IN THE LABORATORY

Routine Analysis of the Alpha Activity of Protactinium Samples

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The classical method of measuring the alpha activity of protactinium consists of the following procedure: The samples containing protactinium are painted on metal discs as uniformly as possible and in extremely thin layers in order to provide minimum absorption for the alpha particles of protactinium. The current produced by the alpha particles of the sample is measured by an electrometer or electroscope and compared with that of a uranium standard.

The current obtained by the protactinium sample, expressed in e. s. u., is used for the determination of the number of alpha particles emitted by the sample:

$$J_{e.s.u.} = N\cdot e\cdot n, \tag{1}$$

where N is the number of alpha particles emitted; e, the electric charge of an ion; and n, the number of ion pairs produced by an alpha particle of protactinium along its path through the ionization chamber.

Knowing the half-life of protactinium, the number of alpha particles emitted by, for instance, 1 mg. of protactinium can be calculated. One mg. of protactinium emits in total (whole sphere) 1.85×10^6 alpha particles/second.

The fraction $\frac{2N}{1.85 \times 10^6}$ gives the amount of protactinium (in mg.) which the sample contains, provided that the absorption of the layer of foreign material can be neglected. However, this assumption can actually never be realized completely. Therefore, the maximum value of $\frac{2N}{1.85 \times 10^6}$ corresponding to a zero absorption has to be determined by extrapolation, measuring samples of decreasing total weight. The ratio $\frac{\text{current}}{\text{weight of sample}}$ increases with decreasing layer thickness and, plotting these values versus the weight of the samples, we obtain a curve whose intersection with the ordinate gives us the actual value of the protactinium content per unit weight of sample material.

It is evident that this method is time consuming and subject to various errors, especially if the sample is very dilute with respect to the protactinium content. Moreover, this method presents a great many difficulties in plants where other radioactive products are present, especially alpha radiators of shorter half-life than protactinium, because even the slightest contamination introduced in the process of painting the discs can give rise to great errors. If, for instance, in a sample of 1 mg. of protactinium/gram, only 10^{-6} mg. of polonium is present, the error in the determination of the protactinium content would amount to more than 10 per cent.

In order to overcome these difficulties, we decided to apply for routine measurements the method of measuring the samples in thick (alpha-saturated) layers, especially since it affords less handling of the material, which, after suitable grinding, is filled into special, strictly uncontaminated dishes.

The theory of the ionization, above alpha-saturated layers of material, due to its emission of alpha particles was given by R. D. Evans (*1*). The number of ion pairs, Nip, produced by the alpha particles emitted by 1 cm.^{2} of the sample layer is given by

$$\text{Nip} = \frac{J}{e\cdot f}, \tag{2}$$

where J is the current due to the alpha particles of the sample; e, the charge of the ions; and f, the surface of the sample. As these samples contain much more material than the painted layers, a slight contamination does not affect the accuracy in such high degree and can be recognized more easily be refilling. The relationship between this number and the content of protactinium per gram of the sample Pa is given by the equation

$$\text{Nip} = \frac{L}{A_{Pa}}\cdot(EKR\lambda_{Pa}Pad\mu). \tag{3}$$

In this equation L is the Avogadro number and A_{Pa} the atomic weight of protactinium; the radioactive constants, EK, R, λ, Pa which refer to the specific ionization and range of the alpha particles and to the disintegration constant of protactinium, have been tabulated (*1*). Therefore, Pa, the protractinium content per gram of sample, could be calculated if d, the density of the sample, and μ, the absorption coefficient for the alpha particles, *i.e.* the ratio between the alpha particle range in the solid to its range in air, are known.

The density of the sample can be obtained experimentally, but would require separate measurements; on the other hand, an experimental determination of μ presents quite great difficulties.

To overcome these difficulties and to establish simultaneously and once for all the value of the product "μd" for our special samples (ZrP_2O_7), we proceeded as follows:

(1) Very careful measurements were made using the first-described method of thin-layer samples in order to obtain directly the content of protactinium per gram of the sample. Thereafter, exactly the same sample of material was used in an alpha-saturated layer, and the product "μd" was calculated, substituting for Pa in Equation 3 the value found in the thin-layer experiments. The average value obtained in four series of measurements is: $\mu d = 2.36\cdot 10^{-3}$.

(2) An accurately known quantity of polonium was added to ZrP_2O_7 samples of low protactinium content and later also

to unactivated ZrP_2O_7 samples.[1] The polonium was (a) added in form of a solution (HCl) and (b) precipitated on ferric hydroxide $[Fe(OH)_3]$.

In case (a) the exact quantity of polonium was determined by spinning a nickel foil in an aliquot part of the polonium solution used for the experiment and by measuring this foil by photoelectric or ionization methods. The solution was thoroughly mixed with the zirconium sample and dried at only slightly elevated temperatures until the original weight was reached. Because of the high specific activity of polonium, no correction factor with respect to density changes have to be made. Two series of measurements gave the average value of $\mu d = 2.20 \times 10^{-3}$.

In case (b) a nickel foil with a well-known amount of polonium was dissolved and the polonium precipitated on $Fe(OH)_3$. By this procedure about 5 per cent of the original polonium quantity is lost, and this factor, as well as the change in density by adding approximately 10 mg. of $Fe(OH)_3$ to 1 gram of sample material, was taken into consideration. Six series of measurements gave the average value of $\mu d = 2 \cdot 10^{-3}$; this value is smaller than that in (a) and 11 per cent smaller than in case 1. In three series out of these six we measured only the quantity of added $Fe(OH)_3$ before the precipitation with polonium and did not observe the weight after having dried the sample. It might be that, because the drying was not complete, the value of density appears too small. On the other hand, the values of series 1 may be too high, as the samples were very dilute with respect to protactinium, so that the active material is covered with an absorption layer.

The average value of the three sets of measurements, $\mu d = 2.22 \cdot 10^{-3}$, was accepted and introduced in Equation 3, which now allows us simply to calculate the content of protactinium precipitated on ZrP_2O_7, if the sample is measured in alpha-saturated layers.

For routine measurements, the necessary calculations can be still further simplified by comparing the ionization above the protactinium samples with that appearing over the uranium standard. If the ionization effect is the same in both cases, assuming equal surfaces of the preparation, we find that the content of protactinium in the zirconium sample is 1.3×10^{-5} gram/gram of sample. Therefore, the protactinium content in grams per gram of sample can be expressed by

$$Pa = \frac{J_{ZrP_2O_7+Pa}}{J_{U_3O_8}} \cdot 1.3 \cdot 10^{-5} \text{ gram.}$$

The above considerations, of course, are valid only if the radioactive element is always separated by the same chemical processes, a common occurrence in routine work of this nature.

The use of polonium for the determination of the factor μd in the case of protactinium samples is self-indicating because of the nearly equal range of the alpha particles emitted by these radioactive elements. However, the same procedure can be recommended in the case of other alpha emitters of appreciable half-life, e.g. plutonium and other transuranic elements, since the addition of polonium, due to its high specific activity, does not alter the absorption in the sample.

Reference

1. EVANS, R. D. *Phys. Rev.*, 1934, 45, 29.

[1] We are grateful to W. R. Horn and M. Pavey, of International Rare Metals Refinery, Inc., for the preparation of all of these samples.

A Simple Quantitative Method for Intravenous Injection of Small Volumes of Fluid

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The intravenous injection of small, accurately measured quantities of solutions is frequently required in physiological investigations. In most of the reported studies the technic has consisted of using a syringe calibrated to contain the solution and final rinsing of the syringe with blood several times to insure complete delivery (*1*). In the course of a study of the distribution of radioactive and nonradioactive substances injected intravenously into small infants, a simple technic was adopted which has several advantages.

The arrangement of the apparatus, which consists of two ordinary glass syringes with Luer locks and a metal three-way stopcock,[2] is shown in Fig. 1. One or more solutions are delivered from sterile volumetric pipettes of desired capacity into the upright syringe barrel, A, with the stopcock handle, B, in position 1. After the vein is entered, the handle is turned to position 2, and the plunger of the horizontal syringe, C, is withdrawn, bringing the solution into the barrel. Returning the handle to position 1 and advancing the plunger into the barrel delivers the solution through the needle into the vein. The upright barrel can then be filled with saline to the height of the original fluid level and washed through the apparatus as many times as desired by repeating the above maneuvers.

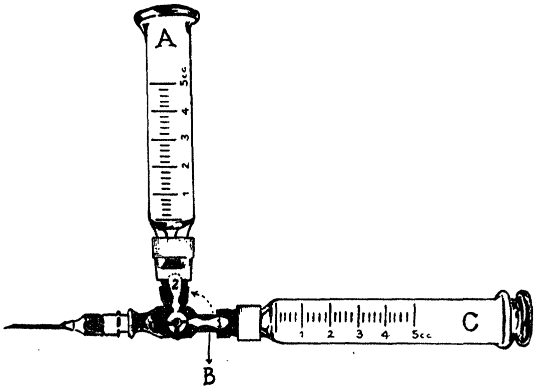

FIG. 1

One-ml. aliquots of four different solutions (thiocyanate, creatinine, mannitol, and sodium-*p*-aminohippurate[3]) were pipetted into the upright barrel of the assembly and then delivered through the apparatus into 2,000-ml. flasks. With two 4-ml. rinsings of the assembly into the flasks, concentrations of the four substances were found to be identical with those in flasks into which similar aliquots were delivered

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[1] With the technical assistance of Dorothy Weber.

[2] This is the type commonly used for intravenous injections in pediatrics and is available from Becton-Dickinson & Company, Rutherford, New Jersey.

[3] The mannitol and sodium-p-aminohippurate were kindly supplied to us by W. P. Boger, of Sharp & Dohme, Inc., Philadelphia.