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**DESORPTION OF SECONDARY NEGATIVE IONS INDUCED BY FAST HEAVY INCIDENT
ATOMIC AND MOLECULAR IONS⁽¹⁾**

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RESUME

Des ions lourds atomiques et moléculaires ont été utilisés comme ions incidents dans des études de désorption d'ions secondaires. Des ions C^+ , C_2^+ , C_3^+ , Ar^+ , CH^+ , CH_3^+ , $C_3H_3^+$, $C_4H_9^+$, $C_7H_{15}^+$, CO^+ , O_2^+ , CO_2^+ , CF^+ , CF_3^+ , $C_3F_5^+$ and $C_4F_7^+$ ont été produits et accélérés dans la gamme d'énergie 600 keV - 3.9 MeV. Les ions moléculaires ont des rendements de désorption élevés. Ceci présente un intérêt pratique en spectrométrie de masse. Une interprétation de l'augmentation du rendement est présentée.

ABSTRACT

Fast heavy atomic and molecular ions have been used as incident ions in secondary ion desorption studies. A number of ions such as C^+ , C_2^+ , C_3^+ , Ar^+ , CH^+ , CH_3^+ , $C_3H_3^+$, $C_4H_9^+$, $C_7H_{15}^+$, CO^+ , O_2^+ , CO_2^+ , CF^+ , CF_3^+ , $C_3F_5^+$ and $C_4F_7^+$ have been produced and accelerated in the energy range 600 keV - 3.9 MeV. The molecular incident ions give rise to high measured secondary ion yields which is of practical interest in mass spectrometry. An interpretation of the enhancement in the yield with molecular incident ions is presented.

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Particle induced desorption of polyatomic ions from surfaces is of general theoretical interest, as well as having practical applications in the field of mass spectrometry. A large body of experimental data exist on the use of fast ($v > v_{\text{Bohr}}$) atomic incident ions in desorption studies (1). However, few reports exist on the use of fast molecular ions as the primary ions in desorption experiments. Brown et al. (2) and Thomas et al. (3) have used hydrogen cluster ions to measure the yields of H_2O and Cs^+ ions respectively (yield is defined as the number of the species of interest desorbed per incident ion). We recently reported on desorption experiments with the use of fast heavy molecular ions such as C^+ , C_2^+ , C_3^+ , Ar^+ , CH^+ , CH_3^+ , C_3H_3^+ , C_4H_9^+ , $\text{C}_7\text{H}_{15}^+$, CO^+ , O_2^+ , CO_2^+ , CF^+ , CF_3^+ , C_3F_5^+ and C_4F_7^+ , in the energy range 600 keV - 3.9 MeV (4,5,6,7).

The experimental set up is explained in detail elsewhere (7) and will not be discussed here in any detail. The ions are produced in an electron-ionisation ion source located at the high voltage terminal of a 4.5 MV Dynamitron accelerator. The accelerated molecular ions are unambiguously identified and reach the time-of-flight (TOF) mass spectrometer. The secondary ion yields are measured using conventional TOF techniques. The standard sample has been the amino acid valine (MW = 117) electrosprayed on a thick stainless steel plate. The results presented here are limited to negative secondary ions since the secondary electrons have been used to trigger the start pulse for the TOF measurements.

Figure 1 shows the negative secondary molecular ion yield of valine, Y , as a function of the velocity of the primary C^+ , C_2^+ , C_3^+ incident ions. From the relative secondary ion yields of valine, Y , as a result of impact with C^+ , C_2^+ and C_3^+ , it is apparent that non linearities exist ; the secondary ion yield of valine molecular ions for incident C_2^+ , $Y(\text{C}_2^+)$ is much more than twice that for atomic carbon, i.e., $Y(\text{C}_2^+) \gg 2.Y(\text{C}^+)$. Similarly $Y(\text{C}_3^+) \gg 3.Y(\text{C}^+)$.

We have empirically determined that $Y(\text{C}^+)$ for valine negative molecular ions scale with the square of the electronic stopping power, $\frac{dE}{dx}$ (6,7). Such a relationship would then predict for molecular ions ; $Y(\text{C}_2^+) \sim 4.Y(\text{C}^+)$ and $Y(\text{C}_3^+) \sim 9.Y(\text{C}^+)$. However, the experimental point show much higher ratios over a range of primary ion velocities, i.e., approximately $Y(\text{C}_2^+) \sim 8.Y(\text{C}^+)$ and $Y(\text{C}_3^+) \sim 18.Y(\text{C}^+)$. Similar results have been obtained with other incident ions such as C^+ and O^+ in comparison with CO^+ and O_2^+ (7). It thus seems that other processes may be present.

It has been shown experimentally that the electronic stopping power of an atom in a fast moving cluster in solids is higher in magnitude than the sum of the stopping powers of the atomic constituents in the same medium. The enhancement in $\frac{dE}{dx}$ per atom X in a cluster X_n was defined by Brandt (8) to be, $R = \frac{\frac{dE}{dx}(X_n)}{\sum \frac{dE}{dx}(X_i)}$. Brandt et al reported R values in the

range 1-1.5 for H_2^+ and H_3^+ in thin carbon films. Tape et al. (9) have obtained similar results for H_2^+ and O_2^- incident ions. The enhancement in $\frac{dE}{dx}$ was interpreted as being a "collective" effect due to the temporal and spatial correlation of the fast moving charges giving rise to an interference term in the total electronic stopping power of the cluster (8).

As discussed in detail in references (7), the desorption depth is comparable to the depth required for the incident molecular ions to separate completely and thus, any enhancements in $\frac{dE}{dx}$ would be reflected in desorption yields. The desorption data can be described by assuming a quadratic dependence on $\frac{dE}{dx}$ and using R values in the range 1.1-1.4 for all of the experimental points. This is comparable to the R values reported by Tape et al. (9) for O_2^- incident ions in the same velocity range as our experiment.

Preliminary results with another sample with different chemical properties, chlorophyll a (MW = 893) showed the same enhancements as valine ions. Hedin et al. (10) recently have reported desorption experiments with C_2^+ incident ions at a velocity of 0.49 cm/ns. Their valine negative ion data are in qualitative agreement with ours ; they obtained $R = 1.2 \pm 0.1$. However, for positive valine molecular ions they found $R = 1$ (the conclusions are however, rather sensitive to the choice of $\frac{dE}{dx}$). Since any enhancements in $\frac{dE}{dx}$ are expected to be due to the distant collisions (8) (and not the close collisions), such differences between the

positive and negative secondary ions might present a clue as to their different origins. More experimental results are needed to clarify this.

The implications of this work are two-fold. First, with the large incident molecular ions such as $C_7H_{15}^+$ and $C_4F_7^+$, high secondary ion yields have been measured (up to 60 times that with atomic carbon) which could be of practical interest in mass spectrometry. Second, negative ion desorption studies might be useful in the future for studying the interaction of cluster incident ions with solids.

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