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A STUDY OF FATIGUE IN SILVER-OXYGEN-CESIUM PHOTOCATHODES.

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A STUDY OF FATIGUE IN SILVER-OXYGEN-CESIUM PHOTOCATHODES

P. G. Borzyak, V. F. Bibik and G. S. Kramarenko

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A STUDY OF FATIGUE IN SILVER-OXYGEN-CESIUM PHOTOCATHODES

P. G. Borzyak, V. F. Bibik and G. S. Kramarenko

The article gives experimental results on fatigue of silver-oxygen-cesium photocathodes that do not correspond to existing ideas as to the nature of this phenomenon. There are indications that a major role is played here by volume changes in the oxygen-cesium films due to the effect of illumination.

The existing ideas as to the nature of silver-oxygen-cesium (s-o-c) photocathode fatigue [1, 2, 3] are to a considerable degree hypothetical. It is thus understandable that attempts have been made to subject these ideas to additional experimental verification; there is also a need for this in view of the added fact that the problem of fatigue in practical phototube applications is far from solved, and has remained topical. Moreover, new and even directly contradictory experimental facts, unknown to the authors of the hypotheses as to the nature of fatigue have turned up. This indicates that the widely accepted concepts were constructed on the basis of incomplete experimental data and that the very task of collecting such data is still not completed and is still an urgent problem.

It is to this task that the investigations whose results are reported below are subordinated.

1. INADEQUACY OF THE CONCEPT OF ELECTROLYTIC REMOVAL OF CESIUM FROM THE CATHODE SURFACE

Fatigue of s-o-c photocathodes is frequently accompanied by a reduction in infrared sensitivity and some shift of the red cutoff toward shorter wavelengths. The latter fact is treated as evidence for an increase in the work function - in the amount of energy needed for electrons to escape the cathode. This proposition is not selfevident, however. Thus, for example, Pakswer [4], following De Boer's opinion as to the nature of the s-o-c photocathode, finds the cause of fatigue in the fact that some of the photoionized adsorbed cesium atoms remain on the surface as ions while their field polarizes neighboring atoms so as to change their selective absorption of light, leading to the indicated variations in cathode spectral sensitivity. But in this case, the substitution of positive ions for some of the cesium atoms on the cathode surface should lead to a reduction, rather than to an increase, in the thermionic work function for the cathode and to a corresponding shift in the contact potential.

To check the variation in contact potential, D. Gorodetskiy placed a carefully outgassed miniature thermionic cathode in the center of a spherical phototube envelope; the phototube cathode served to collect the electrons from the thermionic cathode. By taking the volt-ampere characteristics for the thermocurrent for retarding potential differences, it is possible to use the shift in the characteristics to follow the changes in thermionic work function that occur for a photocathode. The results of such measurements are shown in Fig. 1. First the photocathode spectral response (curve 1, Fig. 1a) and the thermocurrent voltampere characteristic (curve 1, Fig. 1b) were measured. The photocathode was then subjected to fatigue, after which the spectral-response and voltampere curves 2 were obtained. After the photocathode was "rested: for a certain period, curves 3 were obtained, and after some more fatigue, curves 4.

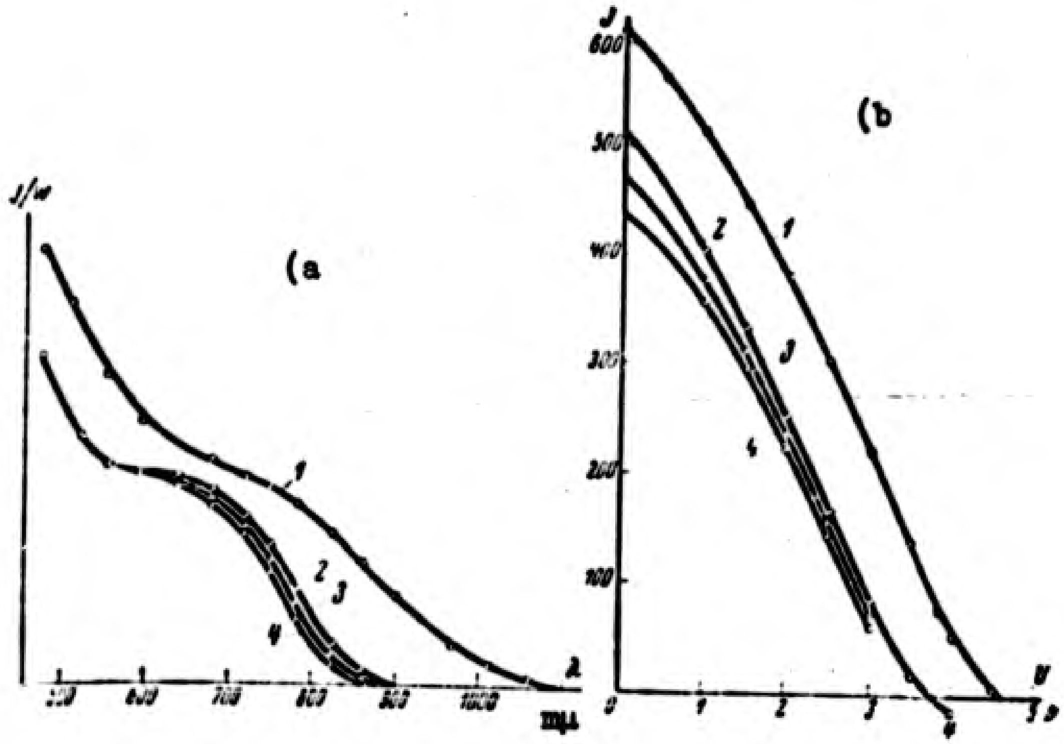

Fig. 1. Variation in spectral sensitivity (a) and contact potential (b) for photocathode under fatigue.

The direction in which the volt-ampere curves shift clearly shows that the shift in the red branch of the photocathode spectral-response curve toward shorter wavelengths is accompanied by an increase in the thermionic work function and vice versa. It should be noted that here there is no quantitative relationship between the shift in the photoelectric-effect red cutoff and the contact potential. This result should not be considered impossible for a semiconductor or composite cathode, which represents a mixture of metal and semiconductor particles; we need only remember that the thermionic work function is determined by the position of the chemical-potential level and the photoelectric work function by the electronic level. This can only indicate that not only variations in cathode surface conditions produce fatigue, but also changes in cathode bulk properties, since it is only in this case that the illumination can affect the position of the chemical-potential level.

In any case, the experimental result indicates directly that the loss of sensitivity near the thresholds is not due to conversion of adsorbed cesium atoms to ions adsorbed on the cathode surface. If the primary cause were photoionization of adsorbed cesium atoms, the cesium ions would not remain on the surface, but would slowly leave; in the final analysis, this should lead to cesium depletion of the surface, to a reduction in the emission-center concentration according to de Boer, and together with this to an increase in the work function. According to de Boer, the mechanism involved in the removal of cesium ions from the surface layer is a process of electrolytic removal to the inside of the cathode due to the field existing within the cathode film.

Direct verification of the notion that cesium leaves the cathode surface during fatigue may be attempted. If the cathode surface is cesium-impoverished, then additional destruction, say by the introduction of a small amount of oxygen, can only lead to both a further increase in work function and decreased photocathode sensitivity. We carried out appropriate simple observations; the results are shown in Fig. 2. The curves of Fig. 2 indicate a change in sensitivity for intensively illuminated photocathodes in the course of time, while the arrows show the direction in which the sensitivity changed when oxygen was introduced. We unexpectedly found cases in which the sensitivity increased both with slight and with well-defined fatigue.

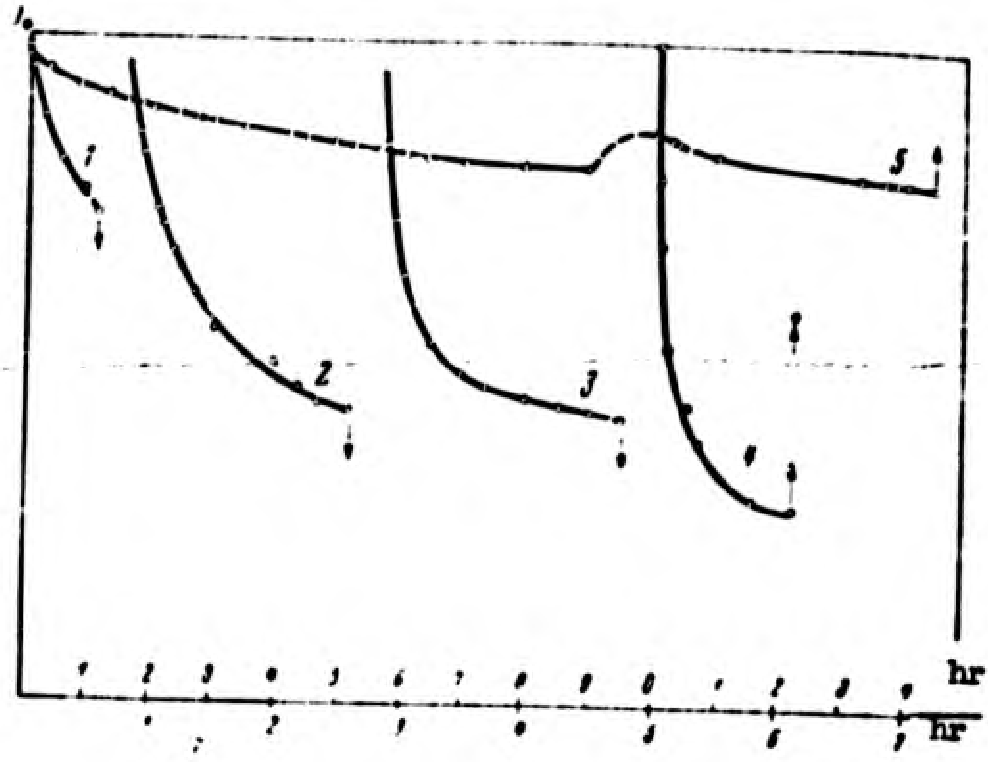

Fig. 2. Variation in sensitivity of fatigued photocathodes (indicated by arrows) with small amount of oxygen supplied to phototube. The lower time scale applies to curves 1-4 and the upper to curve 5.

Without doubt, these results indicate that if electrolytic removal of cesium from the cathode surface together with diffusion back in the form of neutralized atoms played a decisive role in the phenomena of fatigue and recovery, these will not be the responsible factors in every case, or the only responsible factors.

To strengthen this conclusion, we must be convinced that fatigue can also be observed where the electrolysis process is known to be excluded. There are two ways of doing this: we may use oxygen-cesium films that are so thin that it makes no sense to speak of electrolysis in them, or we may illuminate the cathode without applying voltage across the phototube and without drawing current from it. Well-defined fatigue was observed by us in both cases.

Fig. 3 shows the results of one experiment for the second case.

The experiments were carried out in the following manner. Using a special illuminator, we projected a sharp image of a uniformly illuminated round diaphragm onto the photocathode. Where this white-light image fell, we projected the image of the slit of a monochromator delivering a light beam at λ ≈ 960 mμ. The cathode could be illuminated with either beam continuously or just during measurements, and it was possible to follow the photocathode sensitivity variations with respect to both white light and infrared. Where it was not desirable to "spoil" the cathode with white light while there was voltage across the phototube, it was possible to trace the sensitivity variations by means of the infrared test pattern (Fig. 3).

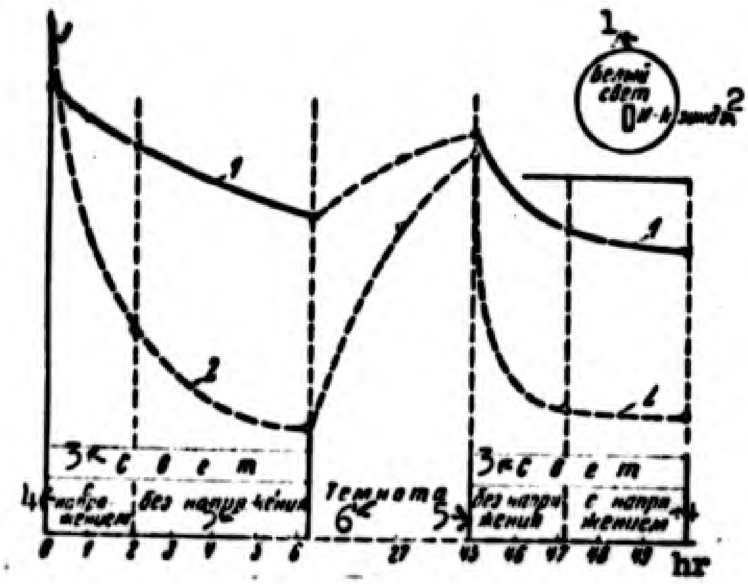

Fig. 3. Variations in integral sensitivity (curves 1) and spectral response for λ = 960 mμ (curves 2) of photocathode subjected to illumination with and without emission-current takeoff.
1) White light; 2) infrared pattern; 3) light; 4) with voltage; 5) without voltage; 6) dark.

It is clear from Fig. 3, where the variation in "white" sensitivity is shown by curve 1 and in infrared sensitivity by curve 2, that they both change under just one illumination of the cathode, with no current being drawn. As with all other fatigue phenomena, we cannot say that this example reflects the universal behavior of photocathodes. But such an effect has been observed after just one illumination both with our laboratory specimens and with commercial type TsV phototubes. As a consequence, electrolytic processes in the cathode due to the voltage applied to an illuminated phototube cannot always be used to account for fatigue. There are also other processes produced by light alone and leading to a change in cathode sensitivity.

2. CHANGES IN SENSITIVITY OF NONILLUMINATED PARTS OF CATHODE

Many observations have shown that under local illumination, the changes occurring in the cathode also extend to neighboring nonilluminated areas.

We first dealt with the question of whether photocathode fatigue depends not only on illumination intensity (and the associated current density) but also on the dimensions of the illuminated surface. To check this, we made an illuminator using a slit from a spectral instrument. A good photographic objective lens was used to project a clear sharp image of the uniformly illuminated slit on the cathode. A type TsV-4 phototube was investigated. The photocathode was illuminated with a light pattern about 0.08 mm wide; by moving the sides of the slit, we then increased the width of the pattern to 4.2 mm while holding the brightness constant. Figure 4 shows the results of observations of cathode-sensitivity variations; here we have plotted the ratio of the observed photocurrent J to the initial current J_1, in per cent, i.e., the relative photocathode sensitivity for both cases.

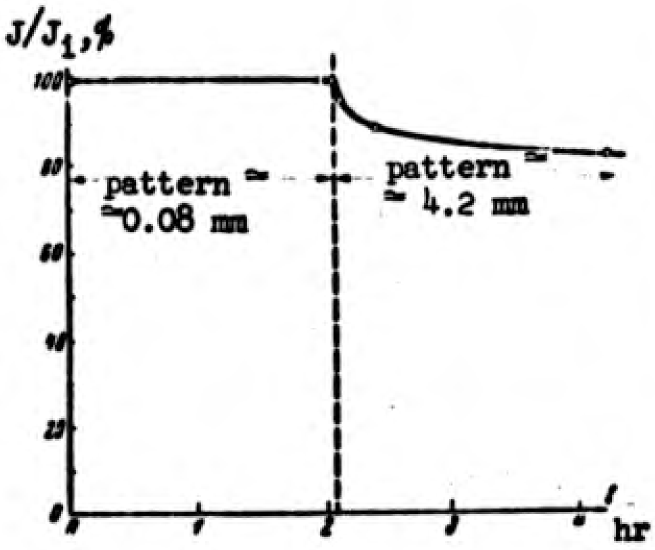

Fig. 4. Effect of size of illuminated surface on stability of local photocathode sensitivity.

The fact that fatigue was observed with a wide pattern but was imperceptible with a very narrow pattern indicates that there are processes in the cathode that lead to "healing" of areas deteriorating under local illumination. Such healing may occur as a result of the participation of the remaining nonilluminated portions of the cathode. Consequently, local cathode fatigue must also affect the neighboring nonilluminated sections.

To check the conclusion that suggested itself, we carried out further experiments. It may be assumed that the phenomenon should be easily observable where there is a considerable relative area of "spoiled" surface. Thus a large spot a on the photocathode was subjected to uniform white-light illumination, while the pattern b (Fig. 5) was used to check the condition of the cathode in a neighboring nonilluminated portion. The illumination on the white spot was of the order of several hundred lux. Before beginning the experiment, we took the spectral-response curve for spot b (with no adjustment to equal energy). It is shown by curve 1 of Fig. 5. Next the pattern was switched off and the "fatigue" light applied to spot a.

After some fatigue of the cathode at spot a, the white light was switched off and the spectral response of area b again measured. The experimental results are shown by curve 2 of Fig. 5. Curve 3 gives the spectral dependence of the sensitivity loss at area b in per cent.

Thus, on the basis of the experimental data we can conclude that the processes occurring in a working cathode involve not only the illuminated part of the cathode but its entire area. If we assume that, regardless of other phenomena, fatigue is somehow connected with cesium depletion of the illuminated cathode surface, the following fact stands out. In a phototube, there is a definite equilibrium relationship among the free cesium contained within the cathode, in the surface film, and in the atmosphere around the cathode. If the action of light causes cesium impoverishment of the surface in some area of the cathode, for example, the upsetting of the equilibrium relationship will cause cesium to be supplied to the impoverished surface from the atmosphere. This decreases the cesium vapor pressure above the film, upsets the equilibrium between the atmosphere and the nonilluminated cathode areas, and causes cesium to evaporate from the latter. Thus there will be an exchange of cesium through the atmosphere between the illuminated and nonilluminated areas.

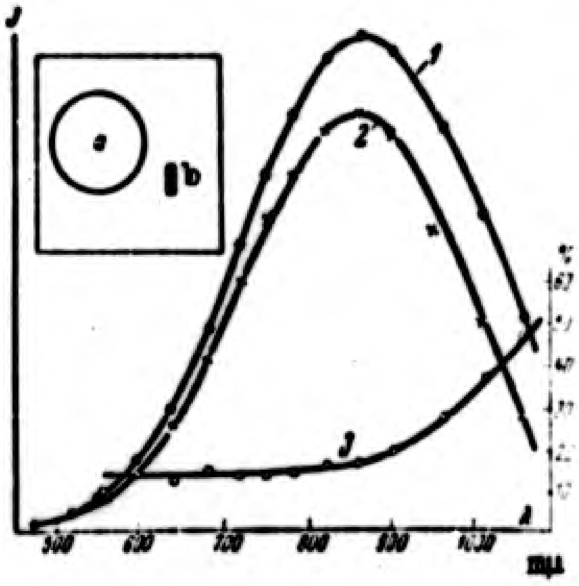

Fig. 5. Variation of local sensitivity in nonilluminated portion (b) with fatigue of cathode by white light in neighboring portion (a). 1) Local spectral response at (b) at beginning of experiment; 2) the same local spectral response at the end of the experiment; 3) spectral dependence of local sensitivity loss at (b).

If there were such a mechanism for interaction of different parts of the cathode, then over the area of a homogeneous cathode parts very close to or far away from a fatigued spot should be under identical conditions and should react in similar fashion to local fatigue. In exactly the same manner, where there are two separate cathodes in a common envelope, the fatigue of one of them should affect the properties of the other. To test this, photocathodes on two separate plates placed side-by-side were mounted in a cylindrical envelope. One of them was fatigued and the condition of the other observed. It turned out that the fatigue of one cathode had absolutely no effect on the sensitivity of the other.

There was another version of this experiment: a photocathode was made with uniform sensitivity over its surface. It was placed on a support provided with a micrometer screw in front of an illuminator that could be used for local fatigue. The sensitivity was first measured at several points located at various distances from the fatigued area; it proved to be roughly the same everywhere. Then cathode fatigue was brought to the saturation point in the illuminated area and the sensitivity again measured at the same points as in the beginning of the experiment. The results are shown in Fig. 6, where the values (in per cent) of local sensitivity loss are given as a function of the distance to the fatigued area. The shape of this curve also fails to support the theory that there is interaction between cathode areas through the atmosphere in the envelope.

The lack of interaction between the separate photocathodes and the nature of the data of Fig. 6 indicates that some sort of diffusion or migration processes are at work, acting along the cathode film both transversely and longitudinally. Through their agency, the interaction between different areas of the cathode takes place. If we assume that this process involves diffusion of cesium atoms, we would need to have an embarrassingly large value for the coefficient of diffusion (or migration), since the process propagates over tens of millimeters within several hours. This fact still leaves the nature of this process an open question.

3. EFFECT OF INFRARED ILLUMINATION

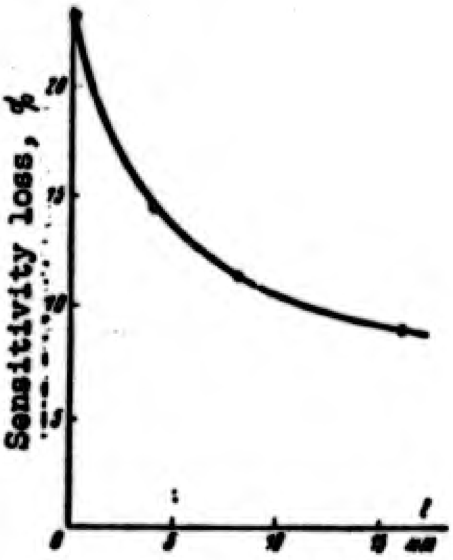

Fig. 6. Local sensitivity loss by non-illuminated areas of cathode as a function of distance between these areas and the illuminated area on a cathode with a uniform initial sensitivity.

Until recently, all investigators usually assumed that red and infrared illumination of a silver-oxygen-cesium photocathode not only caused no fatigue but, on the contrary, facilitated recovery of sensitivity lost under violet-end illumination. At the conference on cathode electronics of 1951, however, the report of Yumatov [5] emphasized that, according to his observations, infrared illumination caused fatigue of S-O-C photocathodes. Our experiments not only confirmed Yumatov's report, but yielded additional data on this question.

The test was carried out on a production specimen of the common TsV-4 phototube, manufactured commercially, rather than on a laboratory specimen. Infrared was obtained from the illuminator with the aid of a light filter, which isolated the spectral region from $\lambda > 820$ mμ from the total radiation of an incandescent bulb. For comparison, the broken line on Fig. 7 shows the sensitivity variation for one of our gold-silver-oxygen-cesium photocathodes [6] illuminated with white light of an intensity such that the initial currents were roughly the same for both of the phototubes compared.

It is clear from the data of Fig. 7 that infrared can produce the same, and sometimes even more pronounced, fatigue effects in a photocathode, including irreversible effects, as white light.

The problem of the red cutoff for the fatigue action of light is undoubtedly of interest. Since we did not have the required set of graduated light filters, we attempted to use a monochromator. In order to hamper the development of a "healing" process in the fatigued area, the operations were carried out with a photocathode cooled by liquid nitrogen and illuminated with light at $\lambda = 960$ mμ. Well-defined fatigue appeared in this case. The processes responsible for interaction of the various cathode areas actually turned out to be "frozen," since small test displacements of the light pattern away from the area of long-term illumination (carried out with the aid of the micrometer screw) led to an abrupt increase in the photocurrent; this indicates that the "injured" area remained isolated from the adjacent areas of the cathode. The results obtained show that the red cutoff for the fatigue action of light is $\lambda_0 \gtrsim 960$ mμ.

It follows from the infrared experiments that the problem of the spectral dependence of the fatigue effect of light has not been accounted for, as is usually assumed in the literature on the basis of the data of de Boer and Teves. Unfortunately, neither this article nor the de Boer book [2] give a detailed description of the exact method used to obtain the results. In any case, however, it is clear that they are incomplete and, as a consequence, cannot be used as a basis for a general theory of fatigue in silver-oxygen-cesium photocathodes.

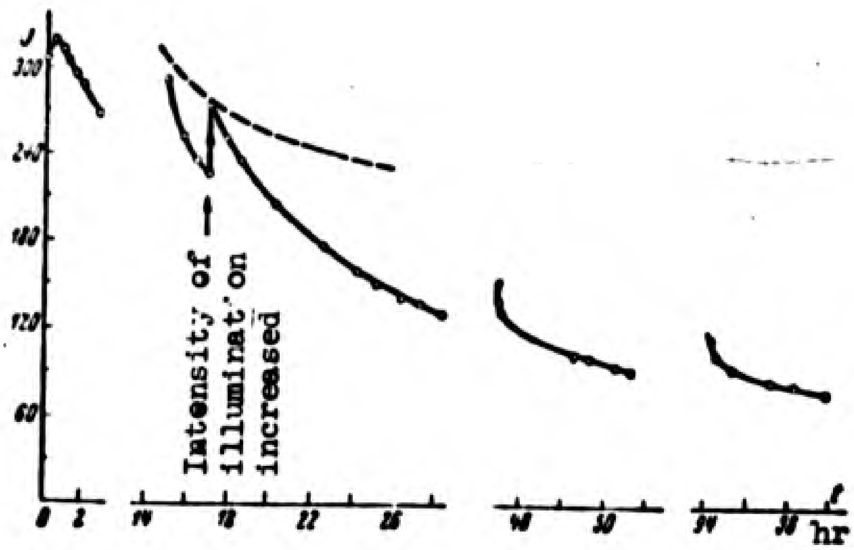

Fig. 7. Variation in photocathode sensitivity with time under infrared illumination ($\lambda > 820$ mμ). Periods of illumination alternated with periods of darkness.

4. ILLUMINATION AFTEREFFECTS

During attempts to ascertain the degree to which photocathode sensitivity losses due to fatigue are irreversible, interesting cases of well-defined light aftereffects were found for both white and infrared illumination. The effect involved the following: after the illumination was switched off, phototube dark sensitivity did not recover, but continued to drop. The data of Fig. 8, also referring to a type TsV-4 phototube, illustrate this. The phototube was installed in front of the monochromator and white-light illuminator so that after white-light fatigue of the photocathode it was possible to check the condition of the cathode by monochromatic light. Before the experiment was begun, we took the spectral-response curve (not adjusted to equal energies); it is shown by curve 1 of Fig. 8. Next the white-light illumination was begun. After moderate over-all fatigue of the cathode, we checked its sensitivity to light at $\lambda = 1060$ mμ. This sensitivity had dropped from the initial value to the value shown by arrow 1.

We next wished to see how the sensitivity recovered while the light was switched off. A measurement lasting about one and one half hours showed, to our astonishment, that there was no recovery of spectral sensitivity in the $\lambda = 1060$ mμ region, and that in fact it dropped further as shown by arrow 2. After 15.5 hr, the result shown by arrow 3 was obtained; curve 3 shows the measurement of the entire spectral characteristic. Thus, instead of "healing" in the dark, the phototube continued to lose sensitivity. Curve 4 was taken 24 hr after curve 3 was obtained, and then white light of somewhat lower intensity than that first used was applied. Here in place of fatigue, we began to observe a gradual increase in photocathode sensitivity; after 6 hr 40 min we obtained the change shown by arrow 5, while curve 5 was obtained later.

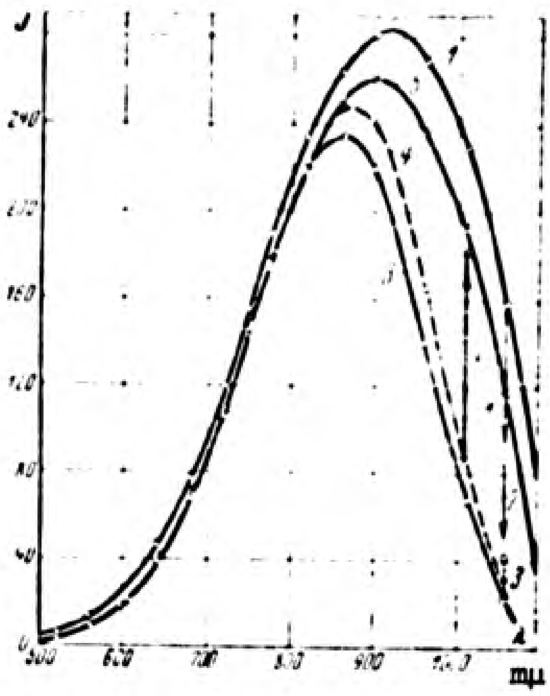

Fig. 8. Appearance of white-light aftereffect during variations in photocathode sensitivity with time. Curve 1) initial spectral response; arrow 1) change in sensitivity at $\lambda = 1060$ mμ owing to white-light fatigue of cathode; arrows 2 and 3) further drop in sensitivity after light is turned off, leading to the response curve 3; 4) spectral response 24 hr after curve 3 was obtained; arrow 5 and curve 5) recovery of sensitivity over 6 hr 40 min period under less intense white illumination.

These unexpected results perplexed us greatly. The phenomenon was observed more than once, and with other phototubes. We had also called attention to this phenomenon in our earlier article [6]. In this case as well, there was no difference between the effects of infrared and white light. The data of Fig. 9, referring to a different TsV-4 phototube, serve as a clear illustration. In the experiments whose results are shown in Fig. 9, we used infrared derived from the total radiation of an incandescent lamp by means of the light filter mentioned previously. At the beginning of illumination, the phototube sensitivity began to rise, passed through a maximum, and again reached its initial value. (The initial segment of the curve is shown separately at the top of the figure in an enlarged time scale.) This part of the experiment lasted 5 hr 15 min, following which the light was switched off and was turned on only to obtain the points. It is clear from Fig. 9 that as with the white light used in the preceding series of experiments, infrared had a strong effect on the cathode, and the sensitivity continued to drop violently in the dark. After 21 hr, constant infrared illumination was again switched on. This case study shows that no "excitation" in the photocathode of the sort suggested by de Boer should be expected (the cathode was illuminated by infrared alone and held in darkness), but subsequent illumination caused the sensitivity to increase from 32.7 to 103 % of the initial value. When the light was turned off, there again was a drop in the sensitivity of the cathode held in darkness.

Thus certain processes sometimes continue in photocathodes even after they have been held in darkness - processes for which the light was merely a stimulus.

In general, these processes include more than just diffusion processes causing recovery of photocathode sensitivity; there are other, frequently overlapping processes at work. There is no way of explaining or understanding this on the basis of existing ideas as to the mechanism for fatigue of s-o-c photocathodes.

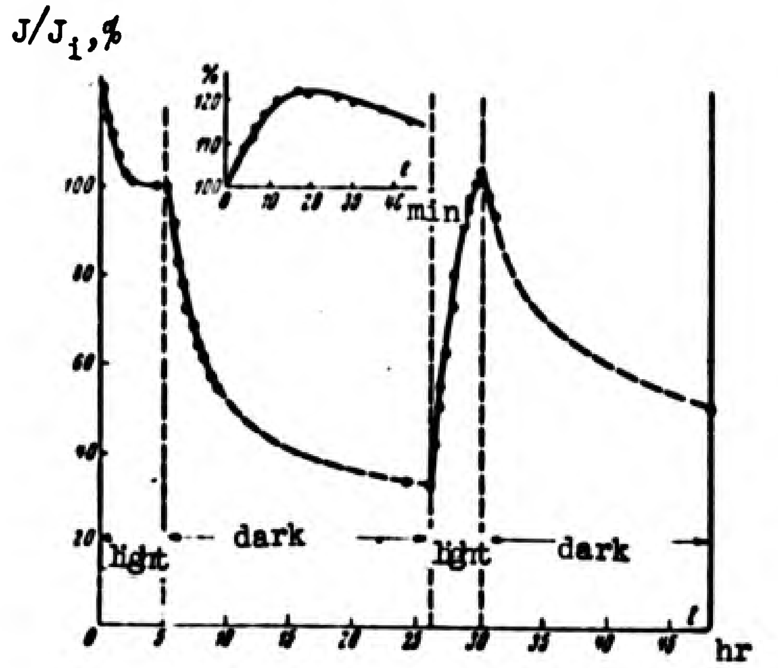

Fig. 9. Appearance of infrared aftereffect during photocathode sensitivity changes with time.

5. VARIATIONS IN SPECTRAL RESPONSE OF PHOTOCATHODES DURING FATIGUE

If fatigue could reduce, as some authors assume, solely to temporary removal of the cesium atoms adsorbed on the cathode surface, which are responsible for the longwave sensitivity of the photocathode, or to a change in the electronic work function, as others think, we would then observe merely a corresponding drop in longwave sensitivity in fatigue or a shift of the red branch of the cathode spectral-response curve toward the violet. It is usually assumed that all of this actually occurs. In fact, the observed variations are more complicated. If we look at the results shown in Fig. 5, we gain additional confirmation for this assertion. This type of result, however, is not the only one observed. Sometimes the relative sensitivity loss in the region very close to the threshold proves to be less than in a region further away toward the violet end. Sometimes we also observe a simple shift toward the infrared of the red cutoff for the photoelectric effect after the photocathode has been in operation.

It turns out that the cases noted by Yumatov [5] in which the drop in red sensitivity, which also determines the integral sensitivity of the cathode, is accompanied by increased sensitivity in the shorter-wave portion of the visible spectrum occur frequently. As our measurements have shown, however, fatigue by visible light has an especially marked effect on cathode sensitivity in the near ultraviolet. This is illustrated by Fig. 10, where curve 1 represents the spectral dependence of the ratio of the photocurrents: J_2 is the photocurrent after fatigue, and J_1 the photocurrent before fatigue, of an s-o-c cathode.

In order to ascertain whether this behavior is peculiar to the silver-oxygen-cesium photocathode or whether it is also inherent in the simpler Cs_2O-Cs system, we studied the effect of "fatigue" on the spectral response of simple oxygen-cesium (o-c) cathodes containing no metallic silver component. For each of these cathodes, we took the spectral-response curves at the beginning and then after operation under strong illumination. As an example, the same Fig. 10 shows results for three o-c cathodes as well (curves 2-4). We note that as for s-o-c cathodes, there are cases in which the red cutoff for the photoelectric effect moves toward shorter wavelengths after operation of the cathode, as well as cases in which it moves toward longer wavelengths. Here it is essential to emphasize that illumination with visible and infrared light (total radiation of incandescent bulb) leads to vigorous variations in cathode spectral response in the ultraviolet region, in the fundamental absorption region of o-c films [7].

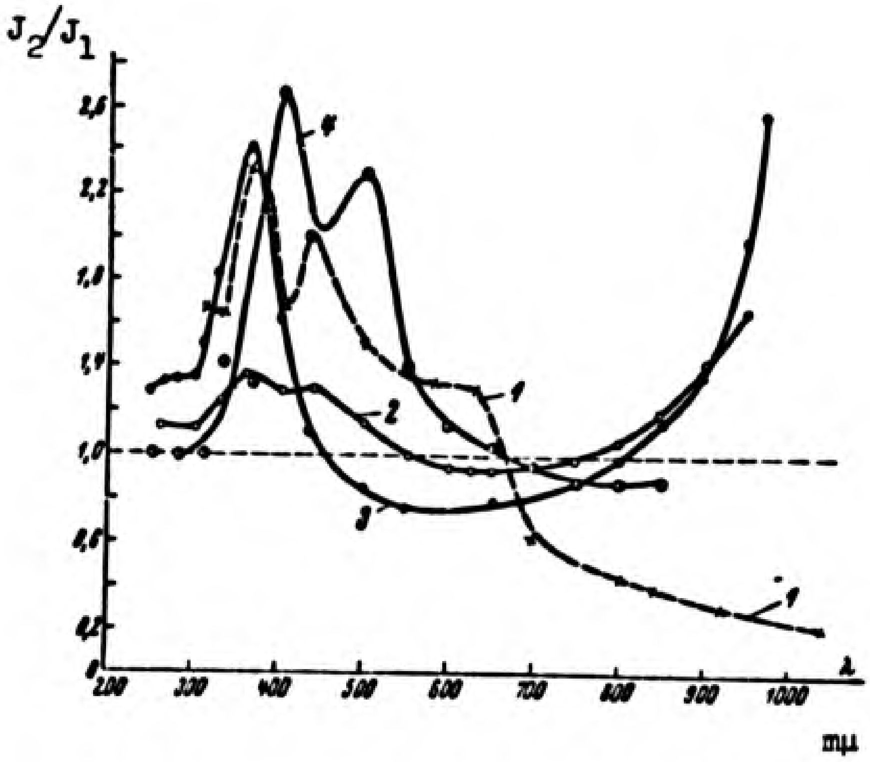

Fig. 10. Variation in photocathode spectral response after "fatigue" (J_1 - initial photocurrent; J_2 - photocurrent after "fatigue"). 1) Silver-oxygen-cesium photocathode; 2,3,4) oxygen-cesium photocathodes.

It is clear from these data that "fatigue" does not reduce simply to a variation in electronic work function and corresponding displacement of the red cutoff for the photoelectric effect. Phototube operation brings about a significant redistribution of spectral sensitivity. Cases are observed in which the integral sensitivity of a cathode remains nearly constant, but the spectral-response curve changes.

If we assume that variations in cathode sensitivity occur owing to changes in the work function, it is impossible to understand how an increase in the work function for photocathodes 1 and 4 (Fig. 10) could lead to the huge increase in sensitivity in the shortwave region, or the decreased work function of the cathode 3 to a drop in sensitivity in the 450-800 mμ band. All of this goes to show that changes in spectral response are associated not with the work function (more accurately not with the work function alone), but with certain other phenomena occurring in the photocathode. In the given case, it is natural to assume that these are volume-type phenomena. Since the change in properties is observed in a region in which we are concerned with the fundamental optical adsorption, which is associated with the lattice of the base material, it is necessary to consider the possible changes that may be produced by light in the base material of an o-c cathode, for example, the photochemical effect of light.

The fact that the variations in spectral response of o-c and s-o-c cathodes are of identical nature leads us to conclude that it is precisely the cesium oxide that is responsible for these phenomena in s-o-c cathodes as well. If light causes any changes in the properties of o-c films, it can produce the same changes in the films coating the silver grains in an s-o-c cathode, thus affecting the electronic emission from these grains. Thus to account for the physical processes producing and accompanying the so-called fatigue phenomenon, it is primarily necessary to undertake a comprehensive study of the physical processes occurring in o-c films.

CONCLUSIONS

1. Observations of the variation in contact potential of a fatigued photocathode show that the cause of fatigue is not the conversion of cesium atoms adsorbed on the cathode surface into adsorbed ions.

2. The effects of small added quantities of oxygen on cathode fatigue show that fatigue is not uniquely associated with cesium depletion of the cathode surface.

3. Photocathode fatigue is also observed when electrolytic processes in the cathodes have been completely inhibited either by the use of very thin cathode films or by illuminating the cathodes while drawing no current.

4. Cathode fatigue depends on the dimensions of the fatigued surface (within small values of these dimensions), which indicates the existence of an interaction between the working and nonilluminated parts of the cathode. Thus fatigue of a given part of the cathode also affects the sensitivities of neighboring nonilluminated parts.

5. Experiments with separate cathodes in one envelope and the sensitivity losses in dark areas of the cathode depending on their distances from the illuminated area show that interaction between separate areas does not take place through the cesium atmosphere in the envelope, but by way of diffusion-or migration-type processes.

6. Infrared illumination ($\lambda > 820$ mμ) has the same, and sometimes a greater fatigue effect on a photocathode than ordinary white light.

7. Interesting cases of light aftereffects have been observed; they appear as a continuing loss of cathode sensitivity in darkness, after the illumination has been switched off. This occurs with both white and infrared illumination.

8. Cathode fatigue, appearing as a drop in integral and longwave spectral sensitivity, is most frequently accompanied by a well-defined increase in shortwave sensitivity, in particular, in the fundamental optical absorption band

of Cs_2O. The result is a significant redistribution of cathode spectral response, owing chiefly to the oxygen-cesium component of the s-o-c cathode.

9. The results obtained show that fatigue cannot be reduced solely to a variation in the state of the cesium film adsorbed on the cathode and a change in electronic work function. Volume processes occurring under the action of light in the cesium oxide play an important role; this raises the necessity for a fundamental investigation of the physical properties of this substance in connection with the problem of fatigue.

The results given in this article have been described, in the main, in a dissertation by one of the authors [8].

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