RADIOACTIVE MEASURING TECHNIQUES OF ION ADSORPTION, I

METHODS FOR THE DETERMINATION OF THE RADIOACTIVE ISOTOPES I-131 AND K-42 IN THE PRESENCE OF EACH OTHER

By

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Introduction

In the investigation of ion adsorption on the face of (111) orientation of germanium single crystals from electrolyte solutions of medium concentration (10^{-1} to 10^{-3} mole/lit.), quantities adsorbed had to be determined on the adsorbent (on a small germanium plate of $15 \times 15 \times 0.5$ mm), on account of the small adsorbent surface available (0.42 cm^2). The radioactive tracer techniques used and experimental conditions have been reported in a previous communication (1). In the study of ion adsorption phenomena the simultaneous measuring of the adsorption of both cations and anions becomes sometimes necessary. There are several possibilities to jointly determine by radioactive indication I—131 and K—42, the radioisotopes most often met in our investigations, because, as can be seen From Table 1, there is a substantial difference in the energy of the β- and γ-radiation, of the two isotopes.

Table 1

Energies characteristic of the β- and γ-radiation of the radioactive isotopes I-131 and K-42

Isotope	E_γ(MeV)	$E\beta$(MeV)
I-131	0.36 (80%)	0.61 (87%)
K-42	1.52 (18%)	2.0 (18%)
		3.6 (82%)

Theoretically, the following methods can be used for the determination of the two isotopes:

1. the measurement of the absorption of the β-radiation with a GM detector;

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2. the measurement of the absorption of the β-radiation by the scintillation method;

3. the measurement of β-radiation by energy selective method involving an integral circuit discriminator;

4. the measurement of γ-radiation by energy selective method involving an integral circuit discriminator;

5. the measurement of γ-radiation by energy selective method involving a differential discriminator (resolution of the composite spectrum),

6. the measurement of γ-absorption by the scintillation method.

Out of the listed possibilities, the measurement of the absorption of γ-radiation was ruled out on account of the required thickness of the absorber. Our present paper reports on results obtained with the methods 1, 2 and 3, while methods 4 and 5 will be discussed in our following communication.

Experimental methods

1. *Investigation of β-absorption with a GM detector*

As is well known, the intensity of the β-radiation of an isotope decreases exponentially in dependence on the thickness of the absorber layer.

$$I = I_0 e^{-\mu' x}; \text{ where } \mu' = \text{const.} E_{\max}^{1.33}$$

and thus

$$I = I_0 e^{-\text{const} \cdot E_{\max}^{1.33} x}.$$

Thus, the degree of diminution will depend also on the $E_{\max}$ energy of the β-radiation. It is to be expected, therefore, that in the case when the $E_{\max}$ energies of the two radiation components investigated differ sufficiently, the intensity of the softer β-component can be neglected after a certain thickness of the absorber beside the harder component. Indeed, this fact evident from data in Table 1, and verified also by experiments is related to the β-radiation of the I—137 and K—42 isotopes.

When the intensity of the preparation containing the isotopes of two kinds is measured first without an absorber, and subsequently in the presence of an absorber of a certain thickness, the first measurement will yield the sum of the intensities of the two isotopes, while in the second case only a portion of the original intensity of the K—42 β-radiation of higher energy will be measured, depending on the nature and the thickness of the absorber.

The required thickness of the absorber layer has been determined in preliminary experiments, using standard preparations of known activity, containing only the I—131 or the K—42 isotope. For an aluminium absorber this thickness was found to be 1 mm.

Table 2

Calculated activities of the reference standards

Reference standard No.	Starting activities μCi	
	I-131	K-42
1	0.095	—
2	—	0.106
3	0.082	0.103
4	0.096	0.101
5	0.094	0.0103
6	0.084	0.0013

Results of our investigations for the determination of I—131 and K—42 in the presence of each other are contained in Table 3. Initial iodine and potassium activities measured for the samples listed in Table 3, are shown in Table 2. The same six reference standards were used in the investigation of the other two methods.

According to data in Table 3, an aluminium sheet of 1.06 mm thickness decreases the intensity of the preparation consisting of pure I—131 isotope

Table 3

The measurement of the absorption of β-radiation with GM detector

Reference standard No.	Thickness of aluminium mm	Average intensity, corrected for background cpm
1	0	2191
	1.06	9
2	0	1403
	1.06	840
3	0	3224
	1.06	835
4	0	1538
	1.06	803
5	0	2278
	1.06	93
6	0	1906
	1.06	21

to 0.4 per cent of its original value without the absorber, while in the case of the K—42 isotope, 60 per cent of the original intensity was indicated. With a preparation containing both isotopes, the measuring procedure will be as follows: The total intensities of the two isotopes is measured first without absorber. Next, the 1.06 mm aluminium absorber is placed over the preparation, and the intensity measured is multiplied by the factor 1.67, compensating for the loss in intensity by absorption of the K—42 β-radiation to yield the intensity without absorption, corresponding to the K—42 content of the preparation. This value is then subtracted from the intensity measured without the absorber. The difference corresponds to the quantity of I—131. If the intensity measured in the presence of the absorber is of the same order of magnitude as the residual intensity of the I—131 radiation calculated, also this latter must be involved into the correction.

2. *Investigation of the β-absorption by scintillation technique*

The principle and the circumstances of this method are the same as those described in the preceding paragraph, only the mode of detection is different. In these measurements a scaler of type NK—108 and a scintillation β-crystal were used. Results are summarized in Table 4. According to data in this table,

Table 4

The measurement of the absorption of β-radiation with scintillation techniques

Reference standard No.	Thickness of aluminium mm	Average intensity, corrected for background cpm
1	0	4450
	1.06	243
2	0	4302
	1.06	2385
3	0	8064
	1.06	2582
4	0	4481
	1.06	2257
5	0	4942
	1.06	464
6	0	4062
	1.06	238

the intensity of K—42 measured after having passed an absorber of 1.06 mm thickness is decreased to 55.4 per cent of the value measured without an absorber, so that the correction factor in this method is 1.8. Measurement procedures and evaluation are the same as described for the preceding method.

3. *Measurement of β-radiation by an energy selective method involving an integrated circuit discriminator*

The intensities of the β-radiation of the two isotopes are measured together with an NK—108 apparatus at a discriminator voltage of 5V.

When the discriminator voltage is adjusted to 40 V, the intensity of the preparation containing only the I—131 isotope is 2.22 per cent of the intensity measured at 5V, while under similar circumstances 73 per cent of the intensity of the β-radiation of the K—42 isotope is detected. Data measured are shown in Table 5.

Table 5

The measurement of β-radiation with integrated circuit discriminator energy selective method

Reference standard No.	Discriminator voltage V	Average intensity, corrected for background cpm
1	5	4450
	40	99
2	5	4302
	40	3152
3	5	8064
	40	3011
4	5	4481
	40	2842
5	5	4942
	40	363
6	5	4062
	40	118

Calculations used for the evaluation

Whichever method is used, data measured are processed according to the following considerations:

Total intensity measured (i_t) is:

$$i_t = i_K + i_I + i_b \tag{1}$$

where i_K is the total intensity of potassium,
i_I is the intensity of the iodide,
i_b is the background intensity;

the intensity corrected for the background (i) is:

$$i = t_t - i_b = i_K + i_I; \tag{2}$$

the intensity measured on "screening" I—131 (i_s) is

$$i_s = i_{K,r} + i_{I,r} + i_b \tag{3}$$

where $i_{K,r}$ is the residual intensity of K—42. and
$i_{I,r}$ is the residual intensity of I—131
Correcting i_s for the background:

$$i_{s,c} = i_s - i_b = i_{K,r} + i_{I,r} \tag{4}$$

In the given adsorption experiments, generally

$$i_t \simeq i_I. \tag{5}$$

The residual intensity of I—131 after "screening" is:

$$i_{I,r} \leq i_t \cdot f_r \tag{6}$$

where f_r is a factor depending on the measuring technique, and involves the ratio of the I—131 intensities with and without screening, respectively. (With a GM tube detecting β-absorption, this factor is e.g. 0.004.)

Thus, the residual intensity of K—42 is:

$$i_{K,r} = i_{s,c} - i \cdot f_r \tag{7}$$

The critetion of K—42 detectability is:

$$i_{K,r} \geq \sqrt{(sd)^2_{i_b} + (sd)^2_{i_c}} \tag{8}$$

where: $i_c \equiv i \cdot f_r$ and (sd) is the standard deviation.

The reference activities listed in Table 2 were calculated from data made available by the Isotope Institute. These activities pertain to the time of pre-

paration of the reference standards. Since, however, the duration of the experiments is comparable to the half life of the K—42 isotope, the activity of this latter, and hence, the ratio of the activities changed considerably during the measurements. Activity data contained in the following tables represent therefore values corrected for the time of preparation of the reference standards, to make possible a comparison. The corrected values were determined with the so-called comparing sample method. The intensities of the reference standards containing both isotopes (3—6) were determined with the given measuring method (taking the average of measurements of 10 ×1 minute), and then the intensity of the reference standard containing only the K—42 isotope (1), and from case to case the intensity of the reference standard containing only I—131 (2) were measured. As the conditions of measuring (thickness of the sample, counting geometry, etc.) were identical in the case of the pure and the mixed reference standards, the starting activity of the components of the mixed reference standards could be calculated from the intensity measured at the time of the investigation on the basis of the i/a_0 ratio (a_0 = starting activity) of the corresponding pure reference standards (1 and 2, respectively).

Example. — On measuring the intensity of the reference standard 2 with a GM detector without an absorber, an average of 1403 cpm was obtained, while in the case of an aluminium absorber of 1.06 mm thickness an intensity of 840 cpm was found (Table 3). The intensity of the K—42 β-radiation has been diminished therefore by the absorber by a factor of

$$\frac{1403}{840} = 1{,}67\,.$$

In the case of reference standard 3, a value of 3224 cpm was measured without absorber, representing the sum of the intensities of the K—42 and I—131 isotopes contained in the reference standard. In the presence of the absorber, the intensity measured was 835 cpm. This latter, multiplied by 1.67, gives 1393 cpm. This value is the contribution of the K—42 isotope to the total intensity of 3224 cpm. Thus, the intensity due to I—131 will be:

$$3224 - 1393 = 1831 \text{ cpm.}$$

At the time of measuring the reference standard 3, the intensity of the reference standard containing only the isotope K—42 has been 1403 cpm, and that of the reference standard containing only the isotope I—131 2191 cpm, so that the activity of the reference standard 3 (referred to the time of preparation of the reference standards) was the following:

$$\frac{1831}{2191} \cdot 0.095 = 0.080\ \mu\text{Ci}\ \ I-131$$

$$\frac{1393}{1403} \cdot 0.106 = 0.105\ \mu\text{Ci}\ \ K-42.$$

In Table 6, these two data are given for the reference standard 3. The activity of 0.095 μCi for reference standard 1, and that of 0.106 μCi for reference standard 2, were taken from Table 2.

Data in Table 2 (and identical data under the designation "dosed composition" in Table 6), were calculated, as mentioned earlier, on the basis of data supplied by the Isotope Institute.

Evaluating in a similar way the measuring results, that value was taken as the limit of detectability of the K—42 isotope, which at the end of the duration of the experiment (after about one half life) could still be detected.

Table 6

Determination of the radioactive isotopes I-131 and K-42 in the presence of each other with three methods measuring the β-radiation. Series I

Activity calculated on the basis of dosage, μCi		Reference standard No. 3		Reference standard No. 4	
		I-131	K-42	I-131	K-42
		0 0820	0 1030	0 0096	0 1010
Activity found, μCi percentage deviation from the calculated value	β-absorption, GM detector	0.080 −3%	0.1050 +2%	0.0087 −10%	0.1010 0%
	β-absorption, scintillation detector	0.0820 0%	0.1040 +1%	0.0090 −6%	0.100 −1%
	β-radiation measured by the energy selective method	0.087 +6%	0.098 −5×	0.0120 + 25%	0.096 −5%

Activity calculated on the basis of dosage, μCi		Reference standard No. 5		Reference standard No. 6	
		I-131	K-42	I-131	K-42
		0 0940	0.0103	0 0840	0 0013
Activity found, μCi percentage deviation from the calculated value	β-absorption, GM detector	0.0930 −1%	0.0106 +3%	0.0820 −2%	0.0015 +15%
	β-absorption, scintillation detector	0.099 +5%	0.0098 −5%	0.0860 +2%	(0.0010)* (−23%)
	β-radiation measured by the energy selective method	0.0980 +4%	0.0089 −13%	0.0860 +2%	(0.0010)* (−23%)

* The value does not satisfy the criterion according to (8).

The results obtained with the other two methods were evaluated in the same way as in the example given above.

Table 6 shows the results obtained with the three methods, and indicates also the percentage deviations from the activities, calculated on the basis of dosage.

Table 7 contains the results of a similar series of experiments, with the difference that the activity ratio of the isotopes I—131 and K—42 was about twice that in the preceding experiment series.

Table 7

Determination of the radioactive isotopes I-131 and K-42 in the presence of each other with three methods measuring the β-radiation. Series II

Activity calculated on the basis of dosage, μCi		Reference standard No 7		Reference standard No 8	
		I-131	K-42	I-131	K-42
		0 195	0 110	0 0195	0 110
Activity found, μCi percentage deviation from the calculated value	β-absorption, GM detector	0.187 −4%	0.106 −4%	0.0172 −12%	0.108 −2%
	β-absorption, scintillation detector	0.201 +3%	0.108 −2%	0.022 +13%	0.103 −6%
	β-radiation measured by the energy selective method	0.209 +7%	0.104 −5%	0.022 +13%	0.103 −6%

Activity calculated on the basis of dosage, μCi		Reference standard No. 9		Reference standard No. 10	
		I-131	K-42	I-131	K-42
		0 195	0 0110	0 195	0 0011
Activity found, μCi percentage deviation from the calculated value	β-absorption GM detector	0.201 +3%	0.0115 +5%	0.196 +0.5%	0.0016 +50%
	β-absorption, scintillation detector	0.206 +6%	0.0094 −15%	0.203 +4%	—
	β-radiation measured by the energy selective method	0.202 +4%	0.011 0%	0.209 +7%	—

Evaluation of the methods used

Results of both the first and second experiment series showed that the isotopes I—131 and K—42 can be determined in general with an accuracy of $\pm$ 10 per cent in the presence of each other, if their activity ratio is comparable or within the same order of magnitude. If the activity ratio of the I—131 and K—42 isotopes is 65 : 1 (reference standard 6), they can be determined with an error of 15—25 per cent by the method of calculation described. If the activity ratio is 175 : 1 (reference standard 10), the error of the K—42 energy selective method measuring β-radiation, will be +36 per cent, while that of the method measuring β-absorption 50 per cent.

The criterion expressed by the inequality (8), which was accepted as the limit of detectability of K—42, is satisfied only by the method measuring β-radiation with a GM detector.

The possibility of measuring I—131 and K—42 quantities of this ratio in the presence of each other is of importance on account of the fact that iodide ions are chemisorbed and potassium ions physically adsorbed on a germanium surface. Consequently (as shown by other measurement) in case of identical solution concentration, the quantity of iodide adsorbed on the surface of germanium is by an order of magnitude greater than that of potassium ions.

Summary

The study of ion adsorption on the (111) face of germanium single-crystals made it necessary to determine simultaneously anion and cation adsorption with radioactive indication techniques.

Out of the six possible measuring methods for the determination of the radioactive isotopes K-42 and I-131 in the presence of each other, the present paper discusses three methods.

Owing to the different character of the adsorption of these two ions on germanium, it becomes necessary to determine in the presence of an I-131 isotope of a given activity a K-42 isotope the activity of which is by 1—2 orders of magnitude lower than that of the I-131 isotope. From this viewpoint, the measuring of β-absorption with a GM detector was found to be satisfactory.

References

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