



Title	CHARGING OF RADIOACTIVE PREPARATIONS
Author(s)	GROMOV, V. V.; SPITSYN, VICT. I.
Citation	JOURNAL OF THE RESEARCH INSTITUTE FOR CATALYSIS HOKKAIDO UNIVERSITY, 19(1), 1-16
Issue Date	1971-04
Doc URL	<a href="http://hdl.handle.net/2115/24916">http://hdl.handle.net/2115/24916</a>
Type	bulletin (article)
File Information	19(1)_P1-16.pdf



[Instructions for use](#)

## CHARGING OF RADIOACTIVE PREPARATIONS

By

V. V. GROMOV\*) and VICT. I. SPITSYN\*)

(Received October 28, 1970)

### Abstract

The charge accumulation in the radioactive dielectrics has been investigated. The theoretical analysis was made from point of view the dielectric properties of crystals and an environment. The calculation of a charge in radioactive metals, semiconductors and dielectrics showed the satisfactory correlation with the experimental determinations of the charge.

The changing of physico-chemical properties as a result of the charge accumulation was illustrated in case of adsorption and kinetics of dissolution of the charged crystals in water.

First information about charging of radioactive preparations as a result of radioactive decay were reported by P. and M. Curie in 1900.<sup>1)</sup> They described their experiments on measuring of a radium-bearing  $\text{BaCl}_2$  charge. However, with the chosen experimental technique it was possible to find only the sign, but not the value of a stationary charge, since only the current of ionizing particles from the sample was determined. Other results in this respect were not published.

Recently, the idea of charging the surfaces of radioactive preparations (salts, oxides) has been widely used to explain some catalytic properties,<sup>2-6)</sup> adsorption<sup>7-13)</sup> and dissolution<sup>14-18)</sup> characteristic features of highly radioactive compounds. The results obtained point to the necessity of a more detailed analysis of the mechanism of formation of a stationary charge at the surfaces of radioactive samples.

Any radioactive preparation should acquire a charge with a sign opposite to the sign of a charge of particles leaving the surface of a sample. This phenomenon is already used in some types of atomic batteries where a difference in potentials of some thousands volts is set up between a radioactive preparation and the environment (collector) due to radioactive radiation. In this case, a radiation source as a rule is surrounded by a medium with low conductivity: a vacuum is set up or a solid dielectric is used.<sup>19,20)</sup> However under conventional conditions, for instance in water or in air, the charge of

---

\*) Institute of Physical Chemistry, USSR Academy of Sciences, Moscow, U.S.S.R.

radioactive preparations turns to be small due to the processes of its neutralization by the environmental ions. Approximate calculations show that for  $\text{BaSO}_4^*$  containing  $\sim 100$  m-curie/g of  $\beta$ -emitter of sulfur-35, this change in distilled water may equal  $+1.10^{-15}$  coulomb/cm.<sup>2,21)</sup> It seems to be of no sense to mention a stationary charge under certain conditions, since it may simply be absent. Apparently, electric properties of the environment and of radiation source itself play an important role in this process.

### Estimation of charge value of radioactive preparations

Let the time of charge relaxation in the solid body be determined by the relationship<sup>22)</sup>

$$\tau = \epsilon / \sigma, \quad (1)$$

where  $\epsilon$  is a dielectric constant and  $\sigma$  is the specific electric conductivity. Then variations with time in charge density ( $q_I$ ) on a radioactive preparation obey the law

$$dq_I/dt = J_I - q_I/\tau_{\text{eff}}. \quad (2)$$

Here and below, symbol  $I$  refers to a radioactive preparation  $II$  to environment;  $J_I$  is current density of changed particles leaving  $I$  on the radioactive decay of an isotope contained in  $I$ . Upon integrating (2), under the condition that  $J_I = J_0 \exp(-\lambda t)$  and  $q_I = 0$  at  $t=0$ ,  $q_I = q_t$  at  $t$  we obtain

$$q_I = q_0 [1 - \exp(\lambda t - t/\tau_{\text{eff}})] \exp(-\lambda t), \quad (3)$$

where  $\lambda$  is a constant of the isotope decay,  $q_0 = J_0/(1/\tau_{\text{eff}} - \lambda)$ ,  $\tau_{\text{eff}} = f(\tau_I, \tau_{II})$ . Thus, the time of setting of a stationary charge and its value are determined by the dielectric properties of a radioactive preparation and environment.

It should be noted that for the system under consideration the availability of the electron conductivity, homogeneity of the dielectric properties and observance of the Ohm's law in  $I$  and  $II$  are assumed.

If the isotope decay in  $I$  may be neglected, i.e.,  $J_I = J_0$  and  $\lambda \sim 0$ , then expression (3) is simplified

$$q_I = q_0 [1 - \exp(-t/\tau_{\text{eff}})] \quad (4)$$

This formula coincides with the empiric law proposed in paper<sup>23)</sup> for calculating the change accumulated in glasses at external  $\beta$ -irradiation.

Without analysing the details of charge distribution in radioactive samples, we shall find the value of  $q_0$  for the case when  $\lambda = 0$ . Under the stationary conditions,  $J_I = J_{II}$ , where  $J_{II} = eu_{II}(n_0 + n_{II})\mathcal{E}$ , and  $J_I = |K_1/\rho Q$ . Hence, taking into account that  $\mathcal{E} = 4\pi q_0/\epsilon_{II}$ , we obtain

*Charging of Radioactive Preparations*

$$q_0 = K_1 \varepsilon_{II} p Q / 4\pi e u_{II} (n_0 + n_{II}), \quad (5)$$

where  $K_1$  is a constant with either plus or minus sign, which depends on the type of particles leaving  $I$  (for instance, if there is a  $\beta$ -radiating isotope in  $I$ , then  $K_1$  is taken with the plus sign);  $\mathcal{E}$  is the electric field strength which is set up on the surface of  $I$  by a charge;  $J_{II}$  is the density of current carriers arriving from the environment to the surface of a radioactive sample;  $p$  is its weight;  $Q$  is the specific radioactivity;  $\varepsilon_{II}$  is the dielectric constant of  $II$ ;  $e$  is the electron charge;  $u_{II}$  is the mobility of current carriers in  $II$ ;  $n_0$  is their stationary concentration without irradiation effect;  $n_{II}$  is the additional number of carriers arising in  $II$  on irradiation. In turn, constant  $K_1$  is equal to  $K_2 e N_Q \varphi(\mu, r) / S_I$ , where  $S_I$  is a surface of  $I$ ;  $N_Q$  is the amount of particles emitted per second by unit radioactivity of the given isotope;  $\varphi(\mu, r)$  is the function defining the portion of particles leaving the sample of  $r$  thickness;  $\mu$  is the coefficient of radiation absorption in a radioactive preparation;  $K_2$  is a constant which depends on dimensionality of values in formula (5). The form of function  $\varphi(\mu, r)$  for samples of various forms can be found in papers.<sup>24,25)</sup>

In order to find  $n_{II}$ , we shall assume that the rate of ionization of the surrounding medium  $v_1$  is proportional to  $Q$ , and the rate of ion recombination  $v_2$  is a bimolecular process. For instance, this is valid for gases and water. Under these conditions,

$$v_1 = \{N_Q p Q \varphi(\mu, r) \sum F(E) |\overline{dE/dx}| \Delta E / w S_I\} \quad (6)$$

and

$$v_2 = d_i n_{II}^2, \quad (7)$$

where  $d_i$  is a coefficient of ion recombination in  $II$ ;  $F(E)$  is the function of particles distribution by energy in the radiation spectrum of a given isotope;  $\overline{dE/dx}$  is the average losses of a radiation energy ( $E$ ) into the surrounding medium; sign  $\sum$  implies summarization throughout the spectrum;  $w$  is a radiation energy which is spent on formation of ion pair in  $II$ . Value of  $\overline{dE/dx}$  ( $x$  is the distance covered by irradiation in  $II$ ) at the surface of  $I$  can be determined on the base of Katz-Penfold formula.<sup>26,27)</sup> It should be noted that (7) is valid if decrease of ions in  $II$  at the cost of their discharging on the surface of  $I$  is negligible as compared with  $v_2$ .

Having determined  $n_{II}$  at  $v_1 = v_2$  and substituting this value into (5), we finally obtain<sup>28)</sup>

$$q_0 = K_1 \varepsilon_{II} p Q / 4\pi e u_{II} \left\{ n_0 + \left[ N_Q p Q \varphi(\mu, r) \sum F(E) \left| \frac{\overline{dE}}{dx} \right| \Delta E / w d_i S_I \right]^{\frac{1}{2}} \right\}, \quad (8)$$

If  $n_0 < n_{II}$ , then formula (8) simplifies

$$q_0 = K_3 Q^{\frac{1}{2}}. \quad (9)$$

As follows from (9), the charge of a radioactive preparation increases directly as the square root of the specific radioactivity. This result for a flat radioactive conductor in air was obtained in the qualitative form in paper.<sup>5)</sup>

When  $n_0 \gg n_{II}$  in the environment, then as can be seen from (8), a stationary charge should be proportional to the specific radioactivity of  $I$  ( $q_0 \sim KQ$ ).

For a flat radioactive preparation<sup>29)</sup>

$$K_{3(\text{fl.})} = K_2 \varepsilon_{II} [N_Q p \varphi(\mu, r)_{\text{fl.}} / S_I]^{\frac{1}{2}} / 4\pi u_{II} A,$$

for the case of sphere

$$K_{3(\text{sphere})} = K_2 \varepsilon_{II} [N_Q p \varphi(\mu, r)_{\text{sph.}} / 4\pi r^2]^{\frac{1}{2}} / 4\pi u_{II} A,$$

where  $A = [(\sum F(E) |dE/dx| \Delta E) / w d_i]^{\frac{1}{2}}$ .

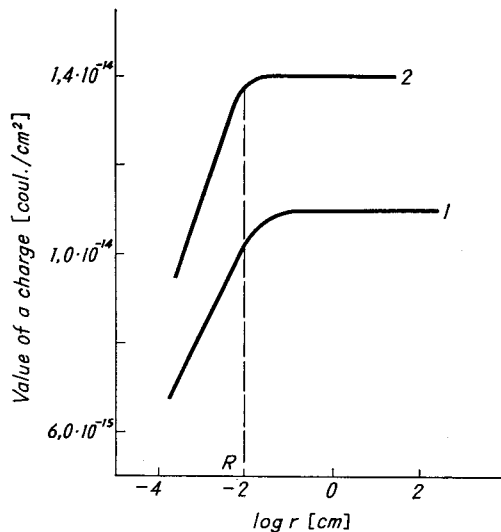
Let us consider some consequences from formulae (8) and (9). If (9) is fulfilled, then for instance for a sphere and a plate, it is valid that

$$q_0^{\text{fl.}} / q_0^{\text{sph.}} = [\varphi(\mu, r)_{\text{fl.}} \cdot 4\pi r_{\text{sph.}}^2 / \varphi(\mu, r)_{\text{sph.}} \cdot S_{\text{fl.}}]^{\frac{1}{2}} \quad (10)$$

In relationship (10), as can be seen from calculations, a numerator is always greater than a denominator, if  $r > R$  ( $R$  is a pathlength of particles of the given type in substance  $I$ ), or if, at least, the thickness of the layer of  $I$ , where particles defining  $J_I$  are escaping from, is equal. Hence, a charge per unit surface of a flat sample is greater than that of a sphere. This is defined by purely geometrical factors. Area of a circle is taken as a unit surface area of  $I$ . Then a flux of particles which leave a given surface of a flat sample will be proportional to amount of a radioactive substance ( $P_1$ ) confined by cylinder in layer  $R$ . For the case of a sphere, this substance ( $P_2$ ) is enclosed in a truncated spherical sector with height  $R$  and a large base equal to the area of a cylinder base. Then, however,  $P_1 > P_2$ , i.e., at the convex surface, the intensity of escaping particles with equal  $Q$  is smaller than that at the flat surface, and it acquires a smaller charge.<sup>\*)</sup> It should be noted that  $q_0$  increases at  $Q = \text{const}$  and at growing  $p$  only till  $r$  will be  $\geq R$ , since further increase of  $p$  no longer affects on  $J_I$ , Fig. 1.

\*) It is assumed that the difference in the degree of ionization for flat and spherical samples with an equal specific radioactivity may be neglected.

## Charging of Radioactive Preparations



**Fig. 1.** Calculated values of a stationary surface charge ( $q_0$ ) of radioactive tungsten with specific radioactivity of 30 m-curie/g,  $W^{185}$ ,  $\beta$ -emitter, air is an environment; calculations were carried out for the normal conditions by formula (9)

1—a spherical sample; 2—a flat sample

As can be seen from Fig. 1, the calculations confirm the conclusion that  $q_0^{\text{sph.}} < q_0^{\text{fl.}}$ , all other things being equal. Besides, calculated and experimentally obtained values of  $q_0^{\text{fl.}}$  agree fairly well (Cf. Fig. 1 and Table 1).

Formula (9) permits obtaining a simple dependence of  $q_0$  in air on pressure ( $P$ ). When it is taken into account that over the pressure range 1 to  $10^3$  mm·Hg,  $d_i$  varies directly with  $P_{\text{air}}$ , and  $J_{II}$  is proportional to the square of pressure at low pressures (this is equivalent to the condition  $dF/dx \sim P^{2,30}$ ), then at constant  $Q$  value we obtain by way of elementary transformations

$$q_0 = CP^{-\frac{1}{2}}, \quad (11)$$

where  $C = K_3(760Q)^{\frac{1}{2}}$ .

As may be inferred from (11), the charge of radioactive preparations should increase with decreasing pressure. For perfect vacuum, this expression seems to be unsuitable, since  $u_{II}$  markedly increases and a linear dependence of  $d_i$  on  $P$  and of  $J_{II}$  on  $P^2$  is disturbed. According to expression (11) changing of  $q_0$  apparently is well kept over the pressure range 1 to 10 mm Hg where  $u_{II}$  may be assumed to be constant (for values of  $\mathcal{E}/P_{\text{air}}$  not greater than 25).<sup>30)</sup>

Time of accumulation of a stationary charge also will vary depending on  $P$ . For bimolecular process of ion recombination in air ( $\tau = 1/d_i \cdot n_{II}$ ), it is easy to show that  $\tau \simeq KQ^{-\frac{1}{2}}$  (a sample weight is constant and  $P = \text{const}$ ) and if  $Q = \text{const}$ , then  $\tau \simeq kP^{-\frac{3}{2}}$ . Consequently, at low pressures, equilibrium between  $I$  and  $II$  is reached slower. This conclusion is valid at restrictions inherent in formula (11).

When deducing formulae (8) and (9), a number of assumptions were made which reduce the accuracy in calculating values of  $q_0$  down to  $\pm 40$  to 60%. In particular, it was assumed that the exponential law was always fulfilled at self-absorption of radiation in  $I$  (this is valid only for gamma-quantum); "softening of spectrum" of the particles at the cost of their deceleration in the radioactive preparation before escaping into the environment was not taken into account *etc.* In spite of it, the expressions obtained permit making a comparative evaluation of the charge of radioactive samples.

### Conductors and semiconductors

In the case of fairly great values of  $\sigma_I$  ( $\sigma_I > 10^{-4}$  to  $10^{-6} \text{ ohm}^{-1} \text{ cm}^{-1}$ ) and  $\sigma_I > \sigma_{II}$ , all the formulae obtained and the conclusions deduced are valid for radioactive preparations. The condition for good electroconductivity of  $I$  is equivalent to the assumption of rapid charge leakage from the volume onto the surface of the radioactive preparation. In this case, if  $\tau_I < \tau_{II}$ , the kinetics of equilibrium established between  $I$  and  $II$  is determined by  $\tau_{II}$ . It should

TABLE 1 The  $q_0$  values of radioactive preparations<sup>28,29)</sup>

Sample and its Isotope and its geometry properties		Pressure, mm Hg	Charge value, $\text{c/cm}^2$	
			calculated	measured
Nickel plate $1.8 \times 0.55 \times 0.78 \text{ cm}$	Ni <sup>63</sup>	760	$+0.8 \cdot 10^{-15}$	$+1 \cdot 10^{-15}$
	2 m-curie/g	100	$+2.2 \cdot 10^{-15}$	$+2 \cdot 10^{-15}$
	$\beta$ -radiator	10	$+7.0 \cdot 10^{-15}$	$+5 \cdot 10^{-15}$
	$E_{\text{max}} = 0.067 \text{ Mev}$	1	$+2.0 \cdot 10^{-14}$	$+1 \cdot 10^{-14}$
Tungsten disk $r = 1.5 \text{ cm}$ $h = 0.2 \text{ cm}$	W <sup>185</sup>	760	$+1.41 \cdot 10^{-14}$	$+0.7 \cdot 10^{-14}$
	30 m-curie/g	100	$+3.9 \cdot 10^{-14}$	$+3.0 \cdot 10^{-14}$
	$\beta$ -radiator	10	$+1.2 \cdot 10^{-13}$	$+5.2 \cdot 10^{-14}$
	$E_{\text{max}} = 0.427 \text{ Mev}$	1	$+3.9 \cdot 10^{-13}$	$+5.5 \cdot 10^{-13}$
Zinc disk $r = 0.5 \text{ cm}$ $h = 0.2 \text{ cm}$	Zn <sup>65</sup>	760	$-1.6 \cdot 10^{-15}$	$-1 \cdot 10^{-15}$
	13 m-curie/g	100	$-1.4 \cdot 10^{-14}$	$-1.2 \cdot 10^{-14}$
	e.c. $\beta$ +-radiator			
	$E_{\text{max}} = 0.32 \text{ Mev}$	10	$-4.6 \cdot 10^{-14}$	$-2.6 \cdot 10^{-14}$

### Charging of Radioactive Preparations

be also noted that if both the media are good conductors, the charge on the surface of  $I$  is practically absent.

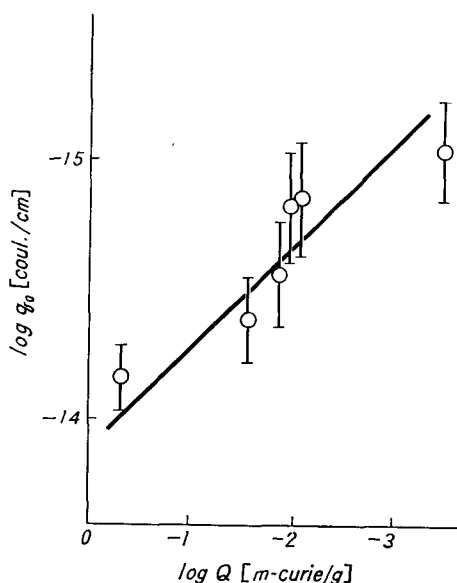
It is most simple to measure the charge in the case of radioactive conductors in the gas medium. Such determinations were carried out for some radioactive metals (Table 1).

An electrometer SG-1M with current sensitivity up to  $\sim 1 \cdot 10^{-16}$  was used as a meter (the method of charge accumulation). Samples surrounded by a metallic collector of ionizing particles were located in a glass chamber which can be readily evacuated. The accumulated charge was distributed between  $I$  and an electrometer filament, from which deviation the charging of radioactive metals can be judged. An average relative error in measurements was  $\pm 52\%$  for small charges.

As can be seen from the table, the charge values determined experimentally and calculated by formulae (9) and (11) satisfactorily coincide. In all cases, the sign of the charge of the radioactive metal surfaces corresponds to the assumptions of the theory. The calculation accuracy was not worse than  $\pm 50\%$ . The main error arises from rather complex conditions of theoretical evaluation of the capacity of the sample ( $I$ )-ionized medium and collector ( $II$ ) system. It is more convenient to perform calculations if the system capacity (this value enters into the constant  $K_1$ ) is found experimentally.

Simultaneously performed experiments confirm the conclusion that a stationary charge on the surface of radioactive samples is set up faster at higher pressures. Indeed, for radioactive nickel at  $P=1$  mm Hg ( $\text{Ni}^{63}$ ,  $\beta$ -radiator, 2 m-curie/g)  $q_I$  becomes equal to  $q_0$  in  $\sim 200$  sec, while at 0.1 mm Hg only in  $\sim 10$  min. This should be expected, since  $\tau \sim kP^{-\frac{3}{2}}$ .

Similar experiments were carried out for germanium plates after diffuse alloying by radioactive antimony<sup>31</sup>), Fig. 2. Antimony-124 is an isotope with a complex spectrum of  $\beta$ - and  $\gamma$ -radiation;  $E_\beta=0.3$  to 2.3



**Fig. 2.** Dependence of surface charge of germanium on its specific radioactivity (antimony-124) air pressure is 2 to 6 mm Hg.



Mev. As was shown by microscopic examination of germanium surface after alloying, antimony is localized mainly on the surface of samples.

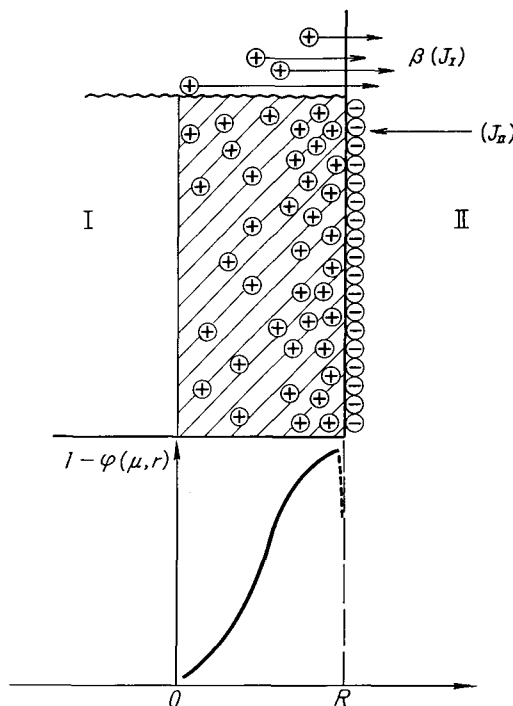
It follows from Fig. 2 that experimental data confirm the linear dependence of the surface charge of radioactive preparations on the square root of specific radioactivity.

To conclude the section on well-conducting radioactive samples, it should be noted that even in air their charge is small, reaching  $\sim 10^{-14}$  coulomb/cm<sup>2</sup> for 0.1 curie/g, and slowly increases with further increase in  $Q$ . A charge on radioactive metals in the medium with specific electrical conductivity, equal to or greater than  $\sigma_r$ , appears to be practically absent.

### Radioactive dielectrics

Two limiting cases are possible depending on electrical conductivity of the environment.

*I. Radioactive dielectric in a well-conducting medium,  $\tau_I \gg \tau_{II}$ .* The appearing charge is compensated (may be completely) by current carriers ( $J_{II}$ )



**Fig. 3.** Charge distribution in the radioactive dielectric containing a  $\beta$ -radiating isotope.

*Charging of Radioactive Preparations*

of the environment. However, a complete neutralization of charge does not occur, since electronic and especially ionic conductivities of a dielectric are small. As a result, even when a radioactive dielectric will be electroneutral as a whole, in its near-surface region, equal to the layer of complete absorption ( $R$ ) of radiation of the given type in material  $I$ , Fig. 3, there is formed "a double charged layer", one side of which is located on the surface, and the other at  $R$ -distance from it in the depth of a sample.

Thus, in radioactive dielectrics, besides a stationary charge  $q_0$ , a double charged layer appears which thickness is determined by the path value of radiation of the given type in  $I$ . To a first approximation, distribution of charge over the layer  $R$  is described by function  $1 - \varphi(\mu, r)$ . In order to estimate the charge value in the double layer  $q_R$ , the use can be made of the following expression

$$q_R = KeN_q p \varepsilon_I \varphi(\mu, r) Q / 4\pi S_I \sigma_I, \quad (12)$$

where  $\sigma_I = \sigma_0 + \sigma_i$ ,  $\sigma_0$  is an electric conductivity of  $I$  without self-radiation effect;  $\sigma_i$  is additional radiative electric conductivity of an radioactive preparations. Expression (12) is obtained from (5) or (8) by way of elementary transformations. If it is assumed that electric conductivity varies linearly under irradiation,<sup>32,33)</sup> then the following empirical relationship is fulfilled rather accurately for  $\beta$ -radiators

$$\sigma_i = \alpha N_q \frac{1}{3} E_{\max} \left[ 1 - \varphi(\mu, r) \right] Q / 6.2 \cdot 10^{13}, \quad (13)$$

where  $\alpha$  is an experimental parameter which is equal to  $10^{-16}$  for dielectrics with the same specific electric conductivity as for quartz.<sup>33)</sup>

Establishment of the stationary state in the layer  $R$  with time is readily evaluated by formula (3), if  $\tau_I$  is taken instead of  $\tau_{\text{eff}}$ .

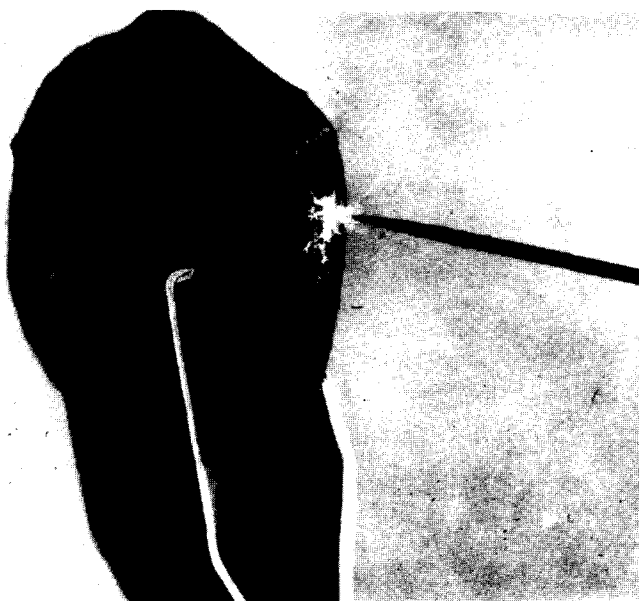
*II. Radioactive dielectric in a slightly conducting medium,  $\tau_I \simeq \tau_{II} \gg 1$ .* Apparently, all the formulae obtained for calculation of  $q_0$  and  $q_R$  are of little use here, since the effect of the charge field in  $I$  on the path of emitted particles should be taken into account, for instance for high vacuum. If vacuum is  $< 10^{-3}$  mm Hg,  $q_0$  may be evaluated in the ordinary way by formula (11). When the environment is a solid dielectric, one should take into consideration the charges in  $\varepsilon$  and  $\sigma$  in  $I$  and  $II$  under irradiation.

As it is seen from the approximate calculations, in the  $R$  layer of radioactive dielectrics fairly great charges may be stored up, which accumulation time and a total value of charging are determined only by dielectric properties of  $I$  i.e., by values of  $\varepsilon_I$  and  $\sigma_I$ . For instance, for crystals of strontium sulphate which contain 1 curie/g of pure  $\beta$ -radiator of sulfur-35,  $q_R$  is  $3 \cdot 10^{-8}$

coulomb/cm<sup>3</sup>. This charge is accumulated during  $\sim 2$  hours. A corresponding electric field strength in the double charged layer amounts to  $10^3 \sim 10^4$  volt/cm ( $\sigma_r$  of strontium sulphate is  $10^{-14}$  ohm<sup>-1</sup> cm<sup>-1</sup>). Consequently, for samples with  $\sigma \simeq 10^{-15}$  ohm<sup>-1</sup> cm<sup>-1</sup> and  $Q \simeq 10$  curie/g, a breakdown voltage of  $\simeq 10^6$  volt/cm may be achieved in layer  $R$ , and as a result of spontaneous discharge, microcracks may appear at the surface. Such a self-cracking should apparently be observed with high-radioactive non-conducting samples, for instance in glass blocks with radioactive waste.

In order to check the occurrence of spontaneous discharges in layer  $R$ , slightly-conducting glasses were prepared ( $\sigma \simeq 10^{-20}$  to  $10^{-22}$  ohm<sup>-1</sup> cm<sup>-1</sup>) which contain up to 50 m-curie/g of radioactive strontium-90 in equilibrium with yttrium-90. In such glasses the field strength in the charged layer may reach  $\sim 10^6$  volt/cm, and spontaneous electric discharge should appear. In fact, such a phenomenon was observed, though in order to stimulate a discharge one had to create a mechanical stress in the glass by pressing on the radioactive block with a metal or glass stick, as it was done in the case of irradiated dielectrics.<sup>23,25)</sup>

The discharge was recorded by a high-speed filmcamera SKS-IM-16 with frequency of 200 frame/sec, Fig. 4. In all the cases, a spark appeared



**Fig. 4.** A spontaneous discharge in glass containing 50 m-curie of strontium-90 in equilibrium with yttrium-90 per gramm of glass.

## Charging of Radioactive Preparations

TABLE 2 The charge of dielectrics,  $q_R$ 

Sample characteristic	Specific resistance	Layer thickness $R$	Difference in potential between measuring contacts*) volt	Pressure mm Hg	Charge in layer $R$ $/q_R/$ coulomb/cm <sup>3</sup>
	ohm·cm	mm			
Strontium sulphate, pressed pellets, con- tains $\text{Sr}^{90}\text{-Y}^{90}$ Specific radioactivity $\sim 10$ m-curie/g	$(3 \pm 1) \cdot 10^{13}$	$\sim 2.5$	0.4	760	$6.0 \cdot 10^{-12}$
			$1.0 \pm 30\%$	200	$1.0 \cdot 10^{-11}$
			3.9	50	$5.5 \cdot 10^{-11}$
			4.2	2	$6.0 \cdot 10^{-11}$
					Calculated value $2 \cdot 10^{-10}$ coulomb/cm <sup>3</sup> , at 760 mm Hg
Strontium glass, as disks contains $\text{Sr}^{90}\text{-Y}^{90}$ Specific radioactivity $\sim 50$ m-curie/g	$1 \cdot 10^{20} - 1 \cdot 10^{22}$	$\sim 2.2$	1.6	760	$7 \cdot 10^{-11}$
			$5.9 \pm 48\%$	200	$3 \cdot 10^{-10}$
			21	50	$1 \cdot 10^{-9}$
					Calculated value $1 \cdot 10^{-6}$ coulomb/cm <sup>3</sup> at 760 mm Hg

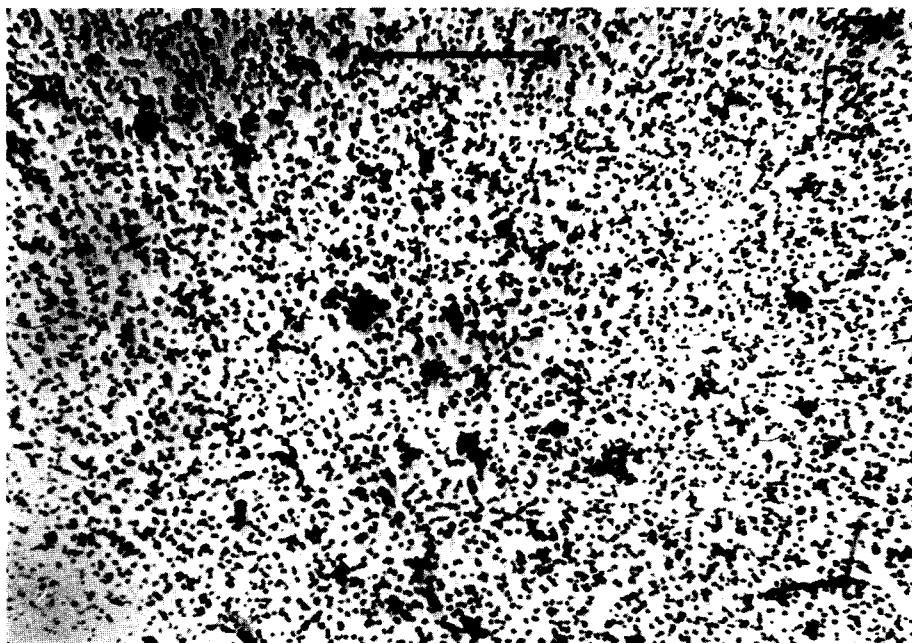
\*) The measuring contact distance is 1.2 to 1.3 mm

after a metal or glass stick (it was not grounded) was taken away from the glass surface.

One of the possible methods for direct measuring of  $q_R$  is the determination of EMF in the charged layer, which appears at the cost of charge accumulation in layer. To do this, "the method of charge accumulation" was used as in the case of radioactive metals,<sup>31)</sup> but one of the measuring contacts was inserted directly into the charged layer, while the other was pressed to the surface of  $I$ .<sup>29)</sup> Table 2 gives the results of determination of the charge localized in the "double charged layer" at the radioactive sample surface.<sup>35)</sup>

The difference in potential is small at normal pressure due to leakage of the scheme through the highly ionized surrounding air, as well as through the constantly irradiated insulation. As the evacuation is performed, the difference in potential between the measuring contacts grows, since the leakage through the air decreases. Apparently, in higher vacuum, the values of  $q_R$  close to these calculated may be obtained.

To measure the charge accumulated in radioactive dielectrics, the method of a vibrating capacitor was used in previous paper.<sup>36)</sup> The experiments were carried out with radioactive oxide of neodymium ( $\sim 7$  m-curie/g of  $\beta$ -radiator of neodymium-147) at pressures of  $10^{-5}$  to  $10^{-6}$  mm Hg. A gold plate was



**Fig. 5.** Microphotographs of the surfaces of spalls of potassium sulphate containing 50 m-curie of sulphur-35 (pure  $\beta$ -emitter).

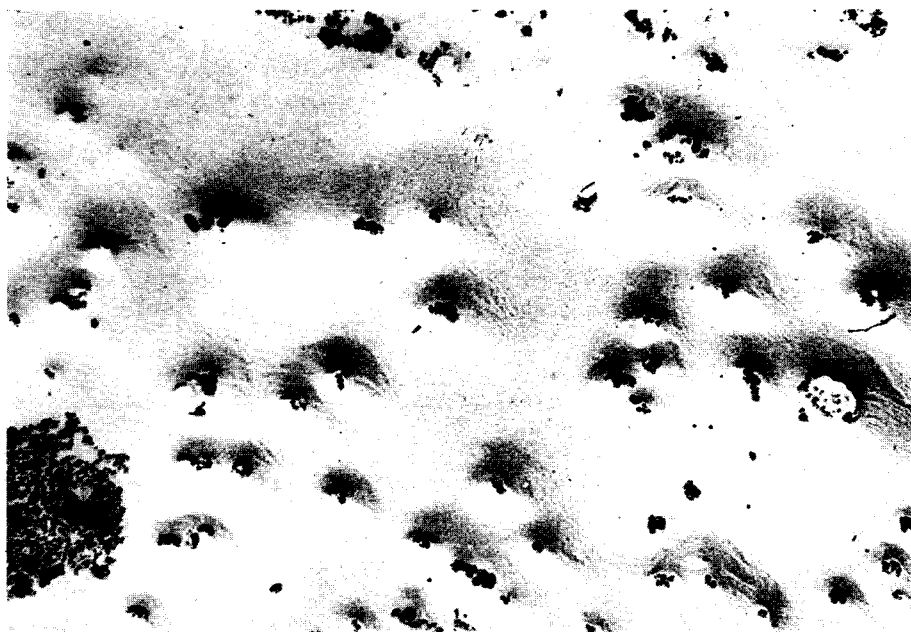
Magnification 20000 $\times$ . The surface is positively charged

a) the sample is covered by particles of negatively charged sol of silver (methyl alcohol as medium)

used as a reference electrode. The difference in potential between neodymium oxide and gold electrode reached  $\sim 100$  volt. However on heating, it decreases linearly with temperature growth and in the range of  $300\sim 400^\circ$  became close to the contact potential difference of non-radioactive neodymium oxide. This depends on increase in electric conductivity of neodymium oxide upon heating and on leakage of the charge off a radioactive oxide. Charges in potential (the sample was charged positively) with the value of specific radioactivity was described with a good degree of precision by the equation of the straight line of the type  $v_k = v_0 + KQ$ , where  $v_0$  is a hypothetical value of the potential difference with zero specific activity.

In certain conditions, the charge accumulated in the layer  $R$  may markedly affects the physicochemical properties of radioactive preparations. For instance, the charged sols of metals are precipitated on the surfaces of spalls of radioactive crystals in different ways.<sup>37)</sup> If a crystal of  $K_2SO_4^*$  (charged positively) is immersed into the solution of silver in methyl alcohol (metal

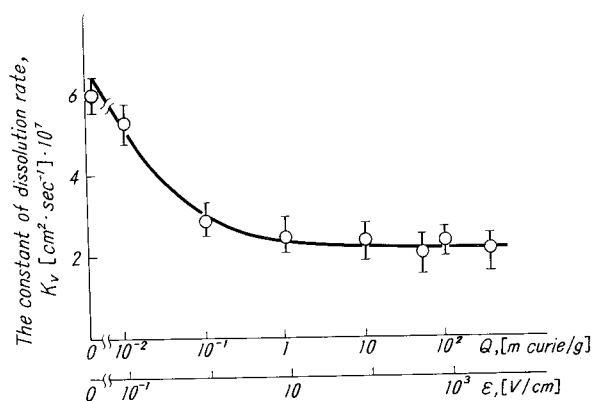
*Charging of Radioactive Preparations*



**Fig. 5.** b) the sample is covered by particles of negatively charged sol of nickel (acetone as medium).

particles are charged negatively) for some time, the whole surface of potassium sulphate is covered by a uniform layer of silver particles, Fig. 5.

In the similar conditions, sol of nickel in acetone (metal particles are charged positively) is hardly liberated on the surface of radioactive potassium



**Fig. 6.** Changes in the rate constant of dissolution of radioactive strontium sulphate (sulphur-35) depending on its specific radioactivity.<sup>38,39)</sup>

sulphate. These experiments indicate that under certain conditions (slightly-conducting environment), charging should profoundly affect the adsorption properties of solid radioactive bodies. Also  $q_R$  has a more profound effect on the kinetics of dissolution of ionic radioactive crystals, Fig. 6.

As follows from the figure, with increasing  $Q$ , *i.e.*, with growth of the field strength  $\mathcal{E}$  in the charged layer, the constant of dissolution rate  $K_v(\text{SrSO}_4^*)$  decreases. This may seem strange at first sight, since, overall, in such a well-conducting medium as water, strontium sulphate is certainly electroneutral ( $q_0=0$ ). This effect was explained in previous paper.<sup>39)</sup> As was found, deceleration in solving is due to the fact that a molecule-dipole of the charged ionic crystal has to make an additional work when passing from the layer  $R$  to the liquid phase. Simultaneously, according to the conclusions of the theory, decrease in  $K_v$  should proceed only to a definite limit and does not depend on the sign of the surface layer charge. These conclusions<sup>39)</sup> are well supported by experiment (See Fig. 6).

### Conclusions

1. Formation and distribution of charge on the surfaces of radioactive preparations are analysed. The formulae are proposed for calculating the charge value on the surfaces of solid radioactive bodies in various media.
2. It is found that directly near the surface radioactive dielectrics have a layer with charge ditribution, non-uniform in depth which thickness is equal to the charged particle path with the maximum energy, over the radiation spectrum of the given isotope.
3. The values of a surface charge of some radioactive conductors and dielectrics ( $\text{Ni}^*$ ,  $\text{Zn}^*$ ,  $\text{Ge}(\text{Sb})^*$ , glass +  $\text{Sr}^{90}$ ,  $\text{SrSo}_4^*$  etc.) are measured. The experimental data obtained agree satisfactorily with the theoretical calculations.
4. It is confirmed that charging of solid radioactive bodies may significantly affect their physicochemical properties (kinetics of dissolution, sorption of charged particles etc.).

### References

- 1) P. CURIE, and M. CURIE, *Compt. Rend.*, **130**, 647 (1900).
- 2) V. I. SPITSYN, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, N **11**, 1296 (1958).
- 3) A. A. BALANDIN, V. I. SPITSYN, N. P. DOBROSEL'SKAYA, and I. E. MIKHAILENKO, *Dokl. Akad. Nauk SSSR*, **121**, 495 (1958); **137**, 628 (1961).
- 4) A. A. BALANDIN, V. I. SPITSYN, N. P. DOBROSEL'SKAYA, and I. E. MIKHAILENKO, I. V. Vereshchinskis, P. Ya. Glazunov, *Actes II Congr. Internat. de Catalyse*, **2**, 1415,

*Charging of Radioactive Preparations*

- Technip, Paris (1961).
- 5) V. G. BARU, and F. F. VOLKENSTEIN, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, **N 11**, 1935 (1964).
  - 6) V. I. SPITSYN, and I. E. MIKHAILENKO, *At. Energiya*, **21**, 277 (1966).
  - 7) V. I. SPITSYN, and V. V. GROMOV, *Radiokhimiya*, **1**, 181 (1959).
  - 8) V. I. SPITSYN, and V. V. GROMOV, *Dokl. Akad. Nauk SSSR*, **159**, 427 (1964).
  - 9) V. I. SPITSYN, and V. V. GROMOV, *Trudy II Vsesoyuznogo soveshchaniya po radiatsyonnoi khimii*, 646, *Izd. Akad. Nauk SSSR, Moskva* (1962).
  - 10) V. V. GROMOV, and F. KEPAK, V. I. Spitsyn, in: "Soosazhdeniye i adsorbtsiya radioaktivnykh elementov", 110, *Izd. Nauka, M-L* (1965).
  - 11) V. I. SPITSYN, I. E. MIKHAILENKO, A. V. KISELEV, and L. D. BELYAKOVA, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, **N 7**, 1311 (1960).
  - 12) F. F. VOLKENSTEIN, *Kinetika i kataliz*, **2**, 481 (1961).
  - 13) W. FULKERSON, C. L. HEARN, and D. T. LELAND, *Chem. Eng. News*, **40**, N 6, 42 (1962); *J. Catalysis*, **4**, N 2, 194 (1965).
  - 14) V. I. SPITSYN, E. A. TORCHENKOVA, and I. N. GLAZKOVA, *Dokl. Akad. Nauk SSSR*, **132**, 643 (1960); **133**, 1111 (1960).
  - 15) V. I. SPITSYN, and N. G. MOSHCHANSKAYA, *Dokl. Akad. Nauk SSSR*, **133**, 859 (1960); **175**, 642 (1967).
  - 16) V. I. SPITSYN, and I. E. ZIMAKOV, *I. Akad. Nauk SSSR*, **138** (1961).
  - 17) C. H. BOVINGTON, and J. INORG, *Nucl. Chem.*, **27**, 1975 (1965).
  - 18) I. G. GLAZKOVA, Candidate Thesis, Moscow State University, Moscow (1968).
  - 19) A. B. GARRETT, *J. Chem. Educ.*, **33**, N 9, 446 (1956).
  - 20) U. CORLIS, and D. HARVEY, *Istochniki energii na radioaktivnykh izotopakh*, *Izd. MIR, Moskva* (1967).
  - 21) V. I. SPITSYN, and V. V. GROMOV, *Dokl. Akad. Nauk SSSR*, **156**, 427 (1964).
  - 22) I. A. STRATTON, *Electromagnetic Theory* 15, Mc Grow-Hill Book Co., N 4 (1941).
  - 23) J. FURUTA, H. HIRRAOKA, and S. OKAMOTO, *J. Appl. Phys.*, **37**, 1873 (1966).
  - 24) A. M. EFREMOV, in: "*Metallurgiya i metallovedeniye*", 382, *Izd. Akad. Nauk SSSR, Moskva* (1958).
  - 25) V. F. KOZLOV, and Yu. S. TROSHKIN, "*Spravochnik po radiatsionnoi bezopasnosti*", Atomizd., Moskva (1967).
  - 26) L. KATZ, and A.S. PENFOLD, *Rev. Mod. Phys.*, **24**, 28 (1952).
  - 27) Ya. CHUDAS, and I. TAURE, *Izv. Akad. Nauk Latv. SSR*, **N 3**, 33 (1959).
  - 28) V. V. GROMOV, *At. energiya*, **26**, 250 (1969).
  - 29) V. V. GROMOV, Doctor Thesis, Institute of Physical Chemistry, USSR Academy of Sciences, Moscow (1969).
  - 30) K. K. AGLINTSEV, *Dozimetriya ioniziruyushchikh izlucheni*, 166, Gosizdat Tekhn. teoret. Lit., M-L (1950).
  - 31) V. I. SPITSYN, V. V. GROMOV, V. S. ARAKELIAN, and N. G. LYSENKO, *Dokl. Akad. Nauk SSSR*, **182**, 390 (1968).
  - 32) B. M. VUL, F. T. T., **3**, 2264 (1961).
  - 33) B. M. VUL, *Dokl. Akad. Nauk SSSR*, **139**, 1339 (1961).
  - 34) T. M. PROCTOR, *Phys. Rev.*, **116**, 1436 (1959).



V. V. GROMOV and V. I. SPITSYN

- 35) V. V. GROMOV, S. N. OZIRANER, V. I. SPITSYN, and A. A. MINAEV, Zhur. fiz. Khim. (in press, 1970).
- 36) V. I. SPITSYN, G. N. PIROGOVA, A. A. SOPINA, and E. Kh. ENIKEEV, Dokl. Akad. Nauk SSSR, **186**, 1358 (1969).
- 37) V. V. GROMOV, V. I. TROFIMOV, V. M. LUK'YANOVICH, and V. I. SPITSYN, Dokl. Akad. Nauk SSSR, **178**, 1307 (1968).
- 38) V. V. GROMOV, and T. N. BESPALOVA, Khimiya Vysokykh Energii, **2**, 263 (1968).
- 39) V. V. GROMOV, and V.S. KRYLOV, Dokl. Akad. Nauk SSSR, **192**, 123 (1970).