Secondary Ion Emission from Metallic and Insulator Targets by Impact of Carbon Ions

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She H^+ secondary emission yield induced by the collision of carbon ions was measured for Fe, Ni, Pd, Au and Phenylalanine targets. Results are shown for secondary ion emission yield as a function of the charge state and the energy of the incident ion. The incident ion charge varies from 0 to $4f$ and the energy varies from 0.03 to 0.25 MeV/u. The results for the secondary emission yield of protonated phenylalanine ions, $(M + H)^+$; phenylalanine depleted of a carboxyl group $(M - COOH)^+$ and H^- from a phenylalanine target in the same range of energy of the incident carbon ions are also is presented. These results are compared with predictions of the thermalized ion explosion model of Seiberling et al[14] and with the model of Sigmund[5] using the normalized nuclear stopping power value taken from Ziegler[15].

I. Introduction

The secondary ion emission induced by collisions of accelerated ions has been used to study the mechanism of the surface-ion interaction. In particular the emission of H^+ ions as a function of the incident ion charge state can give informations about the two main mechanisms involved in the emission process. These mechanisms are usually named the potential and kinetic mechanism[1,2]. The potential mechanism can be described by the Coulomb interaction of the incident ion with the surface and may induce the electron emission before the ion impact[3]. The emission of these surface electrons from the target will leave atoms and molecules in excited states that may decay by the emission of secondary ions. An alternative description was given recently by Bitensky et al[4] assuming that the emission of H^+ ions is due to a Coulomb explosion of a small charged domain at the target surface. This charged domain for low energy projectiles (1 keV/u) results from Auger cascades in the close vicinity of the incident ion and for the MeV ions the ionization is due to the inelastic energy losses. In these description the H^+ emission yield will depend strongly on the charge of the incident ion. In the kinetic mechanism the emission of ionic fragments of the target constituents is a result of the energy deposited by the penetrating ion. For the present experiment we have used thin films as target and high energy projectile and in this case, as suggested by Sigmund[5], the energy deposited by the incident ions is primarily given by the elastic stopping power. Using neutral carbon beam we measured the H^+ emission yield to quantify the contribution due to kinetic mechanism which is independent of the incident ion charge. This contribution was subtracted from the H^+ emission yield obtained for the carbon ions with charge states ranging from 1^+ to 6^+.

There are early results for the relative yield of H^+ induced by low energy (0.45 Kev/u Ar iTl ions)[6-8] and high energy collisions (1.16 Mev/u Ne, Ar and Kr ions)[8-11] onto CsI, Phenylalanine, C foil and SiO_2 surfaces. In those experiments high charge states for the incident ion were used.

In the present work the secondary emission yields for H^+, H^- and Phenylalanine ions are measured from metallic targets of Fe, Ni, Pd, Au and Phenylalanine under the impact of C projectiles with energy in the 0.03 to 0.25 Mev/u range. The charge state of the incident projectile was varied from 0 up to $4f$. Measurements with low charge states of the incident carbon ion help the analysis of the secondary emission yield in terms of the contributions from the potential and kinetic mechanisms.

The target samples were chosen aiming to observe variations in the secondary emission yield of H^+ for materials with different hydrogen affinity. In the energy range of this work, the electronic stopping power is around one hundred times larger than the nuclear stopping power for the above ions and target samples. The data obtained suggest that, for H^+ and H-, the ejection process shows a strong influence of the nuclear stopping power.

Early publication by Hunt and Wien[12] for a valine sample and projectile velocities between 0.10 and 0.25 cm/ns have shown an influence of the nuclear stopping in the OH^- emission yield. Similar result was obtained by Salehpour et al[13] for the secondary ion yields of C-, O-, F^-, OH^- and CH^- for valine and gold targets bombarded with Xe^+, Kr^+ and Ar+ within the velocity range of 0.10 to 0.45 cm/ns. Results for the emission yield of gold negative ions from the *Au* target have also been shown. These results are consistent with the theory of Sigmund[5] assuming a convmechanism of collisional cascades. Since this theory is able to describe the emission yield for such different ion species as charged organic fragments as well metalic negative ions we will attempt to describe the kinetic contribution to the H^+ yield using the Sigmund theory[5]. Note however that the nuclear stopping power in our case is much smaller than the electronic stopping power.

The data obtained in the present experiments were also fitted to the parametrized form for yields given by Seiberling et al[14].

In summary, the present study attempts to contribute for the elucidation of some aspects of the mechanisms involved in the emission of secondary ions induced by the collisions of swift ions. However, as it is well understood, the process of ion desorption involves very complicated mechanisms of ion interactions with surface and bulk of solids and the complete understanding of the full process of secondary ion emission is far from to be achieved.

II. Experimental

The carbon beam was produced by a 4.0 MV Van de Graaff accelerator at Pontifícia Universidade Católica do Rio de Janeiro (PUC-Rio). The mass spectra measurements were performed using a standard Time of Flight (TOF) mass spectrometer shown schematically in Fig. 1. This spectrometer was fitted inside a chamber placed in a beam line 30° to the direction of the beain produced by the accelerator. An analyzing magnet allowed to select the ion beam driven into the reaction chamber according to the ion charge desired. To produce the different ion charge states a thin carbon film stripper was placed in the analyzing magnet entrance. Targets of Fe, *Ni*, Pd and Au were prepared by evaporation of these materials onto thin *Al* films mounted on 1.2 Phenylalanine targets were similarly prepared onto C films. The *Al* and C films used as backing were sufficiently thin to be traversed by the incident beam. The primary ion (PI) beam was counted by a surface barrier detector placed behind the target and furnishes the start signals for coincidence measurements. The secondary ions (SI) desorbed by impact of the PI were accelerated in the electric field between the biased target and a grounded grid, 80% transparency, placed parallel to the front surface of the target at a 4-5 mm distance. The acceleration bias used in the experiments was 3.5 kV, positive or negative according to the charge of the desorbed ion. After acceleration, the SI penetrate a field free flight path and are detected by a tandem microchannel plates detector placed at the end of the flight tube which produces the stop signals. Start and stop signals are fed into a Time to Digital Converter (TDC) that accepts up to 255 stop signals for each start signal with a time bin of 0.5 nsec and dead time of 22 nsec. The TOF spectra were stored in a personal computer (PC) and promptly converted to ion mass spectra. The secondary ion yield was determined as the ratio between the desorbed ions counted and the number of PI imping on the target and counted by the surface barrier detector. All measured yields are relative yields because the detection efficiency of the TOF mass spectrometer is not accurately known. The efficiency depends on several factors such as geometry, grid transparency, efficiency of the channel plates detector which is known to depend on the velocity and mass of the incident particle. Therefore the yields measured are compared relative to each other even considering that all measurements were performed using similar experimental conditions The samples of Fe, Ni, Pd, *Au* and Phenylalanine were investigated using carbon beains with charge states varying from 0 to 4^+ and energies in the 0.5 to 4.0 MeV range. We have measured the H ion for all the above targets and for all ion charges snd energies. SI of Phenylalanine and H^- were also measured in the Phenylalanine target.

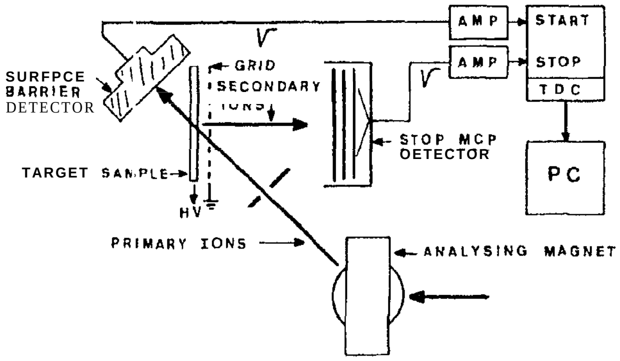

Figure 1: Schematic of the experimental arrangement

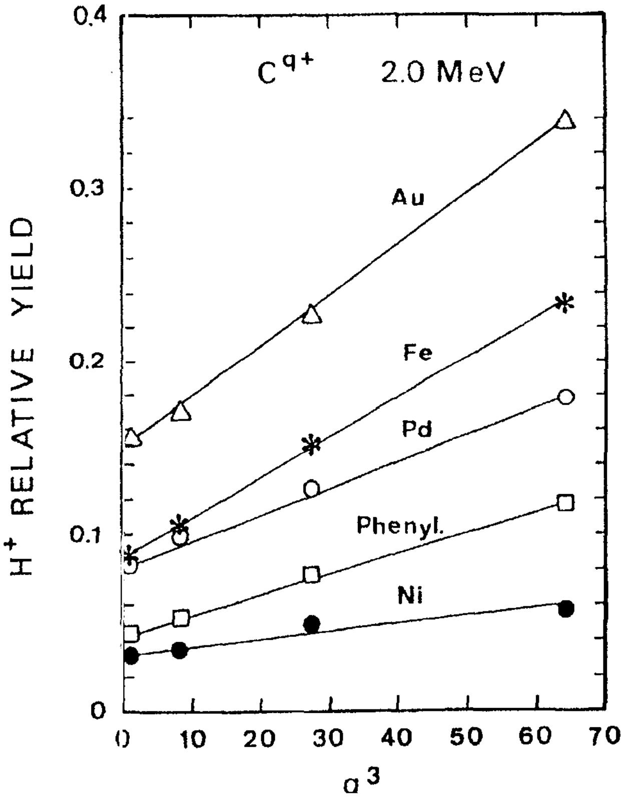

Figure 2: H^+ relative desorption yield as a function of the incident carbon ion charge. Results are shown for projectiles with 2.0 MeV energy and targets of Fe, *Ni*, Pd, *Au* and Phenylalanine. The lines are linear functions of the cubic of incident ion charge.

The pressure in the reaction chamber was always better than $10^{-4} Pa$.

III. Results and Discussion

The relative yield for the desorption of H^+ from target samples of Fe, Ni, *Pd*, Au and Phenylalanine induced by 2.0 MeV carbon projectiles as a function of the cube of the incident ion charge are given as an example in Fig. 2. It is evident the linear dependence of the H^+ yield on the cube of incident ion charge for all the cases shown. This behavior was is in agreement with early results from several authors[6-11].

In recent publication[1,5] it was suggested that the H^+ desorption yield may result from two different mechanisms namely the potential and the kinetic mechanism. The potential mechanism is strongly dependent of the incident ion charge state (Coulomb interaction) and weakly dependent of the energy of the incident ion resulting more likely in the desorption of hydrogen from the target surface.

Accordingly, one may write the following relation for H^+ desorption yield:

$$Y(H^+) = Y_0 + \alpha q^3 \,, \qquad (1)$$

where Y_0 is the charge independent contributions and depends on the energy deposited in the material by collisions of the incident ions, q is the incident ions charge and α is a coefficient which depends on the characteristics of the target and slightly on the energy of the incident ion[5]. Using the above relation we extracted the contribution to H^+ desorptions yield due to the kinetic mechanism. The projectile charge independent yield Y_0 for targets of Fe and Au are shown as functions of the incident ion energy in Fig. 3 and indicated by a C° together with the total H^+ yield for C^{3+} beam. It is also shown the relative Hf yield for a Phenylalanine target obtained with a neutral and double charged carbon beam. One may observe that each pair of curves follows similar trends whatever the charge of the incident ion. For a given target sample the data set for the different ion charge states are practically parallel following the trend of the yield obtained for the neutral carbon beam.

This fact reinforces the above assertion that the charge dependent contributions to the H^+ desorption yield, αq^3, is a slowly dependent function of the incident ion energy in this energy range. The curves are the normalized nuclear stopping power values for the different elements taken from Ziegler[15]. The agreement between these curves and the experimental points suggests that the incident ion charge independent H^+ yield contribution can be described by the elastic collision model proposed by Sigmund[5]. This dependence of secondary emission yield on the nuclear stopping power was also observed by Hunt and Wien[12] for OH^- fragment ions from valine targets and for projectile velocities around 0.1 cm/ns where the nuclear and electronic stopping powers are of the same order of magnitude. Similar results were obtained by Salehpour et al[13] for C^-, O^-, OH^-, CH^-, and Au^-, desorbed from gold and valine samples bombarded with Xe^+,Kr^+ and Ar^+ ions in the energy range of 0.40 to 3.0 MeV similar to the ion energies of the present experiments.

One may further note that the extension 02 will cross the vertical axis corresponding to q = 0. This crossing point will furnish the H^+ desorption yield Y_0 for a neutral carbon beam with of 2 MeV of energy. The values thus obtained can be compared with the corresponding values in Fig. 3. Additionally, the angular coefficients of the straight lines in Fig. 2 correspond to the parameter α described before. In a recent publication[2] a study of the variation of the a parameter with the energy of the incident ion was undertaken and the results show a very weak dependence of the projectile energy for a wide range of energies. The above analysis allows the quantification of the two contributions assuming that the Y_0 contribution results from the elastic stopping power of the projectile in the target, the kinetic mechanism, and that the incident ion charge-dependent contribution is given by aq^3. The Y_0 dependence on the nuclear stopping power is in accordance with the model proposed by Sigmund[5] and the third power dependence of the incident ion charge is supported by the recent models of Silveira et al[3] and Bitensky et al[4].

From the results for Au taken from Fig. 3 we inferred that the contribution due to the kinetic mechanism is around fifty percent in the case of the H^+ emission yield for 2 MeV C^{4+} ions and will decrease to fifteen percent for 2 MeV C^{6+} ions. For the cases were the charge state of the incident ions can reach very high values, the contribution due to the kinetic mechanism can be shaded by the strong potential contribution.

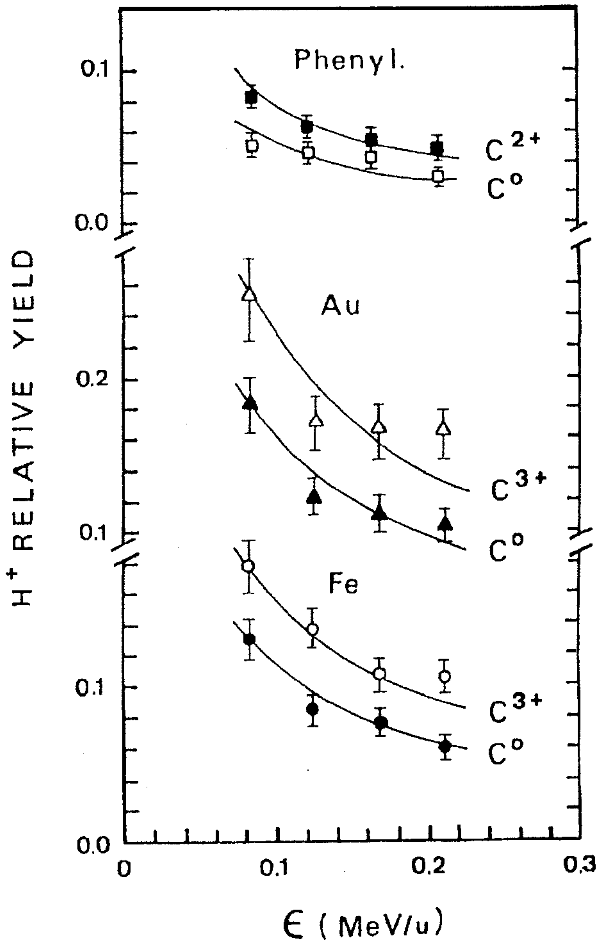

Figure 3: H^+ relative desorption yield as function of the incident carbon ion energy. Results are for Fe, Au aPhenylalanine targets and charges of the incident ion as indicated. The lines were obtained by fitting the normalized nuclear stopping power taken from Ziegler[15].

In Fig. 4 we show the protonated Phenylalanine ion, $(M+H)^+$, Phenylalanine depleted of a carboxyl group; $(M-COOH)^+$, H^+ and H^- as a function the energy of the incident C^{2+} ion impinging on tcan be seen the data for the relative desorption yields for the different desorbed ion are well fitted by the parameterized equation taken from Seiberling et al[4]using the equilibrated charge of the ion as given by Heckman et al[16]. The values of the fitting parameters A and B are shown in the figure.

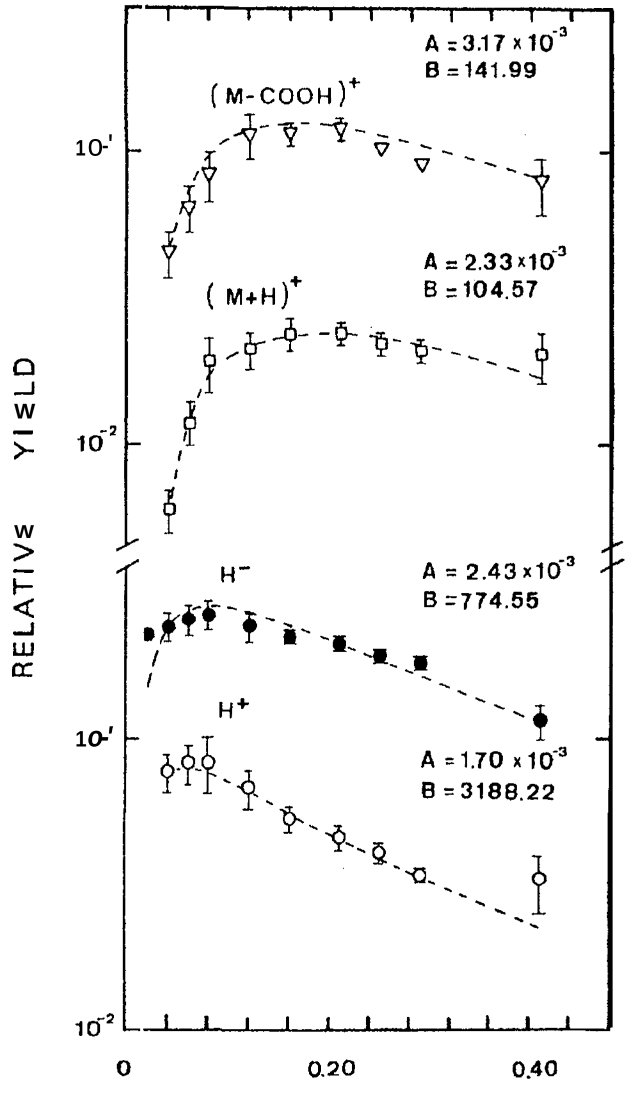

Figure 4: Relative desorption yield for H^+; $H-$;protonated Phenylalanine ions, $(M+H)^+$; and Phenylalanine molecules depleted of a carboxyl group, (M – $COOH)^+$ as a function of ϵ, where ϵ is the energy per nucleon of the incident double charged carbon ion. The full line were ajusted using a parametrized form for the thermalized ion explosion model taken from Seiberling et al[15].

In this model the ejection of secondary ions is related to the number of ionizations per unit of lenght caused directly by the incident ion rather than the electronic or nuclear stopping power. However the validity of this model is difficult to be proved directly because the primary ionization rate is a difficult parameter to measure. For the heavy desorbed ions the fitting constants of the model are consistent with values obtained by Seiberling et al[14] for heavy secondary ions desorbed from their target samples. The energy per nucleon of the projectile used in both cases are within the same range but in the case of H^+ and H^- desorption yields, the values of the fitting constants are quite different from values obtained for the heavy ions. This fact may give further indication that the hydrogen and heavy ions emission are the result of different mechanisms of the ion-solid interaction or are originated from different region of the target.

We further point out that the thermalized explosion model proposed by Seiberling et al[14] uses the primary ionization rate to describe the temperature and radius of the spike left by the penetrating ion, as suggested by Fleischer et al[17]. In the case for which the spike radius expands or contracts according to the increase or decrease of the primary ionization rate, $\frac{dJ}{dx}$, and the spike temperature is determined only by physical and chemical properties of the for the secondary ion emission yield is obtained:

$$Y = (dJ/dx)^4 \ , \qquad (2)$$

with the primary ionization rate $\frac{dJ}{dx} = \frac{AZ_{eq}^2 lnB\epsilon}{\epsilon}$ obtained in the Born approximation where Z_{eq} is the equilibrated charge[7], ϵ is the incident ion energy per mass unit and A and B are parameters dependent on the incident ion and target material.

For the case of H^+ secondary emission yield as a function of the incident ion energy the fitting of the above parametrized form, in the incident ion energy range of the present experiment, is almost indistinguishable from the nuclear stopping power fitting with values taken from Ziegler[15]. This result is rather unexpected since the unscreened nuclear stopping expression shows an energy dependence similar to the above $\frac{dJ}{dx}$ expression, although without the adjusting parameter. In addition the normalized stopping power is fitted directly while the above relation requires the fourth power of the primary ionization rate. The coincidence found in describing the general trends for H^+ desorption yield as a function of the energy of the incident ion using Seibeling model and by the normalized nuclear stopping power was not found for the (M + H)f and (M – $COOH)^+$ ions desorbed from the Phenylalanine target. In this case the secondary emission is well fitted to the above parametrized form and poorly fitted to normalized nuclear stopping power values showing a dependence on the cubic of the total stopping power[18].

We have restricted the comparison of the results obtained in the present experiments to the models of Sigmund[5] and Seiberling et al[14]. However, we will briefly comment on the experimental results within the framework other models frequently used to describe desorption process. The thermal spike model describes the emission of material as the activation and ejection from the surface due to a local energy density. The local energy resulting from a fraction of the electronic energy deposited uses the electronic stopping power of the incident ion as a fitting parameter to ineasured yields. Accordingly the emission yield scales with the square fraction of the energy deposited $(Y\alpha f^2(dE/dx)^2)$ [19]. The multihit rnodel proposed by Hedin et al[20] suggests that certain number of eletrons hit a molecule to cause the secondary ion einission. This model scales the yield for tlie fourth power of the stopping power for small dE/dx. For intermediate values of the stopping power the yield has a more complex dependence on dE/dx and varies nearly linearly for large values of dE/dx. The predictions of these models can fit poorly to the present experimental values.

Fenyö et al[21] described the secondary ernission process as a result of the excitation of molecules in a cylindrical region around the track produced by the incident ion. In this highly excited region the electronic energy transfered into atomic motion can be described by several mechanisms such as Coulomb repulsion between ionized atoms, repulsive decay of excited and ionized molecules and molecular expansion produced by secondary electrons. These processes produce a local expansion of the solid inducing the secondary ions emission. Using this analytical approximation the secondary emission yield can be calculated numerically for finite thickness samples. In this case the yields show a linear dependence on the effective stopping power. For the incident ion target combination of the present experiments the electronic and total st therefore the models described can not describe the results for the H^+ and H^- desorption yield. The results for the desorption of (M + H)+ and (M – $COOH)^+$ from the Phenylalanine target can be partially fitted to the lower values of the incident ion and shows an increasing deviation for increasing energies of the incident ion using the model proposed by Hedin. The yield is poorly fitted to the stopping power values as suggested by Fenyo et al[21].

IV. Concluding Remarks

Results were presented for the H^+ emission yield induced by carbon beams and discussed considering the existence of a potencial and a kinetic mechanism for desorption. The contributions to the yield due to the potential mechanism shows a third power of the incident ion charge dependence and indicate to be target surface emissions. This strong dependence on the ion charge for the H^+ emission yield was earlier verified by several authors[6–11] and described recently by Bitensky et al[4] and da Silveira et al[3]. The contribution due to the kinetic mechanism depends weakly on the energy of the incident ion within the energy range of the present experiments. The low charge states used in the experiments allowed to separate the contributions of the two mechanisms involved in the H^+ desorption yield as function of the incident ion energy. The observed behavior is well described by the Sigmund's model[5]. On the other hand, the desorption of (M + $H)^+$ and (M – $COOH)^+$ ions from a Phenylalanine target are well fitted to the formula of Seiberling et al[14] although the results for H^+ and H^- for the target are only fairly fitted to this formula.

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