

CARBON ACCUMULATION ON TARGET SURFACES IRRADIATED BY PROTONS*

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Summary

We show that the observed beam spot consists of accumulated carbon. The accumulation is the same on the front and back surface of a thin target, and is proportional to the bombardment time at current densities greater than $\approx 2\mu\text{A}/\text{cm}^2$. In our energy range of investigation (100-300 keV), the accumulation rate is independent of energy, but does vary with the temperature of the target. For targets at $\approx 15^\circ\text{C}$, the accumulation rate for an oil diffusion pumped target chamber is $\approx 2\mu\text{g}/\text{cm}^2\text{-hr}$ or $2\text{\AA}/\text{min}$, which corresponds to a retention of one carbon atom per incident proton. Cooling a shield surrounding the target, or heating the target to 150°C , reduces this accumulation rate by an order of magnitude.

Introduction

The buildup of a foreign substance on the target surface is a considerable nuisance for experiments where the true energy of a projectile incident on the target needs to be known. We hope to shed some light on the nature of this phenomenon and describe a simple and practical way of measuring the amount of contamination on the target surface, as well as methods of preventing such contamination.

Experimental Procedure

The method employed in determining the accumulation rate for thin ($\approx 20\mu\text{g}/\text{cm}^2$) and thick (0.02 mm) aluminum targets is shown in Fig. 1. A brass shield, which is thermally and electrically isolated, surrounds the target, and serves as an electron suppressor and cold shield. For the thick targets, the holder is thermally and electrically isolated and is connected directly to the current integrator. A surface barrier solid state particle detector and a Si(Li) x-ray detector viewed the target. Their counts were recorded for a preset integrated dose. The target chamber was evacuated by a 4" oil diffusion pump (using Dow Corning 705 pump oil) or a 280 l/sec turbo-molecular pump. The pressure in the target chamber was typically 10^{-6} Torr.

With this experimental technique, we monitored the elemental composition of the accumulated material. We observed some characteristic x rays of silicon. Their yield did not change with time and hence was taken as a constant background equivalent to a target thickness of $\sim 0.2\mu\text{g}/\text{cm}^2$. There was no indication of any other high atomic number elements in the accumulated material. In fact, large angle Rutherford scattering measurements indicate that the only deposited element which changed with bombarding time was carbon.

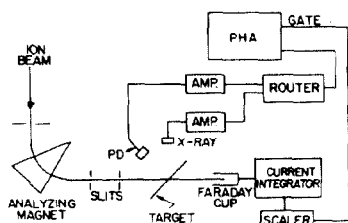


Fig. 1. Schematic of the experimental arrangement.

Direct measurements of the carbon accumulation rate on aluminum can be made by large angle (135° in our case) Rutherford scattering at 300 keV and higher incident energies. Carbon-scattered protons then have a mean energy which can be differentiated by a solid state particle detector from aluminum scattered protons. Furthermore, due to the energy loss suffered by the proton in the aluminum target, the protons scattered from the back surface carbon layer distinguish themselves, in energy, from those scattered by the front surface carbon layer. The accumulated carbon layer can be measured at any given moment by measuring the yield of carbon scattered protons relative to the aluminum scattered protons. Using Rutherford scattering yield equation, we obtain

$$X_C = X_{A\ell}^K \frac{N_C}{N_{A\ell}} \quad (1)$$

where X_C is the carbon layer thickness, X_{Al} is the thickness of the aluminum foil, both in $\mu\text{g}/\text{cm}^2$, K is a constant containing Rutherford scattering cross sections, and N_C and N_{Al} are scattering yields per incident ion from carbon and aluminum respectively. The accumulation rate, $\dot{X}$, is then determined by the time rate of change of Eq. (1) during continuous bombardment. This measurement is only possible at the maximum energy of our accelerator.

At lower incident energies we can determine the accumulation rate by measuring the change in aluminum K x-ray yields due to the energy loss suffered by the projectile in the carbon before striking the underlying aluminum target. By simultaneously measuring the number of emitted x rays, Y_X , and the number of Rutherford scattered protons from the aluminum, Y_R , one can obtain a ratio, $R = Y_X/Y_R$. The rate of carbon buildup, $\dot{X}$, is then related to the time rate of change R by

$$\dot{X} = \frac{1}{(m+2)} \frac{\dot{R}}{R} \frac{E}{S_c(E)}, \quad (2)$$

where $m \approx 3$ and is determined from the energy dependence of the x-ray cross section, $\sigma_x = KE^m$, and $S_C(E)$ is the stopping power of carbon for protons of energy E .

Similarly, for a thick target, the buildup rate, $\dot{X}$, can be deduced from the time rate of change of the characteristic target x-ray yield per proton, $\dot{Y}_Y$, as

$$\dot{X} = \frac{1}{\ell} \frac{E}{S_C(E)} \frac{\dot{Y}_x}{Y_x}, \quad (3)$$

where $\ell \approx 4$ is the energy dependence of the thick target x-ray yield, $Y_x = KE^\ell$. The only restriction that applies to Eqs. 3 and 4 is that the stopping power of carbon, $S_C(E)$, is constant. This requires that the carbon layer be thin and hence frequent changes of the beam spot are necessary. Despite all precautions to pretreat the targets in an identical manner (the thick targets were washed with alcohol and distilled water followed by a heat treatment at 600°K for one hour before irradiation), we still observed a $\pm 30\%$ variation in the accumulation rate of carbon. However, for a particular target, this variation was less than $\pm 10\%$.

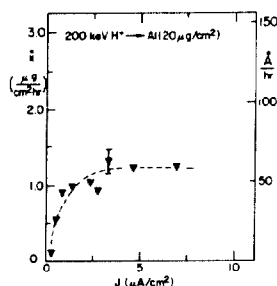


Fig. 2. Ion beam current density dependence of the carbon accumulation rate, $\dot{X}$, on a thin aluminum target.

Results and Discussion

If we assume that carbon accumulates linearly with time, and that this rate rises with current density J , our data have a consistent pattern. We find that $\dot{X}$ increases with the current density of the beam, and reaches a saturation value at $J \approx 2 \mu\text{A}/\text{cm}^2$, and remains constant thereafter. Figure 2 displays a typical current density dependence of the carbon accumulation rate. All subsequent measurements were made at high current densities to insure that we were measuring the plateau values of the accumulation rate.

At an ion beam energy of 300 keV and a Rutherford scattering angle of 135° , we measured $\dot{X}$ by using the x-ray and scattering methods simultaneously. The results are shown in Fig. 3. Our x-ray measurement of the accumulation rate was corroborated by the scattering measurement of $\dot{X}$ for the front surface. Carbon accumulates on the back surface at the same rate as on the front surface of the target. This was also demonstrated by the x-ray yield method where no discontinuity of the x-ray yield slope was observed for a target rotation of 180° degrees.

The current density dependence of the carbon accumulation rate was measured at several energies for thin targets. These data are shown in Fig. 4 and represent the average of several high current density measurements, and indicate no energy dependence of $\dot{X}$. A similar energy independence of the accumulation rate was observed by Khan et. al.¹

When the shield surrounding the target was cooled to liquid nitrogen temperature, the plateau value of the accumulation rate decreased by an order of magnitude. A simple cold surface in the vicinity of the target had the same effect. The use of a liquid nitrogen trap above the oil diffusion pump, but some 14

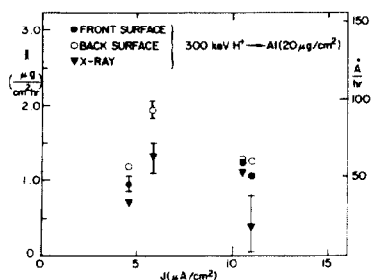


Fig. 3. The accumulation rate of carbon, $\dot{X}$, as determined by the Rutherford scattering and x-ray method described in the text.

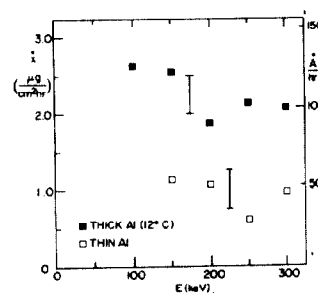


Fig. 4. The energy dependence of the carbon accumulation rate for thin and thick targets as a function of the incident beam energy.

inches distant from the target, had no measurable effect on the accumulation rate. The accumulation rate is higher for an oil diffusion pumped system than for a turbo-molecularly pumped system by a factor of ≈ 1.5 .

For thick aluminum targets, we found a similar current density dependence and lack of energy dependence, but the maximum accumulation rate was higher by a factor of two (see Fig. 4). The thick target was maintained at 12°C as was the shield around the target. There was no similar knowledge of the thin target temperature. We therefore attempted to measure the carbon accumulation rate as a function of target temperature. As can be seen in Fig. 5, there is pronounced temperature dependence. However, as the temperature of the target block was decreased, the cryogenic pumping of our target block increasingly masked the effects of the molecular dwell time on the target with the result that a maximum in the carbon accumulation rate occurs at approximately $10\text{--}20^\circ\text{C}$.

Using this temperature dependence, we deduce that the thin targets are approximately $10\text{--}20^\circ\text{C}$ above room temperature. Such a temperature rise is not unreasonable considering the heat conductivity of thin aluminum films. We also measured the accumulation rate for a thin carbon target and it was lower by an order of magnitude. This difference can be resolved if we consider that the thermal conductivity of the carbon is much lower ($\sim 1/50$) than that of aluminum. The carbon accumulation rate was also studied for a variety of thick targets (Si, Ti, and Cu) which yielded rates similar to the aluminum targets, indicating that the accumulation rate is insensitive to the material of the target surface. The accumulation rates determined in the present work are about a factor of six lower than those determined by Khan et. al.¹ and contemporaneously by investigators in this laboratory.² We attribute this reduction in the accumulation rate to advancements of

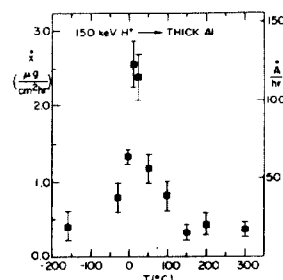


Fig. 5. The target temperature dependence of the carbon accumulation rate for 150 keV protons incident on thick aluminum targets.

diffusion pump oil technology in the past decade.

These results indicated that a study with varying carbon-molecule partial pressures would be useful. We raised the total pressure in the target volume by an order of magnitude (from 10^{-6} Torr to 10^{-5} Torr) by introducing various gases (air, CO_2 , CH_4 , and C_3H_6). With the target maintained at 12°C no appreciable change in $\dot{X}$ for any of the test gases was observed. Cooling the target to -160°C and using propylene (C_3H_6) as the test gas, a thirty fold increase in $\dot{X}$ was noted. However, upon visual inspection, the beam spot was a brownish tarry substance quite unlike the usual black beam spot. The other test gases exhibited no appreciable change in the accumulation rate shown in Fig. 5.

We find that we can parameterize all of our results by an equation of the form

$$\dot{X}(T, J) = \alpha(T) \dot{N}_0 (1 - e^{-\sigma \tau J}) \quad , \quad (4)$$

where $\alpha(T)$ is a sticking coefficient³ that depends on the target temperature, T , $\dot{N}_0$ the arrival rate of the specific gas molecules at the target surface that can be converted to carbon by proton irradiation, σ the cross section for this conversion process, and τ the dwell time of the molecule on the target surface. At high current densities, the accumulation rate becomes current-density independent, i.e., $\dot{X} = \dot{N}_0 \alpha(T)$ and is the result shown in Fig. 5 for the target temperatures $T > 300^\circ\text{K}$. The sticking coefficient, or probability, varies between 0.05 and 1 and decreases with increasing temperature of the target. This decrease is reflected in our data. Assuming that the temperature dependence of sticking probability can be expressed in terms of an activation energy, E_a , as $\alpha(T) = \exp(-E_a/KT)$ we find $E_a \approx 0.13 \text{ eV} \approx 3 \text{ kcal/mole}$ and $\dot{N}_0 = 10^{13}$ carbon atoms/ $\text{cm}^2 \text{ sec}$. An upper limit for the partial pressure of hydrocarbons in our oil diffusion pumped system was measured to be 10^{-7} Torr, or 5×10^{13} atoms/ $\text{cm}^2 \text{ sec}$, which is consistent with an arrival rate determined by assuming an activation energy or an inverse temperature dependence for the sticking probability.⁴ To determine the cross section for the hydrocarbon-to-carbon conversion process, σ , and the dwell time, τ , of the hydrocarbons on the target surface, requires precise measurements of the incipient slope of the accumulation rate at low beam current densities. Such precision was not attainable with the present experimental technique.

Conclusion

The dynamics of carbon accumulation on a solid target can be summarized as follows. Hydrocarbon gas molecules are adsorbed on the surface of the target and are converted to solid carbon by proton irradiation. This conversion process allows new gas molecules to be adsorbed and converted in turn. The accumulation rate can be decreased by either lowering the partial pressure of the hydrocarbon gas (and hence decrease the arrival rate of the molecules onto the target surface) or increasing the target temperature (which decreases the sticking probability of the gas molecules on the surface of the target). The cross section for this conversion process, as well as the dwell time of molecule on the target surface, influence the accumulation rate only at low beam current densities.

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