fragmentation of nuclei which have been connected to the existence of a nuclear multifragmentation process. In the same way, the distribution corresponding to low-energy collision (Fig. 2b) where mainly evaporation is involved can be compared to nuclear evaporation reaction resulting from collision with protons of about a few hundred MeV energy[1a].

For a given number of incident clusters H_n^+, the number of charged fragments (from p=3 to n-2) is larger than the number of dissociated clusters (except for n=9). The ratio of the number of charged fragments over the number of dissociated clusters is found to be 0.73±0.07, 1.09 ± 0.11 , 1.32 ± 0.13, 1.3 ± 0.13, and 1.28 ± 0.13 in the H_9^+, H_{13}^+, H_{21}^+, H_{25}^+ and H_{31}^+ cases, respectively (we note that the fragments H_2^+ and H^+ have not been taken into account in these calculations). This shows that a great number of the events correspond to a simultaneous production of at least two charged fragments after ionization of the incident cluster.

In conclusion, an important production of fragment ions with a size of $p/n \leq 0.5$ is observed. The molecular-evaporation process cannot explain the whole mass distribution of the charged fragments. Moreover, the distributions for $p/n \leq 0.5$ are found to have scaling properties and to follow a power law $A^{-\tau}$, where A is the normalized fragment mass and τ an exponent close to 2.6. The present cluster distributions appear to have a similar shape to the ones corresponding to nucleus fragmentation induced by a few GeV proton impact. The importance of the charged-fragment production and the behaviour of their inclusive mass distributions let expect a cluster multifragmentation resulting from the collision in this energy range. Exclusive measurements should uncover cluster multifragmentation channels.

The authors wish to thank M. Spighel, X. Campi and C. Bréchignac for helpfull discussions and R. Genre, J. Martin, R. Filliol, J. P. Lopez, H. Mathez for their expert technical assistance.

REFERENCES

1 - a) X. Campi, Proceedings of the 107 course of the Int. School of Physics "Enrico Fermi" on the Chemical Physics of atomic and molecular clusters, (Varenna, 28 Juin - 7 Juillet,1988, Italie); X. Campi, Nucl. Phys. A495, 259c (1989).

b) D. H. E. Gross, A. Ecker, and A.R. DeAngelis, Nucl. Phys. A545, 187 (1992);

D. H. E. Gross, A.R. DeAngelis, H. R. Jaqaman, Pan Jicai, and R. Heck, Phys. Rev. Lett. 68, 146 (1992).

c) Z. Cheng and S. Redner, Phys. Rev. Lett. 60, 2450 (1988).

2 - see a review on this subject by T. D. Märk, Int. J. Mass. Spectrom. Ion. Phys.,79, 1 (1987).

3 - M. E. Geusic, R. R. Freedman, J. Chem. Phys. 88, 163 (1988) and ref. therein.

4 - M. F. Jarrold, J. E. Bower, and J. S. Kraus, J. Chem. Phys. 86, 3876 (1987) and ref. therein.

5 - A. Van Lumig and J. Reuss, Int. J. Mass. Spectrom. Ion Phys., 27, 197 (1978).

6 - D. Kreisle, O. Echt, M. Knapp, E. Recknagel, K. Leiter, T. D. Märk, J. J. Säenz, and J. M. Soler, Phys. Rev. Lett. 56, 1551 (1986).

7 - N. G. Gotts and A. J. Stace, Phys. Rev. Lett. 66, 21 (1991).

8 - C. Bréchignac, Ph. Cahuzac, F. Carlier, and M. de Frutos, Phys. Rev. Lett. 64, 2893 (1990).

9 - I. Katakuse, H. Ito, and T. Ichihara, Int. J. Mass. Spectrom. Ion. Processes 97,47 (1990).

10 - P. Hvelplund, L. H. Andersen, H. K. Haugen, and J. Lindhard, D. C. Lorents, R. Malhotra, and R. Ruoff, Phys. Rev. Lett. 69, 1915, (1992).

11 - I. Ben-Itzhak, K. D. Carnes, S. G. Ginther, D. T. Johnson, P. J. Norris, and O. L. Weaver, Phys. Rev. A47, 3748, (1993).

12 - E. P. Kanter, P.J. Cooney, D.S. Gemmell, K.O. Groeneveld, W.J. Pietsch, A.J. Ratkowski, Z. Vager, and B.J. Zabransky, Phys. Rev. A20, 834 (1979).

13 - M.J. Gaillard, A. Schempp, H.O. Moser, H Deitinghoff, R. Genre, G. Hadinger, A.Kipper, J. Madlung, and J. Martin, Z. Phys. D 26, S 347 (1993).

14 - B. Farizon, M. Farizon, M. J. Gaillard, E. Gerlic, S. Ouaskit, Nucl. Instrum. and

Meth.in Phys. Res. B88, 86 (1994).

S. Ouaskit, B. Farizon, M. Farizon, M. J. Gaillard, and E. Gerlic, Phys. Rev A49, 1484 (1994).

15 - S. Ouaskit, B. Farizon - Mazuy, M. Farizon, M. J. Gaillard, E. Gerlic, and M. Stern, Phys. Rev A48, 1204 (1993).

16 - M. Farizon, H. Chermette, and B. Farizon-Mazuy, J. Chem. Phys. 96, 1325 (1992) and ref. therein.

FIGURE CAPTIONS

Figure 1: a) The mass spectrum of charged fragments from p=3 to 29 resulting from the collision of H_{31}^+ clusters with helium atoms at 60 keV/amu. The highest energy peak (to be multiplied by 6) corresponds to the detection of the transmitted H_{31}^+ clusters.

b) The same mass spectrum measured without gas target.

Figure 2: a - The production cross section of fragment ions H_p^+ with p=3 to 23 resulting from the fragmentation of H_{25}^+ colliding with helium (at 60 keV/amu) versus p.

b - The relative production cross section of fragment H_p^+ resulting from the fragmentation of H_{25}^+ in low-energy collision (5) (few eV/amu) with helium versus p.

Figure 3 : The production cross section of fragment ions H_p^+ with p=3 to n-2 resulting from the fragmentation of H_n^+ clusters colliding with helium at 60 keV/amu versus p/n, the normalized fragment mass.

Figure 4 : The production cross sections σ versus p/n in Log-Log scale (○ for n=9, 13 ; and ■ for n=21, 25, 31). The slope gives the exponent τ of the scaling law $(p/n)^{-\tau}$.

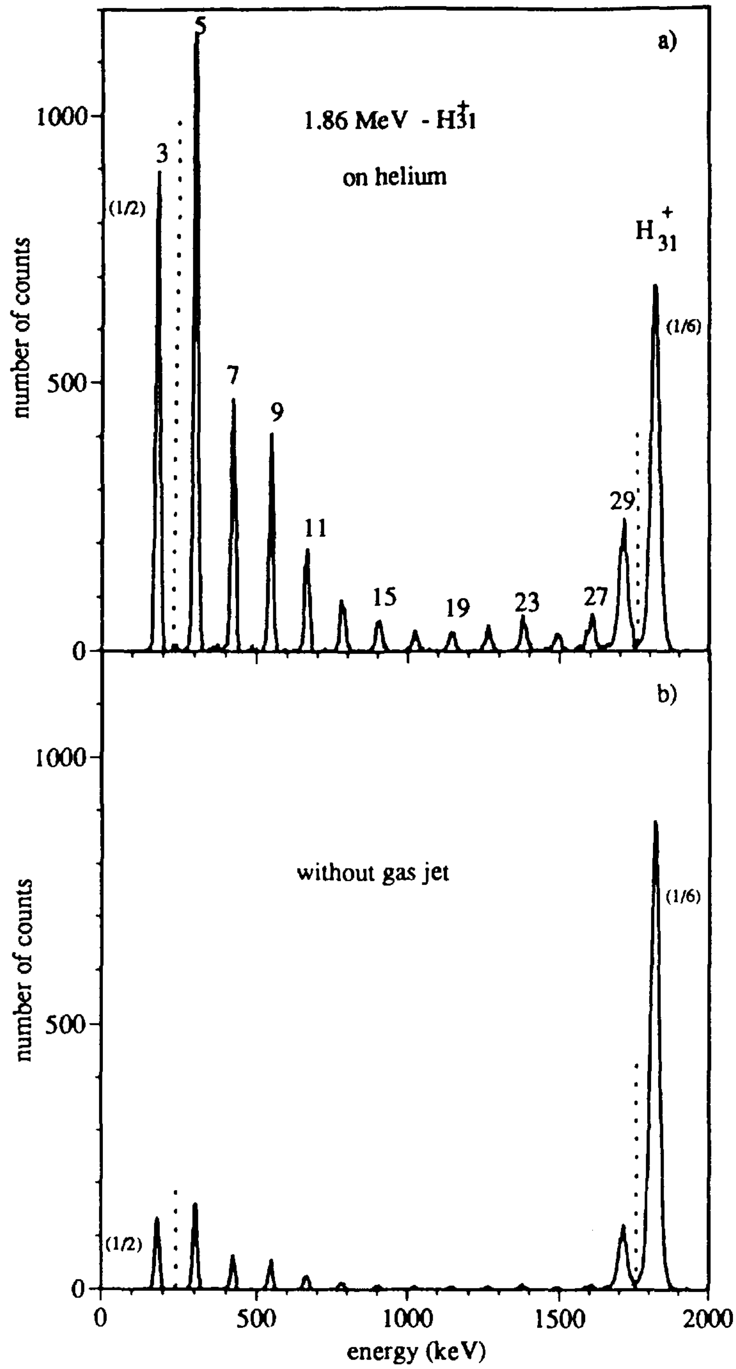

Figure 1 S. Ouaskit et al.

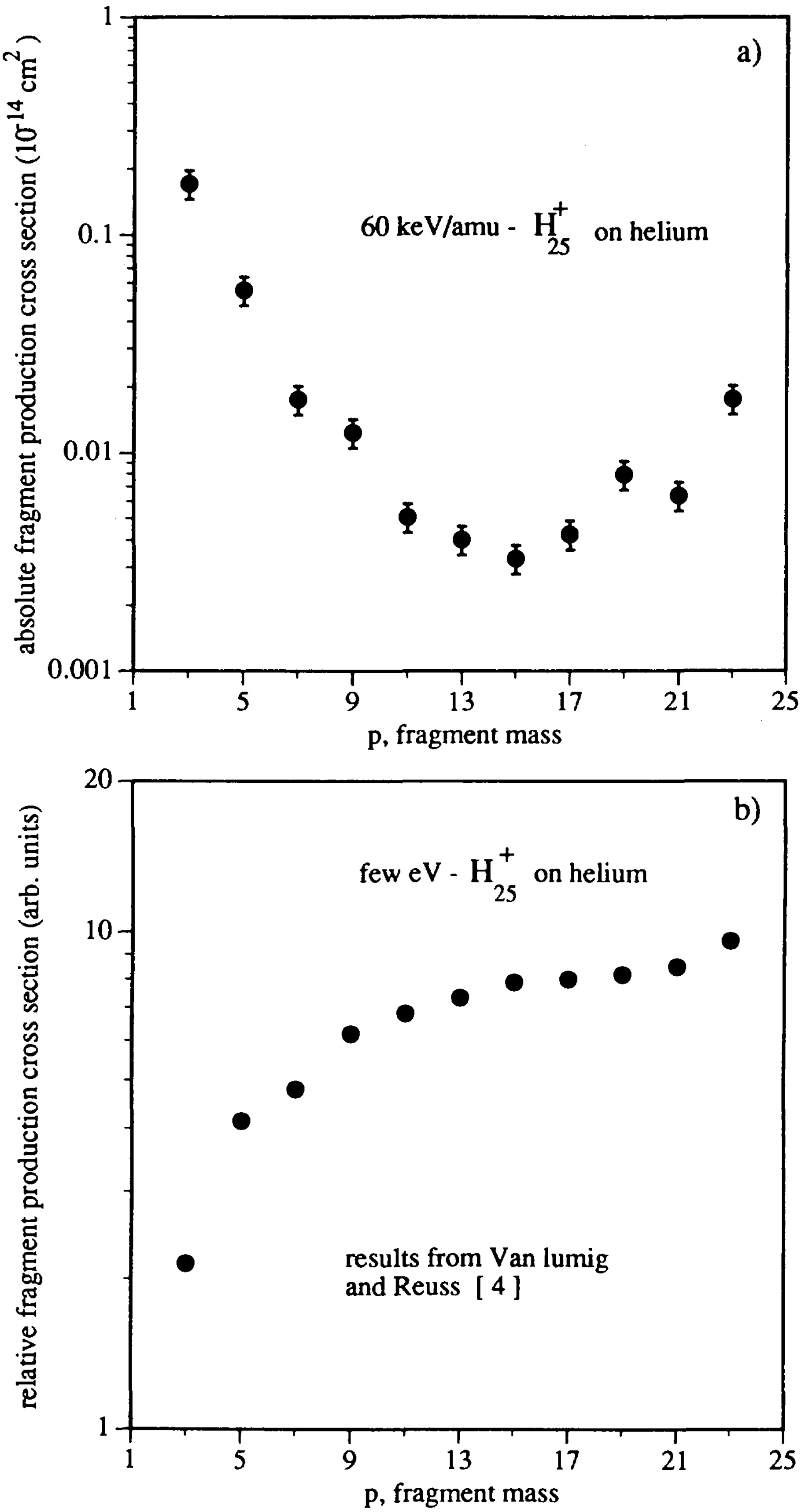

Figure 2 S. Ouaskit et al.

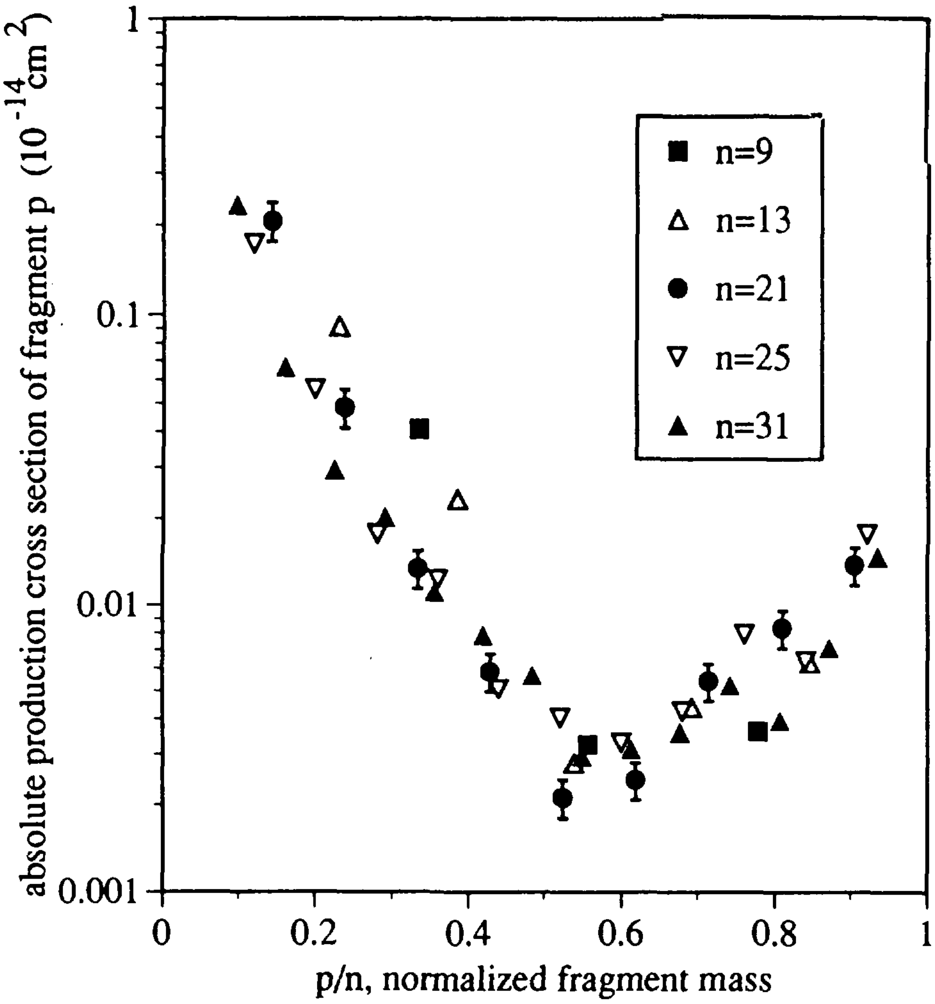

figure 3 S. Ouaskit et al.

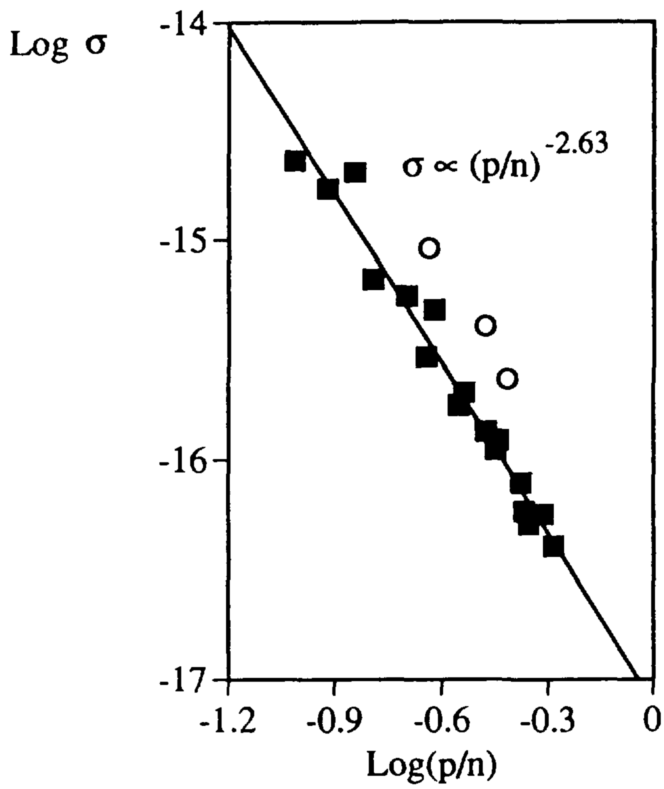

figure 4