INITIAL RESULTS OF POSITRON IONIZATION MASS SPECTROMETRY

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ABSTRACT

The use of monoenergetic positrons for the ionization of organic molecules in the gas phase is described. The ionic products are analyzed with a time-of-flight mass spectrometer and detected to produce a mass spectrum. The ionization mechanisms which can be studied in this way include positron impact at energies above the ionization limit of the target molecules, positronium formation in the "Ore gap" energy range, and positron attachment at energies less than 1 eV. The technique of positron ionization mass spectrometry (PIMS)[1] may have analytical utility in that chemical selectivity is observed for one or more of these processes.

INTRODUCTION

Theoretical predictions by Schrader[2,3] based on molecular orbital calculations indicate that certain organic molecules will exhibit a positive affinity for low-energy (<1 eV) positrons. Higher-energy processes such as positronium formation and positron impact ionization are expected to involve a nearly vertical Franck-Condon transition from the ground state of the neutral molecule to the ionic state, resulting in a highly excited ionic state and fragmentation of the molecular ion.

Work by Surko, et al.[4] indicated that the trapping time of positrons in a Penning trap was strongly correlated with the size (molecular weight) of simple alkanes from butane to hexadecane at a pressure of 10^{-7} to 10^{-6} Torr. Later experiments[5] involving time-of-flight mass analysis indicated that low-energy positron resonances were taking place, along with other processes.

We have designed and constructed a high-quality mass spectrometer to study these processes more systematically and to demonstrate the analytical utility of positron-induced ionization.

EXPERIMENTAL

The positron source for this work is based on the Oak Ridge Electron Linear Accelerator[6-8] and makes use of excess gamma-ray bremsstrahlung which induces pair production in tungsten metal plates. The tungsten also serves as a moderator for the positrons which are re-emitted at approximately 2.5 eV with a narrow energy spread. The positrons are then accelerated to 3000 eV and transported to an experimental room 11 meters away by means of solenoids. The 3000 eV positrons are

then electrostatically deflected by 60 degrees into the ion source chamber of the time-of-flight mass spectrometer (TOFMS).

In our initial studies[1], this 3000 eV beam was allowed to traverse the ionization volume of the TOFMS ion source and strike a tungsten moderator operated in the reflection mode. Numerous spectra were obtained in this way which resulted from positron impact ionization by the 3000 eV beam.

A change in geometry allowed us to eliminate this impact ionization background and concentrate on the low-energy attachment process. This was accomplished by allowing the 3000 eV beam to strike a 1000 Angstrom thick tungsten film which acts as a transmission moderator[9]. A fraction of the positrons (10-30%) emerge on the other side of this film at 2.5 eV. These are then injected into a miniature Penning trap through a molybdenum grid which allowed us to select the energy of the positrons in the trap in the range of 0.1 to 3 eV.

The positron trapping ion source consists of a metal block with two end grids biased +5 V and an axial magnetic field of 10-60 Gauss to confine the low-energy positrons. Ions which are formed in the trap are extracted after 10 to 30 microseconds of positron trapping and are accelerated into the TOFMS flight tube which is 1 meter long. The ions are detected with a channelplate and the anode signal acts as a stop signal for a bank of 8 time-to-digital converters operated in ripple fashion.

RESULTS AND DISCUSSION

Figure 1 shows the spectrum of toluene obtained with the trapping ion source, where the positron energy was estimated to be less than 1 eV. The spectrum consists of only one peak corresponding to the molecular ion (but possibly at m/z - M-1) and virtually no fragment ions. This is strong evidence for a "soft ionization" process in which the intermediate state (before annihilation) resembles the ionic state. The positron impact spectrum of toluene using 3000 eV positrons resembles the conventional electron impact spectrum in which fragment ions are seen at lower masses.

CONCLUSIONS

The initial results with our PIMS system are encouraging. We have successfully demonstrated positron impact ionization and have seen strong evidence for low-energy positron attachment. Future studies will center on measuring the crossections for a number of organic molecules as a function of positron energy.

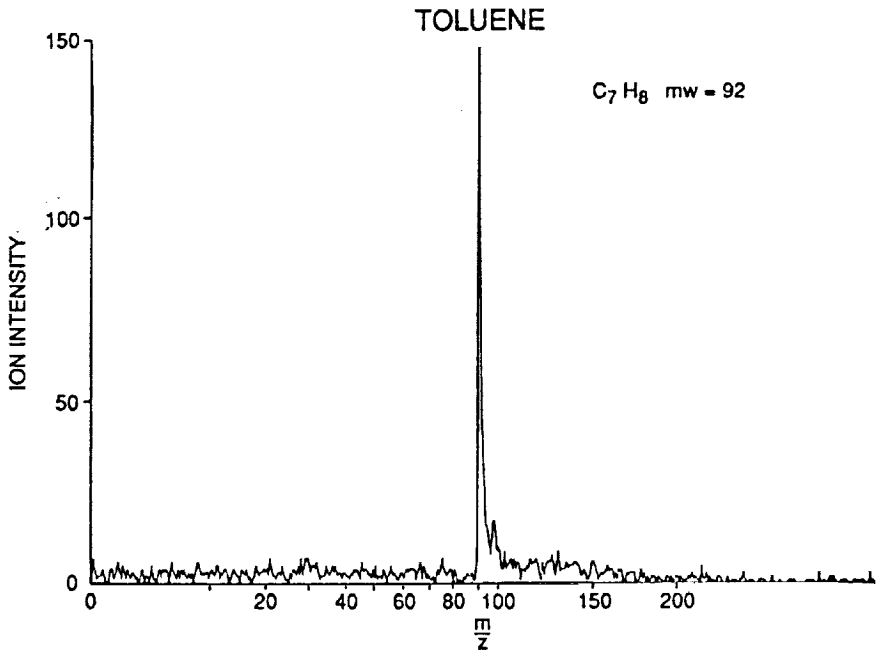

Figure 1. Low-energy positron ionization spectrum of toluene.

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