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Hydrogen Cluster Ions

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Molecular weight distributions of hydrogen cluster ions produced by the expansion of refrigerated ionized gas in a supersonic nozzle are reported. Relatively narrow distributions with most probable masses up to a mass-to-charge ratio of 369 have been observed.

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The purpose of this paper is to report results of studies on the generation of hydrogen cluster ions by expansion of cooled hydrogen plasmas in supersonic nozzles. The objective in this work was the development of a technique for production of singly charged, high-molecular-weight hydrogen clusters with sufficiently narrow mass distributions to permit efficient acceleration in multiple-stage acceleration systems. The long-term goal is to develop an ion source that could produce an intense cluster-ion beam that could be used to heat plasmas for the ignition of controlled thermonuclear reactions.

The importance of energetic hydrogen clusters for fueling thermonuclear plasmas was recognized by Henkes and co-workers.[1,2] Pioneering studies on the generation of neutral cluster beams have been carried out by this group. Efforts were also made to ionize neutral clusters and accelerate them with a single-stage terminal. This work produced relatively broad mass distributions with ions having most probable mass distributions at mass-to-charge ratios of roughly 10 000 or less. For example, with a cluster size peaking at mass 13 000 the full width at half maximum extended from approximately mass 6000 to mass 26 000 (Ref. 1); with clusters peaking at mass 1000 the half maximum points were found at about 600 and 2000, respectively.[2] An increased width of cluster-ion beam mass and charge distributions is expected to result from almost any efficient neutral-cluster ionization technique. The deposition of ionization energy in the very weakly bonded neutral cluster is accompanied by energy transfer which would tend to partially destroy the cluster. Such effects would be greatest for relatively small clusters. For this reason it seemed reasonable to explore the process of the growth of cluster ions around ionic nuclei introduced into the cooled gas undergoing adiabatic expansion in a supersonic nozzle.

The initial production of ions in a high-pres-

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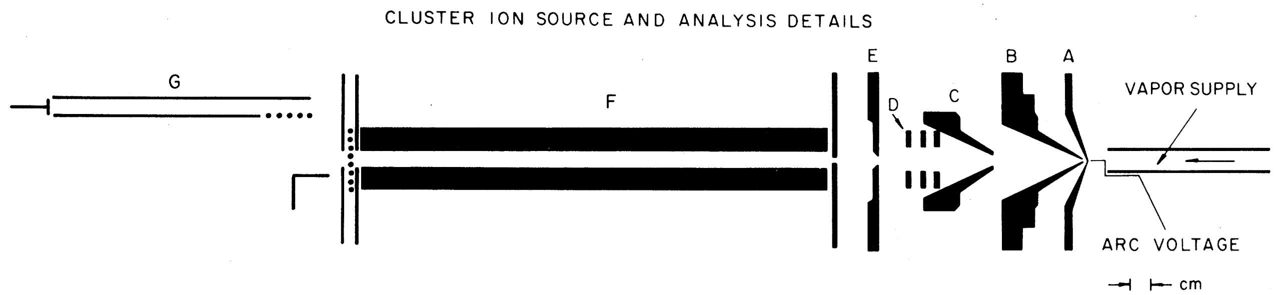

FIG. 1. Schematic of apparatus used to generate hydrogen cluster ions. Refrigerated hydrogen in which a corona discharge was generated was expanded in the nozzel in A and cluster ions passed through the pumped region between A and the skimmer B to the differentially pumped region between B and C. C and D were extraction and focusing electrodes that guided ions into the quadrupole mass analyzer F. Ion detection took place at the secondary electron multiplier G.

sure gas, cooled to a low temperature, serves to provide a means for the moderation of excitation energy deposited in the atomic or molecular system during the ionization process by collisions with cold neutral gas, and also avoids the problem of production of multiply charged ions. Hiroaka and Kebarle[3] have investigated the thermodynamics of the formation of $H\sigma^+$ with n as high as 11 or 13, in a cooled high-pressure mass-spectrometer ion source. Searcy and Fenn[4] have made small clusters of water containing up to 28 water molecules and a single proton using a corona discharge to produce ions in gas at room temperature which was subsequently expanded in a supersonic nozzle. We have carried out similar experiments with water in nitrogen carrier gas and have produced water cluster ions containing more than 4000 water molecules. A high-molecular-weight mass analysis and detection facility capable of distinguishing between singly and multiply charged ions[5] was used to identify argon and nitrogen clusters prepared by expansion of these elements in helium carrier gas at liquid-nitrogen temperatures. In these experiments only singly charged ions were observed. The results of these experiments in which clusters with molecular weights of between 1000 and 100 000 were produced will be described in a separate publication. Experience with high-molecular-weight cluster ions produced in a liquid-nitrogen cooled source was essential in the development of an ion source capable of operating at temperatures down to liquid-hydrogen temperatures.

A schematic of the apparatus used in these experiments is shown in Fig. 1. The ion source consisted of a pointed rhenium ribbon which was the origin of a corona discharge to the walls of a conical nozzle labeled A in the figure. The nozzle had a 0.0075-cm throat followed by a 0.3-cm-long conical divergent section (total divergent angle ~ 118°). A Gentry–Geise-type skimmer[6] with an 0.05-cm opening was located 0.5 cm from the nozzle exit. The point of the rhenium ribbon was ~0.5 cm from the nozzle entrance with no field-free region defined by grids between the ribbon and nozzle. Ionized gas was expanded into the differentially pumped region between A and B, the skimmer of the supersonic beam system; ions were extracted with lens C, and mass analyzed with a quadrupole mass filter before detection with a secondary-electron multiplier. The nozzle and region in which the electrical discharge was generated was cooled with a closed-cycle helium refrigeration system which was cap-

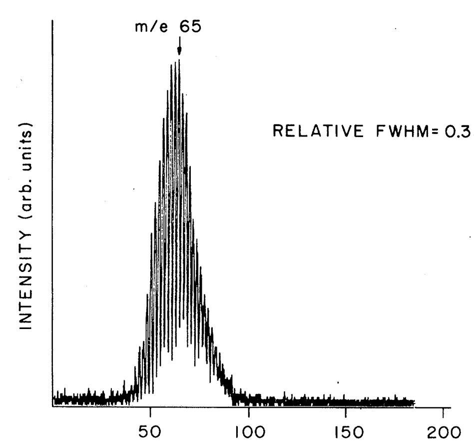

FIG. 2. Mass spectrum of cluster ions produced in a source refrigerated to 40 K with a source gas pressure of 105 Torr. The full width at half maximum of this distribution is approximately 22 mass units or 0.35 of the most probable mass in the distribution.

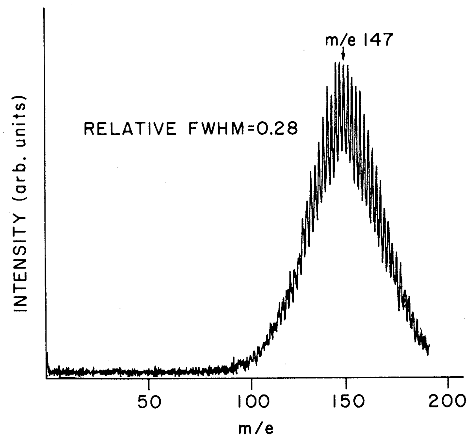

FIG. 3. Mass spectrum of cluster ions produced at 35 K with a source pressure of 125 Torr.

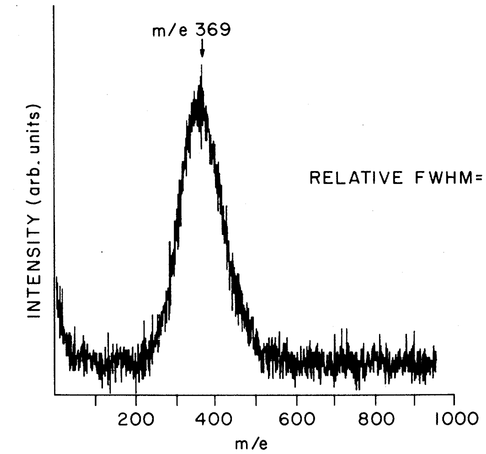

FIG. 4. Mass spectrum of cluster ions produced at 18 K with a source gas pressure at 110 Torr. The peak at very low mass is signal observed with zero quadrupole ramp voltage, the quadrupole "starting signal," and not low-molecular-weight hydrogen cluster ions.

able of cooling the gas and the ion source to temperatures as low as 10 K.

The major problem in the production of hydrogen clusters by the technique of expansion of ionized gas in a supersonic nozzle apparatus is associated with the low temperatures required for cluster growth and the fact that all impurities except helium can plug the nozzle or build up insulating films which render the ion source inoperative.

The results obtained in this work are presented in Figs. 2–4. Distributions of cluster ions with most probable masses ranging from 65 to 369 are shown. In Figs. 2 and 3 a mass separation of two mass units is resolved showing the buildup of hydrogen clusters by addition of molecular hydrogen. The absence of large yields of low-molecular-weight ions, i.e., mass 1 or 3, in the observed mass distributions is an important observation that distinguishes the ion product generated by our technique from reported mass distributions[4] obtained by ionization of neutral clusters. However the most important distinction is the relatively narrow full width at half maximum which is of the order of 123 mass units at mass 369, a full width at half maximum reflected by $\Delta m/m$ of 0.33 compared to the value of roughly 1.5 found in the studies of Henkes and co-workers.

The identification of these cluster ions as molecular systems that consist exclusively of hydrogen molecules and a proton was based on observations of mass distributions as a function of temperature and pressure of gas in the ion source. When trace impurities were present they could easily be observed as components of the mass spectrum with, for example, masses 19, 21, and 23, etc., with minute amounts of water in the system. Reduction of hydrogen gas pressure or elevation of source temperature then produced an isolated mass 19 with no ions below it until mass 3 or 5. Pure hydrogen mass distributions were first observed with a peak at mass 3 which could be smoothly and continuously shifted up to the masses shown in the figures. Relatively large levels of impurities, aside from helium, could not remain as gases in the ion source at the temperatures of most of these experiments.

In conclusion, the formation of hydrogen cluster ions with relatively narrow mass distributions, formed by the expansion of ionized gas in a supersonic nozzle, has been demonstrated. These ions have sufficient stability for mass analysis in a quadrupole mass filter and subsequent detection.

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