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PRODUCTION OF 44-Å CARBON
X RAYS BY 15-110-keV PROTONS

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ABSTRACT

Characteristic K-shell x rays (44 Å) from carbon were produced by proton bombardment of a thick target. A flow-mode proportional counter (20 cm Hg pressure: 96% He, 4% isobutane) was used to observe the photons. The thick target x-ray yield was measured over the bombarding energy range from 15 to 110 keV. With estimates of the stopping power of the carbon for the protons and the mass absorption coefficient for self absorption, the x-ray production cross section was obtained. With a value for the fluorescence yield of 0.007, the ionization cross section is obtained. This varies from $6 \times 10^{-24} \text{ cm}^2$ to $4 \times 10^{-20} \text{ cm}^2$ at 15 keV and 110 keV, respectively.

With direct observation of carbon x rays as a tool, contamination buildup on copper and aluminum was studied as a function of proton current and time. A two-parameter empirical relation was obtained of the form:

$$n = n_0 + \beta \left(1 - e^{-\alpha I_p t} \right) \text{ in units of carbon atoms/cm}^2,$$

where $\beta = 0.7 \times 10^{16} \text{ atoms/cm}^2\text{-min}$, $\alpha = 0.35 \text{ sec}/\mu\text{A}$ with t in minutes.

(System pressure was $1.5 \times 10^{-6} \text{ mm Hg}$, I_p taken at 1.5, 5, and 20 μA .) The results indicated that carbon was the principle contaminant.

INTRODUCTION

X rays produced by heavy-ion bombardment have been observed for five decades.¹ During this period of time the investigations have appeared principally in two contexts, stopping power evaluations and nuclear excitation experiments.² Recently, ionization of specific shells by protons have been studied.³⁻⁹ Models for the interaction of the bombarding particles with the atomic electrons have been constructed and calculations have been made. The general approach is that of a time-dependent, first-order perturbation calculation. The earlier calculations (K and L shells) employing the high-energy (Born approximation) description of the proton trajectory appeared valid above 0.5 MeV bombarding energy. As the measurements have been extended to lower energies, the Born approximation has been shown to predict a value of the ionization cross section higher than the observed by as much as several orders of magnitude. A more recent calculation has been made for the K-shell ionization cross section by Bang and Hansteen which considers deflection of the projectile.¹⁰ In the cases where this has been applied it has represented a significant improvement over the Born approximation calculation.

In the present work carbon was used as the target for protons of energy 15 to 110 keV. The measured quantity is the thick-target yield (x rays per proton). From this and estimates of the stopping power, mass absorption coefficient for self absorption, and the fluorescent yield, values of the ionization cross section have been calculated over the bombarding energy range. A comparison of these results with calculation shows that the Born approximation values are high by a factor of 15 at 110 keV and over 1000 below 25 keV. There are no values from the Bang and Hansteen calculation

currently available for comparison, but it is felt that the agreement should be better than for the undeflected trajectory calculation.

In the appendix to this paper is presented a brief study of carbon contamination buildup on copper and aluminum. By comparison of the carbon yield from copper and from the carbon target it seems evident that the predominant ($\geq 90\%$) contaminant is carbon. The experimental method was to simultaneously observe the carbon K x rays from the contaminant layer and the copper L x rays from the target. Absolute measurements of the copper yield have been made previously and were used to monitor any energy degradation in the proton beam. Negligible energy degradation implies thin target conditions for the contaminant, allowing a determination of the carbon thickness in atoms/cm². Buildup was measured at 1.5, 5, and 20 μ A as a function of time. Sputter cleaning (argon gas at 100 μ Hg, 600 V, 5 mA-6 min) was employed with and without physical cleaning. Lowest carbon surface densities were obtained when physical cleaning was followed by sputter cleaning. Buildup rates were the same for copper and aluminum.

APPARATUS

The experimental equipment employed in making the reported measurements consisted of an electrodeless discharge proton source, dc accelerating column (to 140 kV), bending magnet, target chamber, flow mode proportional counter, associated amplifying and scaling circuits, and a pulse-height analyzer (in coincidence with counted pulses). The experimental apparatus has been described in previous publications and will not be presented in detail here.⁹

After the positive ions (H_1^+ , H_2^+ , H_3^+ , ...) are produced in the source and accelerated, they are passed through an analyzing magnet. The proton

component is isolated and introduced into the target chamber. The beam strikes the target at 45° to the normal. Similarly, the characteristic x rays are observed at 45° . The target material was a sheet of graphite. The surface was made smooth by sanding and polishing, although some pits were unavoidable ($< 1\%$ of total surface area). The target resided in a 5-in. cube which served both as a secondary electron shield and as a part of the charge collecting system. A typical proton current was $5 \mu\text{A}$.

The characteristic x rays were detected by means of a side window proportional counter (2-in. i. d. by 12-in. length with a 0.003-in. center wire). The counter window was a $1/4$ -mil aluminized Mylar foil. The thin aluminized surface served to maintain the center wire field across the $1/2$ -in.-diameter window opening. The counter was operated at 20 cm Hg pressure in a flow mode (100 cc/min). The counting gas was 96% helium + 4% isobutane. At 20 cm Hg pressure 56% of the carbon x rays were stopped in the counter.

With a mass absorption coefficient for carbon of $3600 \text{ cm}^2/\text{g}$ and a path length of 5 cm the absorption in the counter was calculated as a function of pressure. The x-ray production was then measured at 12.5, 13.0, 14.5, 17.0, 19.5, and 20 cm Hg. These pressures corresponded to absorptions varying from 41.5 to 56%. These absorptions fell on the calculated curve to within 3%, and served as an overall test of the pressure and absorption values. (As a part of the preliminary measurements P-10 gas — 90% argon, 10% methane — was employed at 17.5 cm. It was found that for the same counter configuration, not only were the total counts low by a factor of nearly four, but the shape of the pulse-height distribution was very asymmetric with a large tail extending toward lower pulse heights. The belief is held that due to the high absorption of the argon for the carbon x rays there must be

significant numbers of recombinations taking place in the vicinity of the counter window. This suggests that the operation of a proportional counter should be maintained in such a mode that the absorption of the x rays be distributed over a large part of the diameter.)

At low bombarding energies target surface contamination has generally been a serious concern, having the effect of reducing the energy of the beam particles.⁹ The term contamination implies addition of material different than the target. On the basis of the current experiment, it is felt that most (>90%) of the usually observed contamination is carbon. This was demonstrated by two experiments. A carbon target was bombarded for several hours at 5- μ A current. The carbon x-ray yield stayed constant to within $\pm 5\%$. Under similar conditions, a copper target was placed in the beam and carbon x rays were observed. The carbon yield doubled in 12 min (at 60 keV) and appeared to be increasing linearly in time. After 20 min the copper x-ray yield (simultaneously observed) started to decrease corresponding to an energy loss in the carbon film of over 1 keV. A flaw in the reasoning might lie in the existence of some other contaminant with an x-ray line indistinguishable from carbon, and having the same x-ray production cross section over the energy range from 15 to 110 keV. This seems most unlikely. In the appendix of this paper is presented the contamination data collected. By employing the x-ray production cross section the surface density of carbon (atoms/cm²) is presented as a function of time and beam current. The dependence of rate upon current has been empirically fit by a curve. This empirical relation lends itself to physical interpretation in terms of the residence time and impingement rate of decomposable hydrocarbons upon the surface. The generalization has not yet been given a series of rigorous tests, but may be useful as a starting point for further investigation.

MEASUREMENTS

The quantity experimentally measured was the thick-target yield (I_μ) of x rays from a carbon target. From this and from estimates of the stopping power of the target material for the protons and the x-ray self-absorption coefficient, it is possible to obtain a value for the x-ray production cross section by the following formula⁸:

$$\sigma_x = \frac{1}{n} \frac{dI_\mu}{dE_p} \frac{dE_p}{dR} + \frac{\mu}{pn} I_\mu.$$

With a value of the fluorescence yield ω , the ionization cross section can be obtained:

$$\sigma_I(E_p) = \frac{1}{\omega} \sigma_x(E_p).$$

Values for the stopping power dE/dR and the absorption coefficient μ/p are estimated to be within $\pm 30\%$. The fluorescence yield ω is estimated to be 0.007. The experimental results are shown in Table I.

DISCUSSION AND CONCLUSION

The ionization cross section for the K shell can be calculated by two methods: the Born approximation description of the proton trajectory and the Bang and Hansteen deflected trajectory description. The success of the Born approximation for high-energy bombardments (> 0.5 MeV) has been established. As the proton energy is decreased, the Born approximation predicts an ionization cross section higher than that observed. This error has now been observed for a number of elements in the K and L shells, where calculations are available. In the case of carbon the Born approximation is severely in error by as much as a factor of 1000 in the range of the present measurements. This is shown in Fig. 1, where both theory and experiment are compared.

The Bang and Hansteen calculation has been performed for a number of higher-Z elements under bombardment by protons of energy greater than several hundred kilovolts. In those cases the deflected trajectory calculation represented an improvement over the Born approximation.¹⁰ It might be that the predictions for carbon would also lie closer to the observed.

APPENDIX: CONTAMINATION STUDY

Experimental Arrangement

The experimental apparatus employed to make the contamination measurements is the same as was used for the carbon yield measurements. The principle addition was that two separate differential discriminator-scalar systems were employed to simultaneously record the counts from both the copper and carbon x rays. Both scalar systems were gated on the output of a current integrator to turn off at a preset charge. As was the case during the carbon yield measurement, the proportional counter was operated in a flow mode at a reduced pressure. Therefore, the carbon counts can be considered as absolute yield numbers. At the reduced pressures only a small fraction of the copper L x rays are stopped within the counter. (Although only copper is referred to in the work, tests showed that the results were qualitatively the same for an aluminum target.)

The purpose of monitoring the copper yield was to test for change in energy of the proton beam. In the region of measurements 30 to 100 keV, a 1-keV change in energy of the protons would result in at least 10% change in the copper yield. During the course of the contamination studies, the contaminant thickness was such that less than 1 keV was lost. This defines a truly thin target condition for the contaminant.

The contaminant was identified as carbon on the following basis: The yield from a carbon target was observed for several hours. The yield remained constant to within $\pm 5\%$ (the stability limit of the apparatus). When a copper target was employed the carbon x-ray yield (or x-ray yield of lines of approximately 44-Å wavelength) increased steadily with time. After a period of approximately 20 min (at 5 μ A, $E_p = 60$ keV) the copper yield started to decrease slightly (10%), and continued to drop with increasing time. Thus, if the contaminant were other than carbon (with different x-ray production cross section as a function of energy) the carbon target number should have decreased also, or at least changed. The preceding reasoning, and the assumption that it is unlikely that there exists other contaminants of similar x-ray production and energy characteristics, implies that carbon must be the predominant ($> 90\%$) component.

The vacuum environment of the target has a direct bearing upon the results. During the course of the experiments the pressure, measured 12 in. from the target, was 1.2×10^{-6} mm Hg. This gauge was in close proximity to a cold trap. Therefore, information regarding the condensable component of the gas impinging upon the target was largely lost. However, the value of the work presented here is two-fold -- to indicate typical values for contamination buildup and to propose a functional relationship that might describe the process. With this relationship as a starting point further tests might be performed under more controlled conditions.

Data

The contamination measurements are presented in four figures. The ordinate in all cases is in units of 10^{16} atoms of carbon per cm^2 . This number is derived in the following manner. The observed counts (corrected for

geometry), window and counter absorptions are measured as x rays per proton (I_μ). Under the thin-target conditions,

$$I_\mu = n(\text{atoms/cm}^2) \sigma_x(\text{cm}^2/\text{atom}).$$

The values for the x-ray production cross section are taken from Table I.

The equation is solved for (n).

Fig. 2. Sputter cleaning. Operating at 60 keV with a proton current of 5 μ A, the carbon thickness vs time is presented during a series of bombardment and cleaning cycles. The sputtering was performed at 100 μ Hg pressure of argon gas (600 V, 5 mA) for 6 min. It is interesting to note that the contamination rate and the base line remained constant.

Figs. 3, 4. Contamination rates. In these two figures the carbon buildup is plotted against μ A-min (or protons onto target) and against time. It is clear that in neither case are the slopes for the three proton currents the same. This implies that in this range it is neither simply a conversion process actuated by the protons nor purely a surface decomposition of incident hydrocarbons.

Fig. 5. Energy dependence. In this figure are shown two sets of contamination buildups covering 30, 60, and 100 keV. Each set was measured at essentially the same proton current, showing that the contamination rate is largely independent of proton energy.

Surface cleaning is a subject of some interest. The values shown in Figs. 2-5 were taken as reproducible values following 6-min sputter cleanings; however, it was found that values of the base line at $E_p = 60$ keV lower than 8×10^{16} atoms/cm² could be obtained when the sanding and polishing was followed by sputtering. This improvement represented only a 20% decrease and was not wholly reproducible.

Interpretation

From the data represented by Figs. 3 and 4 it is possible to construct an empirical relation which contains the buildup dependence upon proton current and time. The expression obtained was:

$$n(\text{carbons atoms/cm}^2) = n_0 + 0.7 \times 10^{16} [1 - \exp(-0.35 I_p)] t,$$

where I_p is measured in μA , t in minutes, and n_0 is the base line - i. e., initial value of carbon surface density.

The general form of the time-dependent portion of the right side of the equation can be written:

$$n - n_0 = \beta(1 - e^{-\alpha I_p})t.$$

The following interpretation is put upon the constants β and α :

$\beta = v(\text{molecules of hydrocarbons/cm}^2\text{-sec}) \epsilon(\text{efficiency of conversion to carbon contaminant})$

$\alpha = \sigma(\text{interaction cross section}) \tau(\text{residence time for hydrocarbon})/A(\text{target area})$

or

$$n - n_0 = v\epsilon(1 - e^{-\sigma\tau I_p/A})t$$

Let us consider the asymptotic form of the equation.

When $I_p \rightarrow \infty$,

$$n - n_0 = v\epsilon t,$$

which is independent of proton current. This implies that the proton impingement saturates the surface with sites for conversion of hydrocarbons to carbon + gases. Under these conditions the contamination depends solely upon the rate at which the hydrocarbons strike the surface, and, of course, the conversion efficiency.

When $I_p \rightarrow 0$,

$$n - n_0 = \nu \epsilon \left(\frac{\sigma \tau I_p}{A} \right) t = \epsilon \left(\frac{\nu \tau \sigma}{A} \right) I_p t$$

which indicates a linear dependence upon I_p . It may be interpreted then that under these conditions there is an apparent saturation with hydrocarbons and now the process depends upon the protons available for conversion. It has been observed that after 16 h elapsed time with no beam on the target the contamination surface density remained constant. This carries the relation to the extreme $I_p = 0$.

As further possible tests of this relation experiments should be performed where ν is varied and where τ is varied. The tests required and the controls necessary fall deeply into the realm of surface science (chemistry and physics of surface phenomena) and are outside the scope of the present experiments.

REFERENCES AND FOOTNOTES

* Work done under the auspices of the U. S. Atomic Energy Commission.

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Table I. C_K - thick-target yield and ionization cross sections, 15-110 keV.

E_p (keV)	$N(E)$ (x rays per μC before geomet- rical and absorp- tion correc- tions)	$I_\mu (\pm 15\%)^a$ (x rays per proton)	$S(E)^b$ (keV- cm^2 per mg)	$\frac{1}{n} \frac{dI}{dE} S(E)$ (cm^2)	$\frac{1}{n} \frac{\mu}{\rho} I_\mu^c$ (cm^2)	σ_x (cm^2)	σ_I^d (cm^2)
15	7.8	1.50×10^{-7}	75	5.11×10^{-26}	6.50×10^{-27}	4.48×10^{-26}	6.40×10^{-24}
20	27	5.18×10^{-7}	100	1.77×10^{-25}	2.25×10^{-26}	2.00×10^{-25}	2.85×10^{-23}
25	62	1.19×10^{-6}	125	5.07×10^{-25}	5.18×10^{-26}	5.58×10^{-25}	7.97×10^{-23}
30	177	3.40×10^{-6}	150	1.68×10^{-24}	1.47×10^{-25}	1.83×10^{-24}	2.61×10^{-22}
40	610	1.17×10^{-5}	200	5.82×10^{-24}	5.08×10^{-25}	6.33×10^{-24}	9.05×10^{-22}
50	1,705	3.28×10^{-5}	250	1.40×10^{-23}	1.42×10^{-24}	1.53×10^{-23}	2.19×10^{-21}
60	3,694	7.09×10^{-5}	300	3.10×10^{-23}	3.07×10^{-24}	3.40×10^{-23}	4.86×10^{-21}
70	6,620	1.27×10^{-4}	350	5.39×10^{-23}	5.51×10^{-24}	5.94×10^{-23}	8.49×10^{-21}
80	12,038	2.32×10^{-4}	400	1.07×10^{-22}	1.01×10^{-23}	1.17×10^{-23}	1.66×10^{-20}
90	18,682	3.58×10^{-4}	450	1.51×10^{-22}	1.56×10^{-23}	1.66×10^{-22}	2.37×10^{-20}
100	25,641	4.93×10^{-4}	500	1.86×10^{-22}	2.14×10^{-23}	2.07×10^{-22}	2.96×10^{-20}
110	38,897	7.46×10^{-4}	550	2.42×10^{-22}	3.24×10^{-23}	2.74×10^{-22}	3.92×10^{-20}

a) $I_\mu = \Lambda N(E)/T_W A_c$; $\Lambda(\text{Geometry}) = 3364$, $T_W(\text{window transmission}) = 0.050$, $A_c(\text{counter absorption}) = 0.56$

b) S. D. Warshaw and S. K. Allison, Rev. Mod. Physics, 25, 100 (1953)

c) $\mu/\rho = 2170 \text{ cm}^2/\text{g}$; Handbook of Chemistry and Physics (Chemical Rubber Publishing Co., Cleveland, Ohio, 1962).

d) $\sigma_x = \omega \sigma_I$; $\omega = 0.007$

Fig. 1. Carbon K-shell ionization cross section.

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Fig. 2. Sputter cleaning.

Fig. 3. Contamination vs charge.

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Fig. 4. Contamination vs time.

Fig. 5. Energy dependence of contamination.

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