

**Peculiarities Of Measuring The Alpha Particle Activity Of Flat Samples
Of Metals, Alloys And Powders Using Gas Flow Proportional Counters
Specifically the Model 1950 Manufactured by “Spectrum Sciences”, USA**

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ABSTRACT

Naturally occurring radioactive emissions from solder (Pb/Sn or Pb-free) in semiconductor devices may result in memory device malfunctions and possible consequential product liability. These “soft errors” occur randomly but do not cause permanent physical damage to the memory cell. The principal product developed to reduce the probability of soft error malfunctions in memory devices is low alpha lead/tin alloys and low alpha lead-free alloys.

As standards for determining alpha emitter emissions have not been adopted by the Semiconductor industry, proposed for discussion are actual testing methods and standards of measurement.

Of interest, and the subject of this research is the description(s) of testing procedures and measurement techniques for accurate surface alpha emission counts from flat metal and alloy material, metal alloy powders, tin and lead oxide powders. These materials are used, principally, in solder paste, solder spheres, electroplating anodes, and plating chemistry. Discussion is focused on test equipment, sample preparation, alpha counting techniques and the accuracy of alpha emission measurements.

INTRODUCTION

In 1978 there appeared the early discussions on casual failures in memory circuits caused by alpha particles released by the material in or within proximity of these circuits. Refer to the Literature Article (1) Advancing technology of chips, presupposes that the new devices will be even more sensitive to the influence of alpha particles. Refer to the Literature Article (2) For decreasing the frequency of soft error failures of integrated circuits, especially at the linewidth of less than 0.18 microns, it is necessary to have careful and exacting alpha emitter measurements of the materials used on, or in proximity to, the chips with respect to the level of impurities that are capable of releasing alpha particles.

For the alpha emitter measurement of materials used in microelectronics, the gas proportional counter, Model 1950 developed and produced by "Spectrum Sciences" (or "Alpha Sciences" USA) is widely used. This surface emission counter is intended for the measurement of the surface activity of samples with the area of up to 1000 cm² (28 cm x 35.5 cm). The range of energies of the registered particles is from 1 up to 10 Mev. The working background of the device according to the manufacturer's technical description is less than 5 α /hour

In this article there is a description of the procedures for the preparation and measurement of alpha emission of flat samples of metals and alloys, alloy powders, tin and lead oxide powders, and also the technique of processing measurement results.

2. PREPARATION OF SAMPLES

2.1. PROCEDURE FOR THE PREPARATION OF FLAT SAMPLES

There are a number of methods for preparing flat samples from metals and their alloys for measurement of the specific surface alpha activity in the Model 1950 gas proportional counter. Common sample preparation methods include sputtering films onto copper wafers, rolling the metal and casting flat-shaped ingots.

Manufacturing samples using the method of rolling and the method of sputtering on wafers from copper in vacuum melting showed their apparent defects. As a rule, when rolling metal, the surface alpha emission increased even for samples having the level of alpha emission of 0.02 cph/cm² and higher. After the re-melt of the samples, according to the procedure described below, the level of alpha emission returned to the initial level of alpha emitter quality and over time it did not change.

The sample preparation method of sputtering is rather costly, labor-consuming and is not always reproducible. Test samples made by sputtering, as all thin films, are active on the surface and are inclined to oxidation, even during brief storage duration and they need protection from the external atmosphere. Barring measurement of film samples, it is required to take into account factors of back-scattering and self-absorption of the wafers and films. It is therefore necessary to have the additional alpha measurement of the surface of wafers and careful physical measurement of the width of films.

The simplest and most reliable method of producing flat samples is the method of manufacturing ingots in the form of flat plates. It is also important to note that this operation, formation or casting, is the last operation after production of the metal or its alloy. When manufacturing samples by the method of casting, the probability of introducing impurities from the surfaces of the preparation equipment is minimal. Also, in this case, the probability of a non-uniform distribution of the polonium impurity (because of its low concentration) is also small. Traces of polonium corresponding to the level of alpha emission, are basically received on the counter of the Model 1950 device. In the Literature, article [3] it is shown that in the process of lead solidification, the polonium solidifies at a constant rate prior to accumulating on the surface of the metal. The surface accumulation solidifies last. When measuring the alpha emission of this surface, it is possible only to overestimate the results of the alpha measurement. The

results of the measurement of alpha emission of various surfaces of one-and-the-same sample should differ by no more than 10% which testifies to the even distribution of the polonium in bulk. Usage of rather thick samples allows for the simplification of the procedure of processing of alpha measurement results..

Research on the influence of the test sample mould date, comparing many reference samples, on the level of alpha emission, showed that here there is no noticeable variation in secular equilibrium. The reference samples from one lot of metal or its alloy, having different mould dates, have virtually identical parameters of alpha activity.

Thus it is quite reasonable to manufacture samples by the casting method. The preparation procedure of samples for testing the level of alpha emission of a production lot of low alpha metal (alloy) is the following:

From a given lot, some ingots (for example from the lot of lead with the weight of 250 kg, three test ingots with a total weight of about 7.5 kg) are selected at random. The metal is re-melted (lead is re-melted on air at the temperature of 450C in the crucible made of stainless steel). Time of contact of the melted metal with the environment is no more than 20 minutes. Then the metal is poured, in a thin layer of 4 to 5 mm, into flat stainless steel molds. The ingots cool down to room temperature in a closed metal container which reduces, to a minimum, the volume of atmosphere contacting with the sample ingot. After cooling, the samples are packed in vacuum into polyethylene film. All the materials, with which the sample can contact (polyethylene film, the material of steel molds, the table, etc.) should be clean and should not contain radioactive impurities, including dust particles in the atmosphere containing radon gas. All operations are conducted in treated surgical gloves.

The sample ingots of metals or their alloys prepared for alpha emitter testing, are moved into a controlled room environment in which filtering and recirculation of air are provided and strictly maintained and the temperature and humidity are controlled. The room is a clean room, in which the basic technological processes of chip manufacturing (Literature article [4]) are executed.

2.2. PROCEDURE FOR THE PREPARATION OF SAMPLES FOR THE ALLOY, TIN AND LEAD OXIDE POWDERS

For alpha emitter measuring powders of alloys, oxides of tin and lead (with the size of the particles of 20 to 150 microns) a random sample of the powder, in the amount necessary for an even covering of all the surface of the Model 1950 powder tray (1000 cm²) with the layer of about 4 to 5 mm, is taken from the subject low alpha lot of the material. The tray is first carefully wiped with a napkin, wetted with distilled ethyl (isopropyl) alcohol. The clean tray is then placed in the Model 1950 installation to purge radioactive impurities (count rate of the device with the tray is measured for no less than 24 hours and should be different from the count rate of the installation without the tray (also measured for no less than 24 hours) by no more than 10 %). Along the whole top perimeter wall of the prepared tray, Scotch Tape (double stick) is stuck. The powder is uniformly poured and leveled with the help of carefully "wiped" palette-knife. Then the Scotch Tape protection layer is removed and the tray with the powder is covered with a

mylar film. The film should be uniformly stretched over the whole surface of the tray. The tray with the powder is packed into a clean polyethylene package and is moved into the clean room for preparation for alpha testing.

3. ALPHA EMITTER MEASUREMENT OF SAMPLES

3.1. OPTIMIZATION OF COUNTING EXPERIMENTS AND MEASUREMENT OF FLAT SAMPLES

Measurements of the level of alpha emission on Model 1950 gas proportional counter are measurements of the differences in count rates of the installation with a low alpha sample R_t [Rate of Total Count (sample + background)] and of the empty installation R_b (Rate of Background). The value of the rate of the total count of the installation with the powder sample is subject to distribution based on the law of the Poisson Distribution; and the kind of distribution depends on the preliminary items of information, which the researcher possesses prior to the measurement (Literature article [5]). Therefore, for receiving precise and authentic results, the measurement of materials should be conducted in several stages (no less than three). The first stage consists of determining the count rate of the sample. At this stage the count rate of the background is measured not less than 24 hours, the cleanliness of the tray is checked (the count rate of the installation with the tray measured for not less than 24 hours should differ from the count rate of the installation without the tray by no more than 10 %) and the value of the alpha count rate of the sample (for no less than 24 hours) is determined. The second stage is the determination of the alpha count rate of the sample. The third stage is checking the authenticity of the recorded level of alpha emission of the sample.

When holding the second stage of the alpha measurement of the samples, it is necessary to minimize the associated statistical uncertainty to optimize the counting experiment. The optimization of counting experiments is presented in the Literature articles [6,7]. According to Knoll (Literature article [6]) and practical experience using the Model 1950 installations, the minimum of the relative error of measurement of the alpha count rate of a sample is reached, if the full time of measurement is distributed between the total count rate and the background count rate in such a way that the following ratio is executed:

$$\frac{t_t}{t_b} = \sqrt{\frac{R_t}{R_b}} \quad (1)$$

where

- t_t – Time of Total Count (sample + background)
- t_b – Time of Background
- R_t – Rate of Total Count (sample + background)
- R_b – Rate of Background

The minimum full time “T” necessary for measurement of the material with the allowable relative error of measurement of 25 % corresponds to the following:

$$T = t_t + t_b = \frac{R_t + R_b + 2\sqrt{R_b \cdot R_t}}{0.0625 \cdot R_s^2} \quad (2)$$

Thus, for measurement of the alpha count rate of the Model 1950 with the sample and for measurement of the alpha count rate of the background, it is necessary to spend accordingly the following time t_t and t_b

$$t_t = \frac{R_t + \sqrt{R_b \cdot R_t}}{0.0625 \cdot R_s^2} \quad (3)$$

$$t_b = \frac{R_b + \sqrt{R_b \cdot R_t}}{0.0625 \cdot R_s^2} \quad (4)$$

Thus, at the second stage of the alpha measurement, first it is necessary to count the time t_t and t_b which is indispensable for testing the given sample. Then to measure the count rate of the empty Model 1950 installation and of the count rate of the installation with the sample during t_b and t_t or more, to determine the level of alpha emission of the sample, the root-mean-square deviation of the level of alpha emission (confidence interval of the level), relative root-mean-square measuring error of the count rate of the sample. The relative root-mean-square measuring error of the count rate of the sample should be less than 25 %, if the error is more than 25 %, it is necessary to continue the measurement. At this stage of measurement it is necessary to check the permanency or stability of the background of the installation, that is to measure the count rate of the empty installation after the made measurements. The count rate of the installation (background count) after the measurements are made should not differ from the count rate before measurements by more than 10%.

For example, at the first stage of measurement the count rate of the installation with the sample $R_t = 4.5 \alpha/\text{hour}$ (during 24 hours of measurement on the installation with the sample 108 impulses were registered), the count rate of the background $R_b = 3.2 \alpha/\text{hour}$ (during 24 hours of measurement on the installation without the sample, 77 impulses were registered). Then the count rate of the sample $R_s = R_t - R_b$ (the net Rate = sample rate) will be $1.3 \alpha/\text{hour}$. The activity of the sample with the area of 1000 cm^2 at the counting efficiency of 84 % will be 0.00155 cph/cm^2 . It is defined according to the formula

(5)

$$\alpha = \frac{R_t - R_b}{A \cdot F}$$

where

A- area of the sample, cm².

F- counting efficiency of the Model 1950 installation. As a rule, counting efficiency is established on the alpha counter of Model 1950 at about 84 %; with an installation of lower efficiency, the productivity of the installation is reduced and the possibility of errors in the determination of the level of alpha emission can occur because casual fluctuations are increased. With an installation of higher performance (efficiency) there is a possibility to register not only alpha particles, but also beta particles.

The minimum full time “T” necessary for measurement of this material at the second stage of measurement and defined according to formula (2), will make more than 145 hours. For measurement of the count rate of the background R_b it is necessary to count 67 hours (estimated time t_b according to formula (4) is less, but for receiving greater accuracy it should be approximated to the higher number of hours). For measurement of the count rate R_t it is necessary to count 79 hours. If at the second stage of measurement during t_b=67 hours 214 impulses are registered, during t_t = 79 hours 355 impulses are registered, the level of alpha emission of the sample will be 0.00155 cph/cm². The root-mean-square deviation of the count rate of the sample, defined according to the formula,

$$\sigma_0 = \sqrt{\left(\frac{R_t}{t_t} + \frac{R_b}{t_b} \right)} \quad (6)$$

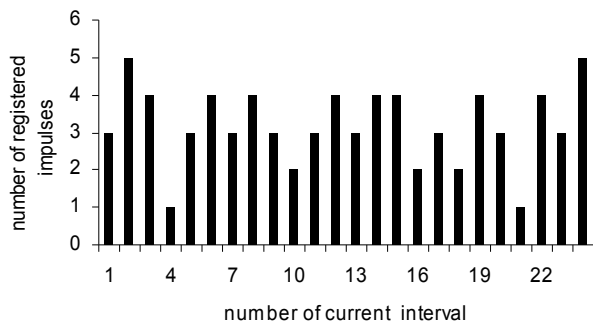
will be accordingly 0.32 or 32%.

The root-mean-square measuring error of the count rate of the sample is defined under the formula

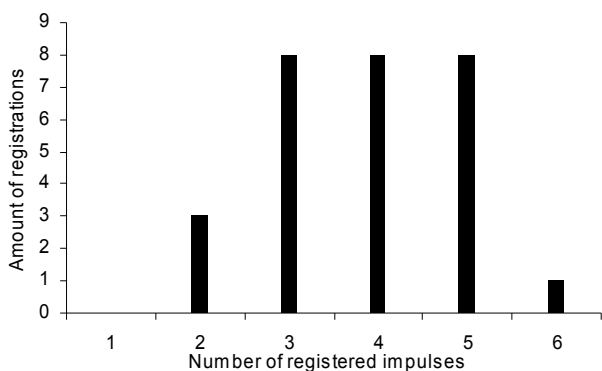
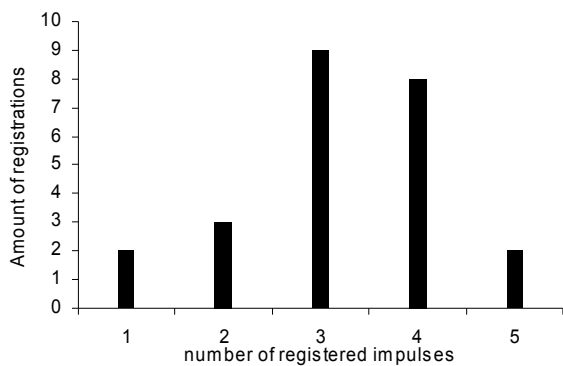
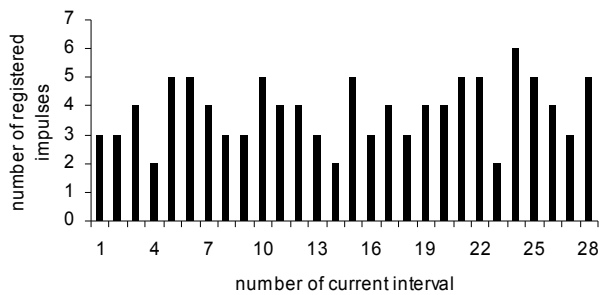
$$\sigma = \frac{\sqrt{\left(\frac{R_t}{t_t} + \frac{R_b}{t_b} \right)}}{R_t - R_b} \quad (7)$$

corresponds 25%. It means that the activity of the measured sample is in the interval from (0.00155 - 0.00038 cph/cm² up to (0.00155 + 0.00038) cph/cm² with the probability of 68 % and the measurements are conducted more or less correctly. Picture 1 Histograms corresponding to the first [a); b)] and second [c); d)] stages of measurement of the flat sample of metal with the activity of 0.00155±0.00038 cph/cm² are shown.

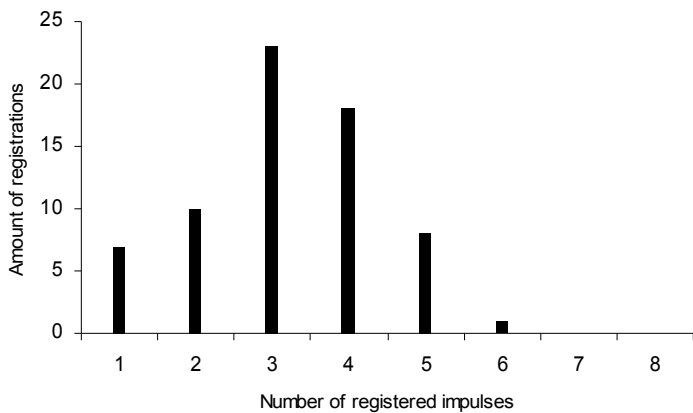
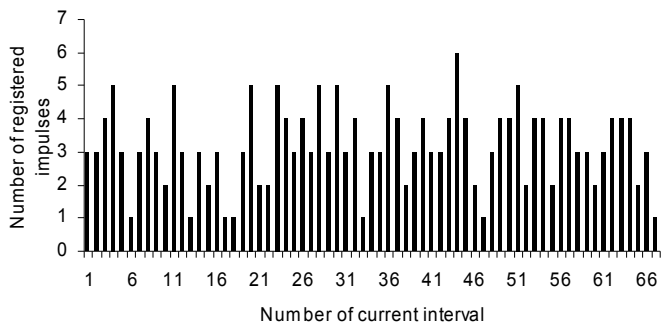
a)



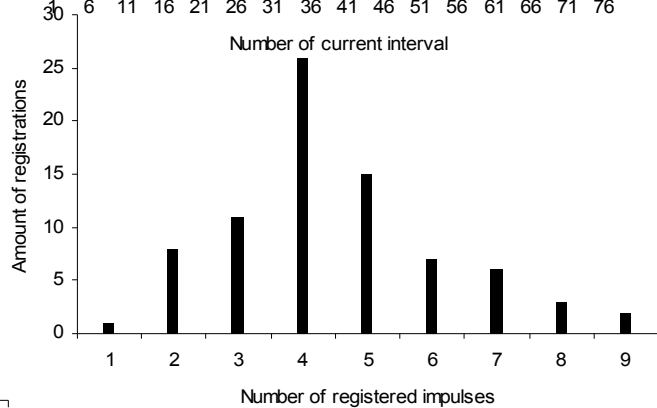
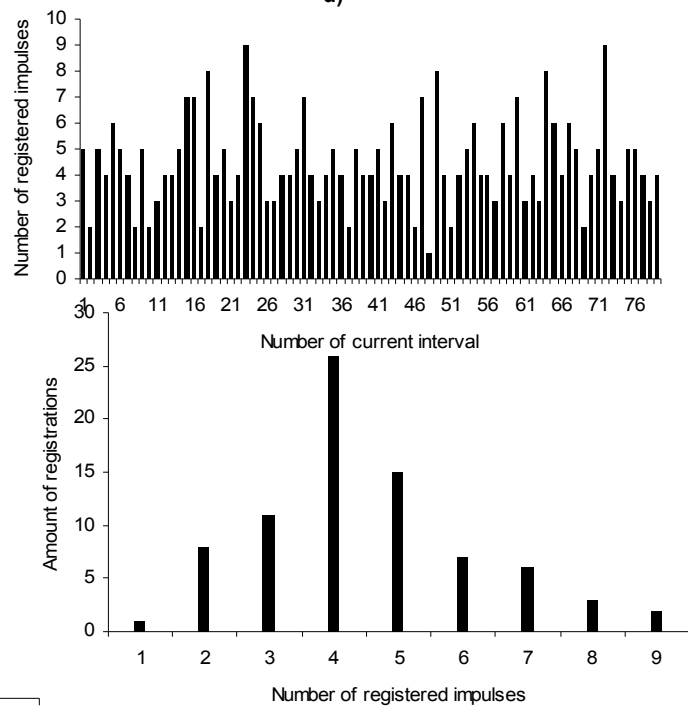
b)



c)



d)



The third stage - the check of accuracy of the received level of alpha emission of the sample - is the repetition of the procedure of the second stage of measurement after the passage of some time. This stage allows to exclude the possibility of an error of the first kind – as a result of the measurement the determination of the fact that the sample is active, when actually it is not, and the error of the second kind - inability to detect the radioactivity of the sample as a result of the measurement, though actually the sample has certain activity. Besides this stage allows to determine whether the secular radioactive equilibrium of the sample is infringed, whether the activity of the sample will increase during its usage.

Table 1 gives the time of measurement of the background and the time of measurement of the samples with the background, which is necessary to spend for having the accuracy of measurement of 25 % for samples with the activity of 0.001 (cph/cm²), 0.002 (cph/cm²), 0.003 (cph/cm²), 0.004 (cph/cm²), 0.005 (cph/cm²), 0.01 (cph/cm²) and the area of 1000 cm², 900cm², 500cm².

Table 1
Time of measurement of the count rate of the empty installation and of the installations with samples using Model 1950 installation with the counting efficiency of 84 %

Back-ground (counts/Hr)	Area (cm ²)	Alpha flux 0.001 $\alpha/\text{cm}^2 \cdot \text{Hr}$		Alpha flux 0.002 $\alpha/\text{cm}^2 \cdot \text{Hr}$		Alpha flux 0.003 $\alpha/\text{cm}^2 \cdot \text{Hr}$	
		t_b	t_t	t_b	t_t	t_b	t_t
		Hr	Hr	Hr	Hr	Hr	Hr
3	1000	145	164	39	48	18	25
	900	178	200	47	58	22	29
	500	563	601	145	164	67	79
4	1000	191	210	50	60	23	30
	900	235	256	61	72	29	36
	500	745	783	191	210	87	100
5	1000	236	255	62	71	28	35
	900	291	312	75	86	35	42
	500	926	964	236	255	107	120

Back-ground (counts/Hr)	Area (cm ²)	Alpha flux 0.004 $\alpha/\text{cm}^2 \cdot \text{Hr}$		Alpha flux 0.005 $\alpha/\text{cm}^2 \cdot \text{Hr}$		Alpha flux 0.01 $\alpha/\text{cm}^2 \cdot \text{Hr}$	
		t_b	t_t	t_b	t_t	t_b	t_t
		Hr	Hr	Hr	Hr	Hr	Hr
3	1000	11	16	7	11	2	4
	900	13	18	9	13	3	5
	500	39	48	26	33	7	11
4	1000	14	19	9	13	3	5
	900	17	22	11	15	4	6
	500	50	60	33	41	9	13
5	1000	17	21	11	15	3	5
	900	20	26	15	18	4	6
	500	62	71	40	48	11	15

If the estimated time necessary for measurement of the alpha activity of the sample is long, it is necessary to conduct alternate measurement of the count rate of the empty

installation, the count rate of the installation with the sample, then again count rate of the empty installation, count rate of the installation with the sample etc., because even minor fluctuations of the background there is a possibility of errors of measurement. At the alternate count the activity of the material is counted under the formula:

$$\alpha_2 = \frac{\left(\frac{\sum_{i=1}^n R_{ti} \cdot t_{ti}}{\sum_{i=1}^n t_{ti}} - \frac{\sum_{j=1}^k R_{bj} \cdot t_{bj}}{\sum_{j=1}^k t_{bj}} \right)}{A \cdot F} \quad (8)$$

where

n - The number indicating how many times the measurement of the sample with the background was conducted,

k- the number indicating how many times the measurement of the background was conducted.

The root-mean-square deviation of the count rate of the sample σ_0 , in this case is defined under the formula

$$\sigma_0 = \sqrt{\frac{\sum_{i=1}^n R_{ti} \cdot t_{ti}}{\left(\sum_{i=1}^n t_{ti} \right)^2} + \frac{\sum_{j=1}^k R_{bj} \cdot t_{bj}}{\left(\sum_{j=1}^k t_{bj} \right)^2}} \quad (9)$$

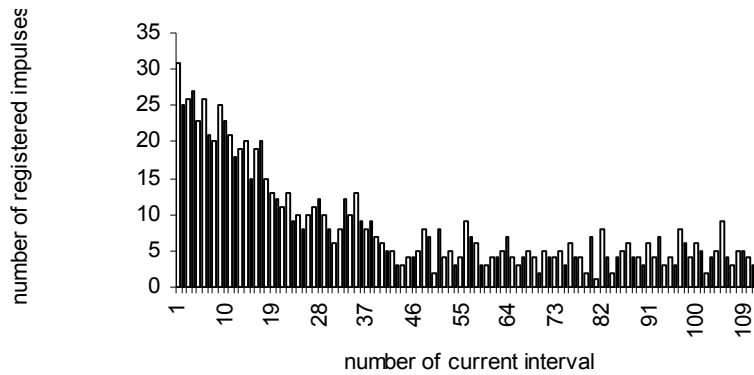
The root-mean-square measuring error of the count rate of the sample

$$\bar{\sigma} = \frac{\sigma_0}{\frac{\sum_{i=1}^n R_{ti} \cdot t_{ti}}{\sum_{i=1}^n t_{ti}} - \frac{\sum_{j=1}^k R_{bj} \cdot t_{bj}}{\sum_{j=1}^k t_{bj}}} \quad (10)$$

For more precise measurements it is necessary to increase the time of measurement of the count rate of the empty installation and of the installations with the sample, here it is necessary to take into account that the minimum of the relative error of measurement is reached, if there is the ratio (1) and the root-mean-square measuring error is considerably less than 25%.

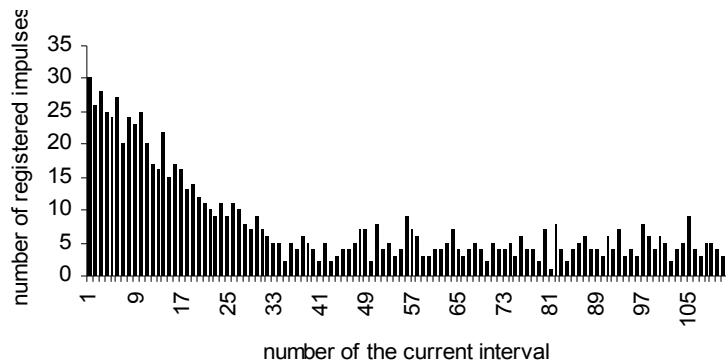
3.2. MEASUREMENT OF POWDERS OF ALLOYS, OXIDES OF TIN AND LEAD

Before measurement dispersible samples are covered with a mylar film. In Picture 2 a characteristic histogram of powders is presented.



Picture 2. Histogram of the powder of the tin oxide with the activity of less than 0.002 cph/cm^2 (count rate of the installation before measurement is $2.99 \alpha/\text{hour}$, after measurement $3.3 \alpha/\text{hour}$, count efficiency of the installation 0.84)

In Picture 3 the histogram of a flat sample of metal with the activity of $0.00155 \pm 0.00038 \text{ cph/cm}^2$ is presented, also covered with a mylar film. From the given histograms it is visible that at the beginning of measurements there is a fall of the level of alpha emission, further the indications of the level of alpha emission fluctuates about the average value.



Picture 3. Histogram of the flat sample of metal with the activity of $0.00155 \pm 0.00038 \text{ cph/cm}^2$ covered with the mylar film before measurement (count rate of the installation before measurement is $3.3 \alpha/\text{hour}$, after measurement $3.1 \alpha/\text{hour}$, count efficiency of the installation 0.84)

From the histogram corresponding the sample with the activity of $0.00155 \pm 0.00038 \text{ cph/cm}^2$, covered with the mylar film (Picture 3) it follows that within the first 34 hours there was a replacement of a residual atmosphere from under the film (preparation of the installation for measurement in this case does not last for 4 hours corresponding to the description of the installation, but for 34 hours).

At measurement of samples covered with a mylar film the time of the fall of the level of alpha emission depends on the degree of tension of the film above the sample, of the dispersibility value of the powder, the time during which the powder was in air and the level of alpha emission itself. That is at measurement of samples covered with a

mylar film, as a rule the preparation time of the installation for measurement is more than 34 hours. This preparation time should be taken into account when calculating t_i .

CONCLUSIONS

The peculiarities of measurement of alpha activity of flat samples of metals and their alloys on proportional counters, specifically the Model 1950 manufactured by "Spectrum Sciences" ("Alpha Sciences"), USA, are in the correct timing of measurements.

To increase the alpha measurement accuracy it is necessary to increase the time of measurement of the count rate of the background and the count rate of the sample. It is necessary to determine the background level of the installation not only before measurement, but also after it to reduce to the minimum the errors possible because of the fluctuations of the background activity over time.

At measurement of dispersible materials it is necessary to take into account, that the usage of a mylar film increases the preparation time of the counter chamber for alpha measurement. At calculating the indispensable time of measurement t_i it is necessary to take into account the duration of preparation of the chamber for measurement.

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