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ELECTRONIC DEVICES FOR NUCLEAR PHYSICS

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PART I

FUNDAMENTAL INVESTIGATIONS

I. Photocathode Research

The work on photocathodes can be conveniently reviewed under two separate headings, i.e., firstly, activation process and characteristics of multi-alkali antimony cathodes (alloys of antimony with more than one alkali metal) and, secondly, experiments with completed cathodes to investigate the mechanism of photoelectric emission. The second heading covers such subjects as chemical analysis of photosensitive layers, structural examination by x-ray diffraction, and the study of the photo- and semi-conducting properties of the materials.

1. Multi-alkali cathodes

a. Sb-K-Na

A reproducible two-stage activation process has been developed according to which an Sb-K cathode is produced first and the K in the layer is then partially replaced by Na. During this replacement process the photosensitivity increases steadily from approximately $2\mu\text{a/l}$ to over $20\mu\text{a/l}$, and the process is stopped when peak sensitivity is reached. Curve (a) in Fig. 1 shows a typical spectral response curve of an Sb-K-Na layer. Owing to its relatively high blue response and low thermionic emission, this cathode appeared suitable for multiplier tubes; however, work on this application has been postponed because the more promising layer, described below, was discovered in the meantime.

b. Sb-K-Li

It was found that if the K in an Sb-K cathode is partially replaced by Li, a rise in photosensitivity is obtained similar to that produced by Na. So far, overall sensitivities have been in the 5 to 10 μ a/l range but the spectral response curve (Fig 1(b)) shows a reasonably good efficiency in the blue range. The great advantage of this layer over the Sb-K-Na layer is its very much lower red response and thermionic emission. Exact measurements of the latter are not yet available, but there are indications that the emission at room temperature is below 10⁻¹⁶ amps. per square cm. In addition, whereas under production conditions it is difficult to introduce controlled amounts of Na into a tube, Li can be conveniently evaporated from a filament.

c. Sb-K-Na-Cs

Of all the investigated multi-alkali cathodes the Sb-K-Na-Cs layer has the highest peak quantum efficiency and red response. (See Quarterly Report No. 12) However, it has also the highest thermionic emission at room temperature and this characteristic makes it less suitable for multiplier tubes, particularly for use in scintillation counters where the high red response does not represent any advantage. Therefore, as far as work covered by this contract is concerned, this layer is now of interest only to the extent that it yields information on the mechanism of photoelectric emission.

Whereas the cathodes described under (a), (b) and (c) have been developed to a stage where they have useful sensitivity

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many other layers of different compositions have been produced which are briefly described below:

d. Sb-K-Cs

Sensitivities of up to $30\mu\text{a}/\text{l}$ were obtained, but the cathodes did not appear to have any advantages over the simpler Sb-Cs layer.

e. Sb-Rb-Na

This composition was tried only once and had very low sensitivity.

f. Sb-Na-Li

One unsuccessful tube with this cathode was made. The failure may be due to the high evaporation temperatures of both Na and Li which makes it impossible to replace in a convenient way one alkali metal by the other.

g. Sb-K-Rb-Cs, Sb-Na-Rb-Cs, Sb-K-Li-Cs

These three layers have Cs and a second alkali metal in common with the layer described under (c), but a different alkali metal is used as the third component. It was hoped that the first two layers in which Rb replaces an alkali metal of higher work function might for this reason have higher sensitivity than layer (c). However, the results were disappointing in all three cases.

Finally, three layers (two of which do not fall under the heading of multi-alkali cathodes) were investigated in which Sb was replaced by another metal. The sensitivities were low in all cases, but the reasons for trying these particular combinations are of sufficient interest to justify a brief description.

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h. Bi-K-Na

In the past Bi has replaced Sb successfully in photocathodes containing Ag, hence it seemed possible that Bi might be useful in multi-alkali cathodes.

i. As-Li

It has often been suggested that As-Cs might have high photoelectric sensitivity, but the technical difficulty of combining two metals of very low evaporation temperature has always been an obstacle to a detailed examination of this compound. This objection does not apply in the case of As-Li because Li evaporates at high temperature. The usual method of evaporating a metal, such as Sb, from a filament and then distilling the alkali metal from a side-tube was, therefore, reversed, i.e., Li was evaporated from a filament first and As was then distilled from a side-tube. Despite the fact that the As had previously been freed from oxide by sublimation, the photoelectric emission was very low at all times.

j. Re-Cs

A recent publication (A. v. Grosse, Z. f. Naturforsch., vol. 8b, (1953) page 533) referred to the formation of a polar compound of rhenium metal with alkali metals in which Re acts as a negative ion. It was, therefore, tried to make such a compound by exposing an evaporated Re layer to Cs vapor in the hope of obtaining a photosensitive compound. However, preliminary experiments showed neither compound formation nor appreciable photosensitivity.

k. Summary

The results obtained with the cathodes described under (a), (b) and (c) prove that in certain circumstances Sb, in combination with two or three alkali metals produces much higher photosensitivity than Sb combined with only one of these alkali metals. It is not yet possible to propose a coherent explanation for this unexpected effect, particularly as the described experiments permit two alternative interpretations: In view of the fact that the three most successful combinations all contain K, it is possible that a specific effect of K produces the high sensitivity. However, a more likely assumption is that under optimum experimental conditions, all combinations containing more than one alkali metal would produce higher sensitivity than single alkali metals. Owing to the great difference in vapor pressure between the alkali metals, it may be very difficult or even impossible to devise an activation process by which the optimum amount of each metal is introduced into the tube in the optimum sequence and at the optimum temperature, so that the failures of some of the described combinations may be due merely to the fact that the best experimental conditions have not yet been discovered.

In order to shed some light on this and related problems, the work reported in the following sections has been undertaken.

2. Basic Experiments

a. Chemical Analyses

Using the micro-methods described in Quarterly Report No. 13, a considerable number of cathodes was analyzed to find the optimum ratio of the alkali metals in multi-alkali cathodes, as well as to determine the ratio of Sb to the total alkali metal content.

The results are summarized in Table I. The samples containing the same alkali metals are arranged in series of decreasing sensitivity. The spread in the results does not yet permit any final interpretation of the analyses, but the following provisional conclusions may be worth recording.

(1) The Na:K ratio in all Sb-K-Na cathodes of high sensitivity (Nos. 4, 5, 6, 7 and 8) covers the range from 1.5 to 2.5 while the values for low sensitivity cathodes (Nos. 9 and 10) lie well outside this range. This may indicate that the optimum ratio would actually be 2:1.

(2) The Na:K ratio in all Sb-K-Na-Cs cathodes of high sensitivity (Nos. 11, 12, 13, 14 and 15) also covers the range from 1.5 to 2.5. The Cs content of these cathodes is very small by comparison and one might speculate that the cathode is basically an Sb-K-Na cathode with a Cs surface to which the high red response may be attributed.

(3) The results of the Sb analyses are inconsistent and very puzzling. It has been accepted for many years that the Sb-Cs cathode consists of a stoichiometric compound of the formula SbCs_3 and it was, therefore, expected

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TABLE I

<u>No.</u>	<u>Sample No.</u>	<u>Composition</u>	<u>$\mu\text{a/l}$</u>	<u>Atomic ratio Na:K:Cs</u>	<u>Atomic ratio alkali metal:Sb</u>
1	S-101	Sb-Cs	60	---	10.5
2	S-107*	Sb-Cs	40	---	2.9
3	S-115	Sb-Cs	25	---	5.2
4	S-147*	Sb-K-Na	32	2.5:1	3.4
5	S-140	Sb-K-Na	27	2.2:1	3.8
6	S-121	Sb-K-Na	18	2.5:1	---
7	S-139*	Sb-K-Na	18	2.0:1	3.6
8	S-129	Sb-K-Na	15	1.8:1	7.3
9	S-92	Sb-K-Na	6	0.8:1	3.5
10	S-100	Sb-K-Na	0.6	3.0:1	---
11	S-110	Sb-K-Na-Cs	140	1.7:1:0.40	3.1
12	S-97	Sb-K-Na-Cs	96	1.5:1:0.26	9.2
13	S-189	Sb-K-Na-Cs	95	1.6:1:0.07	---
14	S-191	Sb-K-Na-Cs	90	2.0:1:0.12	---
15	S-188	Sb-K-Na-Cs	86	2.2:1:0.09	---
16	S-183	Sb-K-Na-Cs	13	3.9:1:0.24	11.6
17	S-170	Sb-K-Na-Cs	<1	4.3:1:0.14	9.0
18	S-108	Sb-K-Na-Cs	0.5	11:1:10	---

*Opaque Sb layers

that the multi-alkali cathode corresponds to a formula SbX_3 , where X is the sum of the alkali metals used. A first glance at the Table shows that the X:Sb ratio in the measured samples varies from 2.9 to 11.6, without any obvious relationship between this ratio and the cathode sensitivity.

It should be noted that the analyses, unless otherwise marked, refer to semi-transparent Sb layers, whereas the earlier evidence for the $SbCs_3$ compound was based on analyses of thicker (opaque) Sb-layers. For this reason, the analyses were extended to thick Sb-K-Na cathodes and, as a control, also to semitransparent Sb-Cs cathodes. The Table shows that in thick layers of the Sb-Cs (No. 2) and of the Sb-K-Na (Nos. 4 and 7) type, a ratio corresponding approximately to the formula SbX_3 was confirmed and that the large variation in ratios was found in Sb-Cs cathodes (Nos. 1 and 3), as long as they are semi-transparent. Many attempts have been made to explain this discrepancy in the analytical results between thick and semi-transparent cathodes, in particular by checking the accuracy of the Sb-analysis, but so far no reasonable interpretation has emerged.

The suggestion may be added that if the excess over the ratio 3:1 can be proved to be not real, the above-mentioned ratio of 2:1 of Na to K may indicate that the basic unit of the multi-alkali cathodes is represented by the formula $SbNa_2K$.

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b. X-ray diffraction measurements

Work has continued on X-ray diffraction experiments with the purpose of gaining information on the structure of the photoelectric Sb-alloys which might supplement the results of chemical analyses.

A more refined technique was developed to produce the photoelectric compounds in a .01 mm wall glass capillary, to reduce the X-ray absorption in the glass which had proved troublesome in earlier experiments. First results with the Sb-Cs compound yielded the same pattern as previous determinations and did not correspond to the patterns published for Li_3Sb , Na_3Sb and K_3Sb . It was, therefore, decided as a control to examine a sample of Sb-K whose structure has been published in the literature. A pattern was obtained that agreed with the published structure*, despite the fact that the earlier work had been done with samples produced by fusing the two metals together in bulk, whereas in the present experiments fine Sb powder was exposed to alkali metal vapor to simulate the conditions obtaining in the activation of photoelectric cathodes. In view of the satisfactory result with this control experiment on Sb-K, the tests on Sb-Cs will be continued.

c. Measurements of photoconductivity

It has been known for some time that the Sb-Cs photocathode is a semiconductor. Using the conventional energy level diagram, Apker, Taft and Dickey (J. Opt. Soc. Amer. 43, 78(1953)) have suggested that the width of the "forbidden gap" between the valence and the conduction band for this semiconductor corresponds

*Lines due to Sb and KOH were also present in the pattern.

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to the photon energy of the light which produces the maximum quantum efficiency. This model postulates that the vacuum level coincides with the bottom of the conduction band or lies somewhat below it. The "forbidden gap" is given as approximately two volts.

To investigate the photon activation of carriers in the range of the "forbidden gap", the photoconductivity in photocathodes was examined. To minimize the photoemissive effect, opaque layers were deposited (between silver paste electrodes) on glass and illuminated from the glass side. Photocathodes of the Sb-Na, Sb-K and Sb-Cs types were prepared. A photoconductive response was found at room and liquid nitrogen temperatures for Sb-Na, at liquid nitrogen temperature for Sb-K and a feeble response for Sb-Cs at liquid nitrogen temperature. Photoconductivity could be identified electrically distinct from photoemission since the former response was reasonably linear with voltage and the latter saturated with voltage. More conclusive evidence of photoconductivity was a marked difference in spectral response which for all the layers extended into the infrared. This relatively high red and infrared response was not observed when the layer was operated as a photoemitter.

The spectral response curves of photoconductivity and photoemission were measured for Sb-K at liquid nitrogen temperature with 25-cycle chopped radiation.* The photoconductive response rose about two orders of magnitude from $13,000 \text{ \AA}$ to $4,300 \text{ \AA}$. The photoemissive response rose over four orders of

*Measurement by Dr. E. E. Hahn of these Laboratories

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magnitude from 6,500 Å to a peak at 3,600 Å. Identification of an intrinsic energy level from the photoconductive response curve was difficult since the long wavelength cut-off was so broad.

The direction of the thermal emf was then determined for the Sb-K layer. With one contact around room temperature and the other at liquid nitrogen temperature, the measurement indicated n-type material.

The photoconductive spectral response will be measured for the Sb-Na layer. Samples of Sb-K-Na and Sb-K-Na-Cs will be prepared and measured.

While the weight of present experimental evidence points toward a model wherein the vacuum potential is above the bottom of the conduction band, and having a number of ionizable impurity levels lying in the "forbidden gap", much more work must be done before this model can be positively confirmed. This work includes further photoconductive measurements, investigation of surface and thickness effects, and detailed preparation studies.

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II. Emission Time of Secondary Electrons

The experimental work on this project has been completed and will be fully described in a separate report. For this reason, only a brief statement summarizing the results is given here.

After completion of the experiments at 400 mc, the final conclusion was reached that the maximum time spread of secondary electrons is 7×10^{-11} sec. Below this value the transit time spread inherent in the experimental set-up prevents the determination of an exact value, but it is thought that the spread in secondary emission is actually far below the quoted upper limit.

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PART IIPHOTOMULTIPLIERSI. 70 cm² Photocathode (Type H-5037)

In addition to the three tubes mentioned in Quarterly Report No. 13, three more tubes were completed and all gave satisfactory results upon testing. It is not intended to make more tubes of this type until test reports have been received from the laboratories to which the tubes have been sent.

II. 11 cm² Photocathode (Type H-5914)

Since the last report, six tubes have been made which were, on the whole, satisfactory as regards sensitivity and gain. Some trouble was experienced with instability at high potentials and the last tube was, therefore, provided with a charcoal trap so that the effect of excessive residual gas pressure on stability and after-pulses can be investigated. Some of the tubes contained a focusing electrode connected to the last dynode in order to raise the output current. With optimum potential distribution, currents of up to .5 amp. were measured. Some tubes have been sent out, but no reports have been received so far.

III. 9" Photocathode

The design of a multiplier with a 9" cathode has been started.

An experimental tube was activated in which the multiplier was replaced by a phosphor screen which can be moved along the axis of the tube to establish the optimum position for the multiplier in the final tube. This tube is also being studied

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for optimum potentials on the lens system which consists of three conducting layers on the glass wall. First results indicate that the electrons from the whole cathode can be focused easily onto an area of about 1" diameter. This tube will require a fairly large 1st dynode region and, consequently, an electron optical system with the small in-line structure and a new first dynode as described in the next section was studied.

IV. Test Multiplier with 11 cm² Photocathode (Type H-6384)

This tube is similar to Type H-5914, but has several new features. In order to improve the collection efficiency from the photocathode, a large 1st dynode has been designed, on the basis of rubber model experiments, to intercept a photo-electron beam of more than 1/4" diameter. In addition, parts preparation and construction have been simplified by using dynodes which are cylindrical circular arcs. The collector consists of a flat strip which is placed in such a position that it enhances the field at the last dynode without intercepting electrons emitted from the previous dynode.

Two test tubes of this type were constructed and found to perform quite satisfactorily. The first dynode region was investigated in some detail inasmuch as it is the important feature of the structure. The three-dimensional chart in Fig. 2 showing the tube output as a function of the 1st dynode and aperture potentials is included to illustrate one of the methods and to study the processes going on in such systems.

Fig. 3 is a photograph of the 11 cm² Type H-6384 multiplier, while Fig. 4 is a sketch of the structure of this tube.

Sensitivity in
arbitrary units
(Corrected for
absolute energy)

A: Sb-Ka-Na
B: Sb-K-Li

Fig 1 - Color Response of Photocathodes

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Fig 2. - Three Dimensional Chart

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Fig. 3 - Phototube Type H-6384

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Fig. 4 - Photomultiplier Type H-6384

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