The Fogging of Photographic Film by Radioactive Contaminants in Cardboard Packaging Materials

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(Received April 18, 1949)

Following the detonation of the experimental atom bomb at Alamogordo, New Mexico, on July 16, 1945, a radioactive contaminant was encountered in strawboard material used by the Eastman Kodak Company for packaging photographic sensitive films. This paper board was manufactured in a mill situated at Vincennes, Indiana, on the Wabash River. A run of strawboard, produced on August 6, 1945, showed this new and unusual type of radioactive contaminant. X-ray film packed with this board showed fogged spots after about two weeks' exposure.

Measurements of the contaminated spots of strawboard showed no alpha-activity but fairly strong beta-activity. Absence of alpha-activity ruled out naturally radioactive materials. Measurements of the beta-activity showed a maximum energy of about 0.6 Mev. and a half-life of approximately 30 days. Radiochemical studies of the active ash from the strawboard indicated that the material was of the rare earth series. The energy value and half-life of the beta-radiation are compatible with the isotope, Ce 141.

All studies point to the conclusion that the radioactive contaminant was an artificially radioactive material which found its way into the mill through the river water. The most likely explanation seems to be that it was a wind-borne fission product derived from the atom-bomb detonation in New Mexico on July 16, 1945.

EXPERIMENTAL evidence is given in the present paper to show that radioactive fission products resulting from the detonation of the first atomic bomb in July, 1945, in the state of New Mexico, were detected by photographic means on paper stock derived from raw products and manufactured in a paper mill at a location approximately 1000 miles from the point of detonation. The experimental observations on this problem were carried out between September and December, 1945, and to the author's knowledge represent the only case of detection of fission products at a ground station so remote from the point of detonation. The magnitudes of the radioactive effects observed were exceedingly minute and were detected at all only because of unusual circumstances that resulted in very great sensitivity. In fact, the main reason for presenting the data at this time arises from the thought that the results obtained, and the experimental procedure used, might be of interest to other physicists who are concerned with detection of minute quantities of radioactive materials.

This work had its origin in a new and unusual type of radioactive contamination encountered in August, 1945, in cardboard material used by the Eastman Kodak Company for packaging sensitive photographic materials. Before discussing the principal subject of this paper, it will perhaps be of interest to present first certain background information on prior experience of the Eastman Kodak Company with radioactive contaminants, which will serve to introduce the main results to follow.

Preceding the time of appearance of the above-mentioned contaminants and during the war, a great deal of the cardboard in this country became contaminated with radioactive materials as a result of the paper salvage program. Paper and cardboard, salvaged from war plants where radium instrument dials were prepared, became mixed with general paper stocks and as a result, specks of radioactive material could be found sporadically distributed in almost any type of packaging paper. Although the amount of material of this sort was very small, nevertheless it was sufficient to cause fogged spots on film left in intimate contact with it for a period of several weeks. Radioactive contaminants of this type were discovered immediately upon their appearance and the source and nature of these contaminants recognized. Alpha-count measurements of radon emanation from such contaminants identified them with radium paint originating from the greatly expanded radium-dial production of the war period. As a result of this trouble with radium contaminants, it was essential in the photographic industry to obtain a new and uncontaminated source of material for packaging sensitive goods. The Eastman Kodak Company found it necessary to select a mill and to control carefully the raw materials when its orders were produced. One of the mills selected for this purpose was located at Vincennes, Indiana, which is situated in southern Indiana on the Wabash River. Strawboard from this mill was used by the Eastman Kodak Company as interleaving stiffener board between sheets of film. Strawboard procured in this way was completely satisfactory for a period extending over many months. However, a run of strawboard produced on August 6, 1945, showed a new type of radioactive contaminant not hitherto encountered.

Effects of this new type of radioactive contaminant were first observed on film which had been packed in contact with the stiffener strawboard. When a piece of double-coated x-ray film, which had been in contact with a piece of the strawboard for a period of three weeks, was developed, the film showed many small exposed areas. These exposed areas ranged in number from ten to several hundred on a piece of film 14 by 17 inches in size. The exposed areas were of the order of 1-millimeter diameter and were quite dark when developed. In many cases, the radiation had passed

* Communication No. 1253.

TABLE I.

Date	Counter	Description of sample	Beta-count	Time	Beta-count hour
		Vincennes strawboard (August 6 run)			
Oct. 11	(*A*)	200 Bad Spots	310	30 min.	620
		200 Bad Spots	233	30 min.	466
		Background	223	30 min.	446
Oct. 20	(*B*1)	350 Bad Spots	160	1 min.	9600
		Background	80	1 min.	4800
Oct. 24	(*A*)	300 Bad Spots	450	30 min.	900
		300 Good Spots	294	30 min.	588
		Background	238	30 min.	476
Oct. 29	(*A*)	600 Bad Spots	580	30 min.	1160
		600 Good Spots	280	30 min.	560
		Background	260	30 min.	520
Nov. 6	(*A*)	600 Bad Spots		1 hr.	1205
		600 Good Spots		1 hr.	760
		Background		1 hr.	700
		Tama strawboard (September run)			
Nov. 6	(*A*)	500 Bad Spots		1 hr.	1224
		600 Good Spots		1 hr.	760
		Background		1 hr.	604

TABLE II. Calibration of Bad-Spot Samples with Uranium Acetate Samples.

Date	Description of sample	Gross weight	Beta-activity (*A*)	Beta-activity (*B*1)
Oct. 30	400μ g U acetate+silica	0.274 g	1062/hr.	157/min.
	300 Bad Spots (Vincennes)	0.274 g	1014/hr.	154/min.
	Background		488/hr.	86/min.

through several layers of film and cardboard, indicating a penetrating radiation.

The usual type of tests that had been used previously to analyze radioactive contaminants showed conclusively that the new radioactive contaminant was not radium; it was therefore necessary to devise new methods of analysis for it. In order to obtain sufficient concentration of the radioactive contaminants for direct study by physical instruments, it was necessary to concentrate the active material from the strawboard. To accomplish this, a technique was adopted of punching out small circular areas of the strawboard which lay directly behind the exposed areas on the film. By cutting out hundreds of these spots from the strawboard, samples were obtained having a much higher concentration of the radioactive contaminants. These batches of strawboard spots were then ashed by heating in a porcelain crucible at a temperature of 900°C to reduce further the bulk of material to be dealt with. Using material prepared in this manner, measurements of beta-activity and alpha-activity were carried out. All samples of ashed strawboard corresponding to the exposed spots on the film showed a fairly strong beta-activity, easily measurable above the background, while samples of ashed strawboard not corresponding to spots on the film showed practically no beta-activity. Samples of ashed materials measured for alpha-activity gave no detectable effect above the background. These results strongly indicated that the new type of radioactive contaminant encountered was not one of the naturally radioactive elements—uranium, thorium, or actinium—since all of these elements emit alpha-rays in addition to the beta-rays. One other possible source of the radiation considered was potassium, which is a naturally radioactive element giving only beta-rays. However, potassium was ruled out on the basis that the large amounts of this material required to produce the observed beta-activity could be detected readily by chemical analysis. In order to rule out the possibility that alpha-radiation due to naturally radioactive elements was present but below the measurable limit, a synthetic sample of uranium acetate in inert powder was made up to match in total weight and beta-activity a sample of the contaminated ashed strawboard, and measured for alpha-activity. This synthetic sample gave easily measurable alpha-activity.

Measurements of the penetrating power of the beta-rays showed that about 0.2 g/cm^2 of aluminum was required to stop the highest energy beta-particles. The half-life period of decay of the contaminant was measured photographically between October and December, 1945, and was found to be of the order of 30 days though the decay curve was not truly exponential. Strawboard made in August, 1945, at Vincennes, Indiana, that had shown strong activity initially had dropped to a point of considerably less activity photographically by January, 1946. However, exposure of film to this strawboard for two months still gave strong effects at this time.

In September, 1945, production of similar strawboard was commenced at a mill in Tama, Iowa, located on the Iowa River. Strawboard made at this mill also was found to exhibit the same type of radioactive contaminant as that found in the strawboard manufactured at Vincennes, Indiana. This mill is situated several hundred miles away from the Indiana mill and derives its water from a different watershed.

EXPERIMENTAL RESULTS

Beta- and Alpha-Activity

The experimental results obtained during the study of this problem are presented in the Tables I to VI. The samples of strawboard spots used for the activity measurements are designated, respectively, as "bad spots" and "good spots." A bad spot consists of a $\frac{3}{16}$-inch diameter area punched out of the strawboard at a point directly behind a fogged spot on an adjacent piece of film. A good spot corresponds to a similar circular area punched out of the strawboard at a point *between* fogged spots on the film. In all cases, the spots of strawboard were ashed before measurements of activity were carried out.

The measurements of beta-activity were made with two different Geiger counter tubes. The first tube (*A*) was small in volume and had low sensitivity, as indicated by the low background count of about 8–10 counts per minute. This tube and its associated scale-of-2 circuit were far from elegant, but gave fairly consistent readings. The second tube (*B*) was a large volume tube, highly sensitive, and when unshielded gave a background count of about 80 counts per minute. When this tube was shielded with lead, its background count dropped to about 30 counts per minute. When unshielded, it is referred to as (*B*1), and when shielded, as (*B*2). The tube *B*, with its associated scaling circuit, was kindly placed at the writer's disposal by Dr. William Bale, of the University of Rochester Medical School, and measurements were made on these instruments with the co-operation of Dr. Bale, who was consulted frequently on this problem.

The alpha-counts on the samples were made on an air chamber alpha-counter which was also furnished by Dr. Bale. This alpha-counter was connected to a scaling circuit for recording counts, but the counts were also observed visually on the screen of a cathode-ray oscillograph in all cases to make sure there was one-to-one correspondence between alpha-counts and registered pulses.

Table I shows beta-counts made on a number of different samples of ashed spots selected from different samples of the contaminated strawboard. The first five samples were from strawboard manufactured at Vincennes, Indiana, and the sixth sample was from strawboard made at Tama, Iowa. It may be seen that measurements showed slightly higher than background activity for good spots, and an activity of about double background for the bad-spot samples. Also, it should be noted that the beta-activity of the Tama strawboard was about the same as that of the Vincennes strawboard.

Table II shows the results of calibration measurements of beta-activity of Vincennes strawboard spots against beta-activity of uranium acetate dispersed in silica powder. In order to determine the approximate amount of uranium acetate necessary to give the same beta-activity as a sample of 300 bad spots of strawboard, trial and error mixtures of uranium acetate with inert silica powder were made up and tested until a sample was obtained having the same gross weight and beta-activity as the sample of ashed spots. As may be seen, a mixture of silica powder and uranium acetate was obtained whose weight (0.274 gram) and beta-activity were closely the same as those of the comparison sample of ashed strawboard spots, this comparison sample of known radioactivity having about the same self-absorption characteristics as that of the ashed strawboard spots. This was necessary for alpha-particle measurements where it was feared that self-absorption might cut down the alpha-particle emission strongly.

TABLE III. Photographic alpha-count.

Date	Description of sample	Relative count	Relative no. of alpha-tracks
Oct. 29	0.2 g U acetate+silica	1	16.1
	0.2 g Bad-Spot Ash	1	2.3
	0.2 g Good-Spot Ash	—	1

TABLE IV.

		Alpha-Activity (Vincennes)		
Date	Description of sample	Gross weight	Beta-count *B*-1	Alpha-count
Oct. 30	400μ g U acetate+silica	0.274 g	157/min.	14.3/min.
	300 Bad Spots	0.274 g	154/min.	1.6
	Background	—	86/min.	2.3
		Alpha-Activity (Tama)		
Date	Description of sample	Gross weight	Beta-count *B*-2	Alpha-count
Nov. 26	400μ g U acetate+0.8 g Inert Strawboard Ash	~1 g	84.5/min.	4.6/min.
	500 Bad Spots	~1 g	87.1/min.	1.5
	Background	—	30.5	1.3

TABLE V. Activity of Vincennes and Tama strawboards.

Date	Description of sample	Gross weight	Beta-count (*B*-2)	Alpha-count
Nov. 7	400μ g U acetate+silica	0.274 g	385/5 min.	60/5 min.
	Tama Bad Spots	0.1747	237	3
	Vincennes Bad Spots	0.1747	183	6
	Background	—	128	4

With samples of the same weight and beta-activity, it was felt that alpha-measurements from the two samples would give strictly comparable values of emission of alpha-particles from the two types of materials.

Tables III, IV, and V give data on the measurements of alpha-activity from the ashed strawboard spots. Table III shows results obtained by a photographic method carried out by Dr. W. West of these Laboratories. Dr. West made up equal weight samples of uranium acetate and silica powder, bad-spot ash, and good-spot ash. Here again the sample of uranium and the sample of bad-spot ash were adjusted to equal beta-activity. For measurement of alpha-activity, these samples were placed in contact with one of the Eastman nuclear-track plates and left for a sufficient time to give a measurable number of alpha-tracks from the uranium sample. Tracks per unit area obtained from the three comparison samples were counted and the relative numbers are shown in Table III. It may be seen that the alpha-activity of the uranium sample (of equal beta-activity) was seven times as strong as that of the bad-spot sample. While the alpha-activity of the bad-spot sample is about twice that of the good-spot sample, the statistical error here was such that this value has only slight significance and the only conclusion to be drawn

TABLE VI. Beta-count of residue from gallon batches of Wabash River water.

Date	Description of sample	Time of count	Beta-count (A)
Nov. 2	(1) Residue	10 min.	12.1/min.
	Background	10 min.	11.4/min.
Nov. 2	(2) Residue	30 min.	12.1
	Background	30 min.	11.0
	(3) Ashed Residue	30 min.	11.6
	Background	30 min.	11.2

is that the two samples had approximately equal activity.

Table IV gives the beta- and alpha-counts made with electrical pulse counters on Vincennes and Tama bad spots *versus* the synthetic uranium acetate samples. In the first part of Table IV, the beta- and alpha-counts of a uranium-plus-silica sample are compared with the counts from the ash of 300 bad spots of Vincennes strawboard of the same weight. In the second part of Table IV, the beta- and alpha-counts of a uranium acetate plus inert strawboard ash are compared with the counts from the ash of 500 bad spots of Tama strawboard. In both cases, it is seen that, whereas the beta-activities of the synthetic and bad-spot samples are closely equal, the alpha-particle count for the synthetic samples of uranium acetate are much higher, being well above the limit of experimental error. In fact, the alpha-count from bad-spot ash was in no case found to be measurably above background. These results are considered to be strong evidence for the fact that the radioactive contaminant in the strawboard was not one of the naturally radioactive elements, such as Ra, Th, or Ac, all of which are alpha-emitters.

Table V shows beta- and alpha-counts made for different samples of Tama and Vincennes bad-spot ash compared with similar counts for the uranium acetate-plus-silica samples. While the gross weights of these samples are not equal, nevertheless, qualitatively it may be seen that the alpha-activity of the bad-spot ash is not significantly above the background, whereas the alpha-activity of the uranium sample of comparable beta-activity is well measurable above the background.

Table VI gives results of beta-activity on samples of residue from one-gallon batches of Wabash River water and it may be seen that the counts here are not significantly above the background, though in every case the residue count was slightly higher than background. It is felt that this method of testing the water was too insensitive for picking up the radioactive material. On the other hand, when the river water was filtered through the strawboard during manufacture, material particles to which the active material was adsorbed, were filtered out, thereby giving active specks in the strawboard which could and did exert a photographic effect on film held in contact with the speck for several weeks. This constituted a very sensitive method for detecting active specks of material. Photographic registration of the specks made it possible to punch out active spots of cardboard and thereby effect a further multifold concentration of the material for study by electrical means.

Energy of Beta-Radiation

Measurement of the penetrating power of the beta-radiation was carried out by the method of aluminum absorption and Geiger-counter registration. In Fig. 1 are shown measurements made on a sample of ash from 600 bad spots of the Vincennes strawboard. The activity of the contaminant had died down to some extent by the time these measurements were made, which accounts for the low readings. The values plotted in Fig. 1 are beta-counts per ten minutes above the cosmic-ray

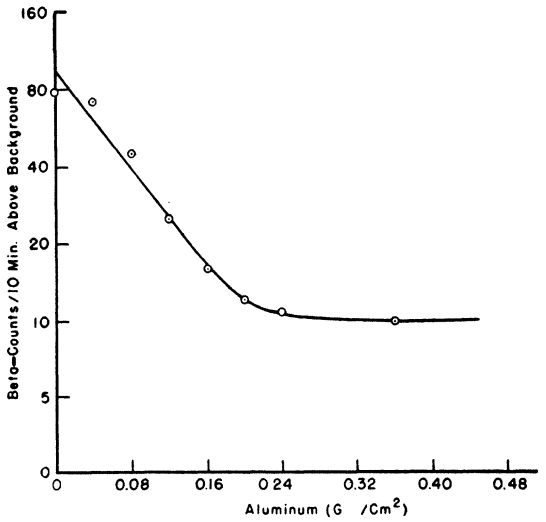

FIG. 1. Absorption measurements on beta-radiation by aluminum absorption and Geiger tube measurements for 600 bad spots of Vincennes strawboard.

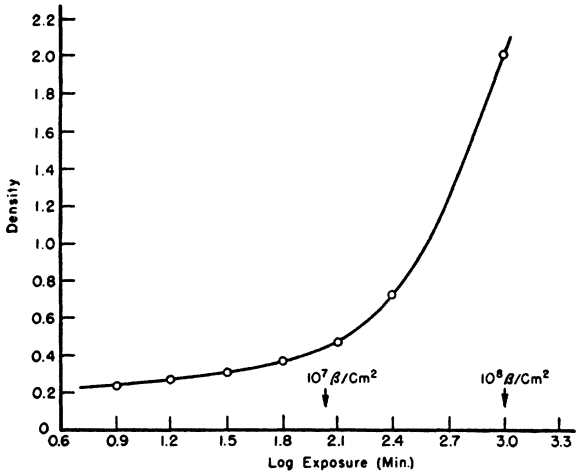

FIG. 2. Calibration characteristic curve for x-ray film exposed to beta-rays of uranium.

background. The abscissa values are given in terms of grams per square centimeter of aluminum. From these measurements it was apparent that there was a low gamma-ray background from the contaminant that was not stopped by a considerable thickness of aluminum. It may be seen that the toe of the curve, corresponding to the highest-energy beta-particles, falls at approximately 0.2 gram per square centimeter. This gives a maximum energy value of the beta-radiation of about 0.6 Mev.

Half-Life of Beta-Activity

The half-life of the beta-activity was measured by photographic methods. In brief, the method consisted in selecting a large piece of cardboard containing several strongly active specks of the contaminant, and exposing a piece of x-ray film to these specks for successive two-week periods over a total time of about two months. Photographic densities produced by these successive exposures showed a steady decline. By comparing the successive densities obtained in a characteristic exposure curve, such as that shown in Fig. 2, for beta-particle exposure, it was possible to read from the abscissa values the declining intensity values of the exposing radiations with time. These intensity values, plotted as a function of time, are shown by the experimental points in Fig. 3. A smooth curve drawn through these points shows an average half-life decay period of approximately thirty days.

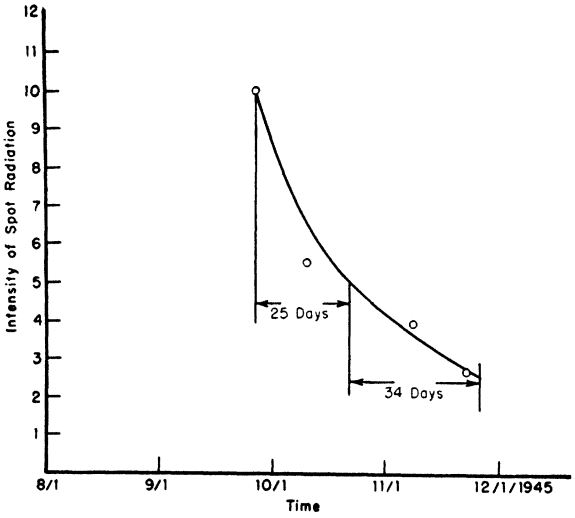

FIG. 3. Decay curve of beta-radiation from radioactive contaminant in strawboard (Vincennes, August 6, run).

DISCUSSION OF RESULTS

The experimental results obtained seem to show definitely that the radioactive contaminant encountered gives beta- and some gamma-radiation but no alpha-activity, indicating an artificially radioactive material. The large amount of material that must have been involved to show up over a period of several months in the water supply for the strawboard mill would seem to rule out the possibility that the material came from a moderate source of radioactive materials, such as a cyclotron. A check on possible sources of contamination of the river water from such nuclear laboratory by-products disclosed no such means of contamination. It was established that the contaminant was not carried by the straw going into the strawboard, since a run of strawboard made using straw that had not been in the field, but stored in warehouses for a considerable time prior to the trouble, showed the radioactive contaminant. Further evidence that the active material came from the river water was indicated by the fact that following each heavy precipitation in this locality, stronger activity occurred in the strawboard. This was attributed to the material being washed into the river water during heavy precipitation periods.

The most likely explanation of the source of this radioactive contaminant appears to be that it consisted of wind-borne radioactive fission products derived from the atom-bomb detonation in New Mexico on July 16, 1945. The evidence that the material was an artificially radioactive isotope, the occurrence of the same unusual type of contaminant in strawboard manufactured in two widely separated areas (Vincennes, Indiana, and Tama, Iowa) at the same time, and the timing of the appearance of this material immediately following the bomb detonation, are thought to lend strong support to the above tentative explanation. It is readily conceivable that radioactive atoms projected into the stratosphere, and carried by winds, could collect on dust particles and be brought down by precipitation and be washed into streams. It is well known that dust projected into the stratosphere from volcanic explosions has been scattered over wide areas in this way. The radioactive contaminants could have found their way into the products of the strawboard mills either by settling on the straw in the fields or through the river water used by the mills in the manufacture of the strawboard. Of these two possibilities, the latter seems to be much the more likely, since strawboard made from straw that was not in the field at the time of the explosion but stored indoors showed the contamination.

The measurements of the energy and half-life of the beta-particle radiation of the radioactive contaminant in the strawboard were found to be 0.6 Mev and thirty days, respectively. These values correspond closely to those for the radioactive isotope, Ce 141, which is one of the more prolific fission products of the atom bomb.

A simple calculation shows that if 50 grams of Ce 141 were formed in an atomic explosion, there would be approximately 2.2×10^{23} atoms. Since the area of the United States is about 7.5×10^{18} sq. mm, there would be about 0.3×10^{5} atoms for every square millimeter. Allowing for some local concentrations of these atoms such as that occurring under the circumstances sur-

rounding the manufacture of strawboard from river water, this number could be increased severalfold. The sensitivity of x-ray film is such that 10^5 beta-particles per square millimeter is just sufficient to produce a visible density. Therefore, it would appear readily possible from a theoretical standpoint for short-life fission products from the bomb to produce the photographic effects described above.

After carrying out the physical study just described of the radiations from the radioactive contaminants, a limited amount of radiochemical analysis was carried out to see if it were possible to follow by tracer techniques the chemical nature of the contaminant. These experiments were carried out by Mr. A. E. Ballard, of these laboratories. Enough work was done to show that the active material was probably one of the rare earth series. However, because of the small amount of material in the sample and because of the limited equipment available for such experiments at the time, a complete analytical determination of the radioactive isotope was not possible.

It is realized by the author that the measurements carried out in the study are not so complete nor so accurate as might be desired. However, the work was carried out hurriedly, and somewhat blindly at first, when it was important to get quick survey measurements. By the time sufficient knowledge was gained to make it possible to conduct the experiments more intelligently, the activity had died down to a considerable extent. In spite of this fact, however, it is thought that the results obtained are sufficiently good and of enough general interest to physicists to warrant their publication.

In conclusion, it is to be emphasized that the amounts of material considered throughout the discussion are exceedingly small and were observed only because of an accidental set of circumstances that proved particularly favorable for the detection of the contaminating substances. The river water carrying the substances was derived from a large area and specks of dust were filtered out of this water onto strawboard during passage of large quantities of water through the strawboard pulp. Photographic film placed in contact with this board for a period of weeks registered the radiation from the concentrated specks of the contaminant. The specific location of the radioactive specks through photographic registration made it possible to obtain large numbers of strawboard spots containing active specks and thereby to increase the concentration further. By ashing down these strawboard spots, the active material was concentrated sufficiently to make direct study possible.

ACKNOWLEDGMENT

The author desires to express his appreciation to Mr. A. E. Ballard, of these laboratories, who participated in some of the experimental work outlined and was frequently consulted on this problem throughout the course of the work. In particular, the analytical radiochemistry experiments were carried out by Mr. Ballard and the preparation of all ashed samples were made under his direction.

It is a pleasure to thank Dr. William Bale, of the University of Rochester, for the kind loan of beta- and alpha-counting equipment during this study and for his generous advice on different aspects of this work.

The photographic alpha-particle counts presented in Table II were carried out by Dr. West, of these Laboratories, and the author wishes to record his thanks to Dr. West for the use of these data.

Finally, the author would like to express appreciation to Mr. Harold Crouch and his co-workers, in the Department of Manufacturing Experiments, at Kodak Park, for the large amount of work contributed on this problem. All contaminated strawboard used in the experiments was selected on the basis of the photographic tests made by this group, who also supplied all of the punchings of the strawboard spots.