

On the Chemical State of ^{32}P and ^{33}P in a Sulphur Target

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The yields of ^{32}P , ^{33}P and ^{35}S by the irradiation of sulphur in the Norwegian D_2O -reactor at Kjeller have been determined for different sample positions in the reflector.

It was found that a large amount of ^{32}P and ^{33}P but hardly any ^{35}S followed the insoluble S_μ -fraction of the sulphur samples on dissolving in CS_2 . Adsorption can be responsible for this phenomenon but an alternative explanation, assuming S_μ formation in "displacement spikes", is also given.

The initial S_μ content of the sublimated sulphur samples decreased on reactor irradiation.

No significant chemical difference was found between ^{32}P and ^{33}P after extraction from a solution of irradiated sulphur in CS_2 to a water phase.

The separation factor for a process where two atoms with recoil ranges r_1 and r_2 recoil from a spherical particle into a catcher phase surrounding the particle has been calculated and applied to the case of ^{32}P and ^{33}P recoiling from colloidal sulphur particles.

For a long time, ^{32}P has been one of the most frequently produced radioisotopes in nuclear reactors and the only isotope used as a radiotracer for phosphorus. Usually the isotope is made by irradiation of sulphur according to the endothermal reaction $^{32}\text{S} (n, p)^{32}\text{P}$ (I). Since the threshold energy for the reaction is 954 keV, fast neutrons must be used (Fig. 1).

In 1951, however, the new radioisotope ^{33}P was discovered in reactor-irradiated sulphur and the exothermal reaction $^{33}\text{S} (n, p)^{33}\text{P}$ (II) was found to be responsible for its formation¹⁻⁴. Now, ^{33}P possesses properties which make it suitable for tracer applications and it is therefore of interest to produce as pure samples of ^{33}P as possible.

When preparing ^{33}P by irradiation of sulphur with neutrons, the only "impurities" introduced are ^{32}P and a large amount of ^{35}S formed by the exothermal reaction $^{34}\text{S} (n, \gamma)^{35}\text{S}$ (III). This last "impurity", however, can always be removed chemically. Therefore the main problem is to get a high $^{33}\text{P}/^{32}\text{P}$ ratio in the irradiated samples.

Now Westermarck³ pointed out that ^{33}P , in contrast to ^{32}P , can be produced also by thermal neutrons, as seen from cross sections pictured in Fig. 1.

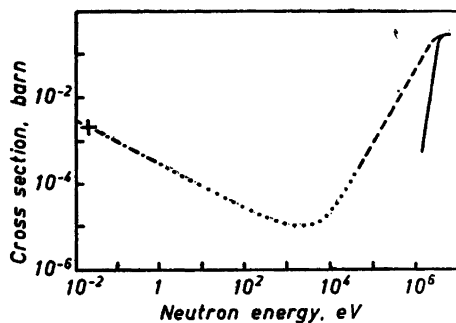


Fig. 1. Cross sections of the reactions $^{33}\text{S}(n, p)^{32}\text{P}$ (I) and $^{33}\text{S}(n, p)^{33}\text{P}$ (II).

- experimental cross section of reaction I according to Klema and Hanson⁵.
 - - - cross section of reaction II calculated from the continuum theory⁶ by B. Elbek⁷.
 + experimental cross section of reaction II according to Westermarck⁸.
 ···· cross section of reaction II according to the "1/v law" for thermal neutrons.

The cross sections indicate that an irradiation of a sulphur sample in the peripheral parts of a reactor where the thermal flux is high (compared with the fast flux) would give an optimum value for the $^{33}\text{P}/^{32}\text{P}$ ratio. One is thus forced to irradiate in a position where the activity of ^{33}P per gram sulphur becomes low. Furthermore, sulphur contains 95.1 % ^{32}S but only 0.75 % ^{33}S . This makes it difficult to establish a high $^{33}\text{P}/^{32}\text{P}$ ratio at all. Any method, chemical or physical which makes it possible to increase this ratio is therefore of value.

If, in order to get a high $^{33}\text{P}/^{32}\text{P}$ ratio, a sulphur sample is irradiated in a position where the ratio of thermal to fast flux is high, then the two isotopes are formed by neutrons with essentially different energies. This means that the most probable recoil energies of the isotopes are also different. As a result they might appear in different chemical states in the target, an effect which could be used to increase the $^{33}\text{P}/^{32}\text{P}$ ratio.

In order to study the $^{33}\text{P}/^{32}\text{P}$ ratio as a function of the distance from the centre of the reactor and the chemical state of the isotopes, several samples of sulphur were irradiated in the Norwegian D_2O -reactor at Kjeller.

EXPERIMENTAL

Measurement of sample activities

The content of ^{33}P , ^{32}P and ^{35}S in the samples was determined by absorption measurements in a standard geometry using aluminium foils as absorbers and an end-window G.M.-tube as detector (1.4 mg/cm² mica window). A thick aluminium backing established saturation backscattering in all cases and counting rates were corrected for dead time and background. Absorption curves were plotted according to Fig. 2, where I_0 is the extrapolated counting rate at 0 mg/cm². From this plot, the different activities were obtained using standard absorption curves for ^{32}P and ^{33}P . The ^{32}P curve was determined by absorption measurements on a nearly pure ^{32}P sample in the standard geometry. The corresponding ^{33}P curve was the result of measurements on samples containing ^{32}P , ^{33}P and very small amounts of ^{35}S .

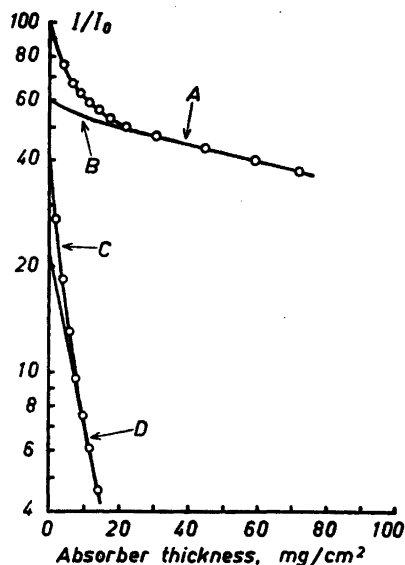


Fig. 2. Absorption curves in the standard geometry.

- A = ^{32}P , ^{33}P and ^{35}S .
 B = ^{32}P standard curve.
 C = ^{32}P and ^{35}S .
 D = ^{32}P standard curve (half thickness $6 \text{ mg}/\text{cm}^2$).

The different activities obtained in this way were corrected for half life (*i. e.* the activities compared refer to a common time) and when necessary also for self-absorption in the samples. This method permitted a good comparison of the activities of the different sulphur samples. Since, however, no pure sample of ^{32}P was available, it was only possible to determine the absolute values of the ^{32}P activity with good accuracy.

Yields of the isotopes for different positions in the reactor

Since the ^{35}S activity was very high compared with the ^{32}P activity, it was not possible to determine the ^{32}P and ^{35}S contents by a direct measurement of the activities of the irradiated samples. From such measurements the ^{32}P activity and the sum of the ^{32}P and ^{35}S activities were obtained.

Samples for irradiation in the reactor were, however, prepared from sulphur containing a small amount of S_μ (*i. e.* amorphous sulphur)⁸. When dissolving such a sample in carbon disulphide and centrifuging the solution, it was found that a large amount of the radiophosphorus followed the insoluble S_μ -fraction. Since there was no or only a very small enrichment of ^{35}S in the S_μ -fraction (Table 1), this is a very simple way of separating a large amount of ^{32}P and ^{33}P from ^{35}S , *i. e.* samples were obtained in which the $^{33}\text{P}/^{32}\text{P}$ ratio could be determined. In addition, these samples had so low a weight per unit area ($<1 \text{ mg}/\text{cm}^2$) that self-absorption for ^{32}P and ^{35}S could be neglected. If ^{32}P and ^{33}P , as seems reasonable, follow S_μ in proportion to their concentrations, then the $^{32}\text{P}/^{33}\text{P}$ ratio in the S_μ -fraction and the first measurement (mentioned above) give the amounts of ^{32}P and ^{35}S in the samples. The results are plotted in Fig. 3. They refer to irradiations in the reflector. Samples irradiated in the reactor tank are omitted, since the ^{32}P content was too low to be determined. The samples were irradiated for about 14 days at a reactor effect of 200–250 kW and the results refer to the end of the irradiation.

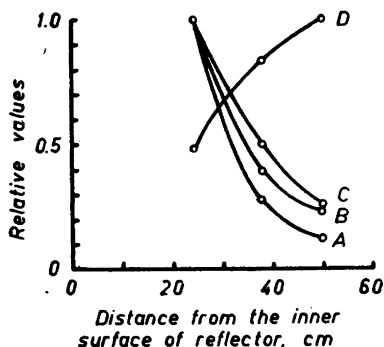


Fig. 3. Yields of the isotopes and the ratio $^{33}\text{P}/^{32}\text{P}$ for different positions of irradiation in the reflector.

A = ^{32}P	1.0	corresponds to	3	$\mu\text{C/g}$	sulphur
B = ^{35}S	1.0	»	»	32	»
C = ^{32}P	1.0	»	»	0.3	»
D = $^{33}\text{P}/^{32}\text{P}$	1.0	»	»	0.25	»

The carrier action of the S_μ -fraction on ^{32}P and ^{33}P

As previously mentioned, a large part of ^{32}P and ^{33}P was carried by the S_μ -fraction. The separation of S_μ was performed as follows:

Irradiated sulphur (0.5–0.6 g) was dissolved in 10 ml carbon disulphide. The insoluble S_μ was then collected on an aluminium foil by centrifuging in a special centrifuge tube. Before weighing and measuring the activity, the foil was placed for 5 min in pure carbon disulphide. The results are shown in Table 1.

Table 1. The carrier action of the S_μ -fraction of different samples on ^{32}P , ^{33}P and ^{35}S .

Sulphur sample	Distance from centre of reactor cm	% S_μ in sample	% Activity carried by S_μ		
			^{32}P	^{33}P	^{35}S
Subl.	Unirradiated	0.47	—	—	—
Cryst.	»	0.02	—	—	—
Subl.	48	0.08	30	—	0.4
»	60	0.08	32	—	0.3
»	124	0.09	62	59	0.04
»	150	0.09	60	71	0.4
Cryst.	48	0.02	19	—	0.2
»	138	0.02	14	13	0.01

The treatment of the foils with CS_2 , as described, resulted in a marked decrease in inactive sulphur and ^{35}S . The influence on ^{32}P and ^{33}P , however, was very small.

Some foils were placed for an additional 24 h in CS_2 . Their weights, however, were unchanged and their total activities decreased by only 2–6 %; *i. e.* the results in Table 1 may be regarded as referring to S_μ .

A number of experiments was performed in order to find if ^{32}P and ^{33}P , in the form of phosphorus or a compound of phosphorus, were reversibly adsorbed on S_μ . This was done by studying the displacement of radiophosphorus from S_μ by phosphorus and different

DISCUSSION

In Fig. 3, A is a graph both of the yield of ^{32}P and also of the decrease of the fast neutron flux in the reflector. This is because reaction I requires fast neutrons. The fast flux, however, decreases more rapidly than the thermal flux and the reactions II and III have relatively large cross sections for thermal neutrons. The yields of ^{33}P (C) and ^{35}S (B), therefore, decrease more slowly and the $^{33}\text{P}/^{32}\text{P}$ ratio (D) increases with distance from the reactor core.

The most interesting result is that a large amount of radiophosphorus but almost no ^{35}S followed the S_μ -fraction (Table 1). Approximately equal amounts of ^{32}P and ^{33}P follow S_μ but the amounts seem to decrease with decreasing S_μ content of the irradiated sulphur.

At first sight this may be supposed to be due to adsorption of either radiophosphorus or a chemical compound of radiophosphorus on S_μ . However, addition to the CS_2 solution of phosphorus or some of the compounds most probably formed by radiophosphorus in this case had only a small effect (except for H_3PO_3) on the carried activity (Table 2). The increase in amount carried was also small after adding S_μ before dissolving the sample in CS_2 . This seems to indicate that adsorption on S_μ of radiophosphorus in these chemical forms cannot alone be responsible for the carrying effect. Adsorption can, however, not be excluded. In any case it may be of value to discuss an alternative explanation:

A recoiling atom is assumed to melt or vaporize a small volume at the end of its track in a solid substance⁹. The local temperature in this "displacement spike" is very high and the quenching is very rapid¹⁰. This means that "displacement spikes" may also be produced in sulphur by recoiling ^{32}P , ^{33}P and ^{35}S atoms. It is further assumed that small aggregates of S_μ are formed by the rapid quenching of the "displacement spikes", in analogy with the formation of S_μ by the rapid quenching of sulphur vapour or liquid sulphur⁸. Radiophosphorus and ^{35}S trapped in these aggregates are difficult to extract by CS_2 and in the chemical operations the aggregates will follow the macro amounts of S_μ in the sample, *i. e.* the radioisotopes follow the S_μ -fraction.

Only radiophosphorus and hardly any ^{35}S follows the S_μ -fraction (Table 1). As seen from the following calculations, an explanation may be found in the different volumes of the "displacement spikes" produced.

The most probable recoil energies of ^{32}P and ^{33}P can be computed from the equation¹¹

$$E_r = \frac{m_p}{M + m_p} \left[E_n \left(1 - \frac{m_n}{M + m_p} \right) + Q \right] \quad (1)$$

where E_n is the neutron energy, m_n the mass of the neutron, m_p the mass of the proton and M the mass of the recoiling atom. The Q -values of the reactions are 925 (I) and 517 (II) keV¹². The equation is valid if the compound nucleus has lost its recoil energy before emitting the proton¹¹. This may be assumed to happen since the reactions take place in a solid substance¹¹.

When irradiating in the reflector, the ^{33}P atoms are most probably produced by neutrons having an energy of 0.025 eV, *i. e.* the most probable energy of the

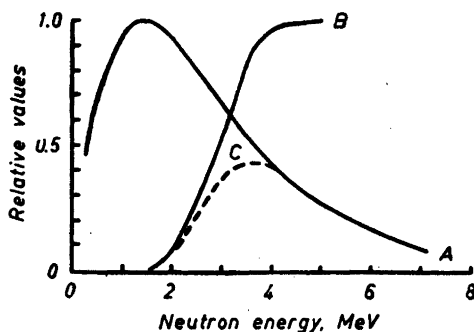


Fig. 4. Graphical determination of the most probable recoil energy of ^{32}P atoms produced in the reaction $^{32}\text{S}(n, p)^{32}\text{P}$ (I) by neutrons of the fission spectrum (fast neutrons).

- A = the flux of fission neutrons. The maximum of A is at a neutron energy of 1.4 MeV.
 B = cross section of reaction I. The top of B corresponds to about 0.3 barns.
 C = the product of flux and cross section. The maximum of C gives the most probable recoil energy of the ^{32}P atoms, i. e. about 3.5 MeV.

thermal flux at 20° C. This value gives a most probable recoil energy for the ^{33}P atoms of 15 keV.

It is more difficult to estimate the corresponding energy for ^{32}P . If the fast neutrons had the energy distribution of the fission spectrum ¹³, a graphical determination (Fig. 4) gives a value of about 3.5 MeV. The maximum of the fast flux in the reflector, however, is very likely at a lower energy. This means that ^{32}P is most probably produced by neutrons having energies between 3.5 and 0.954 MeV (the threshold value). Assuming 2.5 MeV for the neutron energy, the most probable recoil energy is 45 keV.

The recoil energy of the ^{35}S atoms can be calculated from the equation ¹⁴

$$E_r = \frac{536 E_\gamma^2}{M} \quad (2)$$

where E_r is the recoil energy in electron volts, M the mass of the recoiling atom in atomic mass units and E_γ the energy of the emitted gamma ray in MeV. The number and the energy of the emitted gamma rays per neutron capture in reaction III, however, have not been determined. By extrapolation from a diagram by Kikuchi *et al.*¹⁵, the maximum energy of the gamma rays is estimated to be 6 MeV, giving the ^{35}S atoms a maximum recoil energy of 0.55 keV. Since the mean number of gammas emitted per neutron capture in even Z -even N nuclei is about 1.7¹⁶, this value for the recoil energy is too high.

Maximum values of the melted volumes in the three cases can be crudely estimated by assuming that the recoil energy is entirely available for melting (this can be done since the recoil energy is of the same order of magnitude as the transition energy for melting as defined by Brinkman⁹) and that the energy necessary to melt sulphur at atmospheric pressure (about 0.02 eV/atom)⁸ can be used. Under these assumptions the melted volumes produced by ^{32}P , ^{33}P and ^{35}S contain about 2×10^6 , 8×10^5 and 3×10^4 atoms. Since each

sulphur atom occupies a volume v of 26 \AA^3 , these volumes, assumed to be spherical, would have diameters of about 460, 340 and 110 \AA , respectively. Evidently a recoiling ^{32}P atom melts a volume more than ~ 70 times larger than that melted by a recoiling ^{35}S atom. The resolidification occurs with the surrounding S_α lattice acting as a crystallization nucleus. It is therefore most probable that a S_μ aggregate is formed in the centre of the melted volume — the probability of S_μ -formation increasing with the size of that volume. Assuming that the volumes melted by recoiling ^{35}S atoms are too small to produce S_μ -aggregates will explain why ^{35}S does not follow the S_μ -fraction to any great extent.

It may be pointed out that S_μ is also produced by recoil (knock-on) atoms formed in the slowing down of fast neutrons. An assumed formation of S_μ in these ways seems to conflict with the decrease in S_μ by reactor irradiation, the sublimated samples losing about 80 % of initial S_μ content whereas no change is found in the crystallized samples (Table 1).

In the first case the transformation of S_μ to S_α is evidently more effective than the assumed formation of S_μ in "displacement spikes". It is difficult to estimate the amount of S_μ formed but reasons for being small are:

1. Only a part of the energy of the primary knock-on atoms is effective in melting (*i. e.* their initial energy is larger than the transition energy for melting).

2. Only a small part of a melted volume is assumed to form S_μ .

It is known that S_μ spontaneously transforms to the stable modification S_α . The transformation can be accelerated by heating, irradiation by X-rays and so on 8 . In our case irradiation must be responsible for the transformation since the temperature of the reflector was only 10°C . Maier-Leibnitz 17 and Erbacher 18 found a similar effect in the conversion of white phosphorus to the stable red form by the radiation from a Rn-Be source.

The transformation in the reactor of S_μ to soluble S_α means that a part of the radiophosphorus atoms trapped in S_μ are set free. It can be calculated (Appendix 1) that about 50 % are set free in this way assuming that the amount of S_μ formed can be neglected compared with the initial S_μ amount, *i. e.* that the rate of transformation is that shown by S_μ in the sublimated samples.

From the results given in Table 3 it is seen that the maximum amount of ^{32}P extracted from the CS_2 -solution to the water phase was about 50 %. The amount of ^{33}P extracted was found in all cases to be smaller. Owing to the uncertainty in the determination of ^{33}P , it is hard to say whether this has any significance. The amount not extracted probably corresponds to ^{32}P and ^{33}P carried by S_μ .

The valency of extracted ^{32}P and ^{33}P is about the same, *i. e.* the difference in initial recoil energies seems to have no effect on the valency of the isotopes. This is to be expected since the chemical fate of a recoiling atom is determined when most of the recoil energy is lost, the initial energy being too high for any chemical compound to be formed 19 .

The valency of the isotopes is not significantly altered by the precautions against oxidation. It is, however, probable that more rigorous precautions would have changed the results.

Thus, despite the difference in recoil energy, one can hardly expect to find ^{32}P and ^{33}P in different chemical states in the sulphur target. There they may exist as ions or atoms trapped within the lattice or in chemical combination with other atoms.

It is often assumed that the recoiling atoms are ionized and trapped as positive ions in the target. A good approximation to the upper limit of the kinetic energy at which a moving atom can be considered as un-ionized is

$$E = \frac{M}{m} I$$

where M is the mass of the atom, m is the electronic mass and I

is the ionization energy of the atom. The assumption made is that the atom loses all electrons whose orbital velocities are smaller than that of the recoiling atom. The ionization energy of the 3p electrons of phosphorus atoms is about 11 eV, *i. e.* the recoil energies of ^{32}P and ^{33}P must be at least 640 and 660 keV, respectively, in order to ionize the atoms. Comparing this with the maximum recoil energies ($E_n = 15$ MeV) of ^{32}P and ^{33}P (413 and 443 keV, respectively) shows that the ionic state can be excluded.

Owing to the high "temperature" of the atoms, it is more likely that they enter into chemical combination than that they become trapped as atoms in the target.

The phosphorus compounds most probably found in a sulphur target should be the sulphides. The concentration of radiophosphorus atoms, however, is so low (about 10^{13} atoms/cm³) that the sulphides (all of them containing more than one P-atom) formed may be supposed not to have a stoichiometric composition. The atoms may also react with impurities in the target. In fact the chemical state of radiophosphorus in the target must to a large extent be a function of its purity and if the intention is to determine this state it must be very well protected in the separation process.

A better way of increasing the $^{33}\text{P}/^{32}\text{P}$ ratio than the chemical one seems to be a direct utilization of the differences in recoil range of the two isotopes in analogy with the separation of ^{198}Au from ^{197}Au by Magnusson²⁰ and ^{11}C from ^{12}C by Morinaga and Zaffarano²¹. In our case a similar experiment is expected to be more difficult since both isotopes recoil.

An expression for the separation factor s has, however, been calculated (Appendix 2) for the case of two atoms with recoil ranges r_1 and r_2 ($r_2 > r_1$) recoiling from a spherical particle of radius R into a catcher phase surrounding the particle. The separation factor is defined as follows:

$$s = \frac{(N_1/N_2)_I \text{ (When } r_1 \neq 0 \text{ and } r_2 \neq 0)}{(N_1/N_2)_{II} \text{ (When } r_1 = r_2 \text{ or } R = \infty)} = \frac{(R + r_1/4) (R - r_1/2)^2}{(R + r_2/4) (R - r_2/2)^2} \quad (3)$$

where N_1 and N_2 are the concentrations of the atoms in the particle. The separation factor varies between the R -axis ($r_1 = r_2$) and the solid curve ($r_1 = 0, r_2 \neq 0$) in Fig. 5. (The latter case corresponds to the separations made by Magnusson²⁰ and Morinaga and Zaffarano²¹). When $r_1/2 < R < r_2/2$, then all atoms with recoil range r_2 will leave the particle and the separation factor will be infinity. When $R < r_1/2$ both kinds of atoms will leave the particle. It is seen that s is more sensitive to variations in R than to variations in the value of r_2/r_1 .

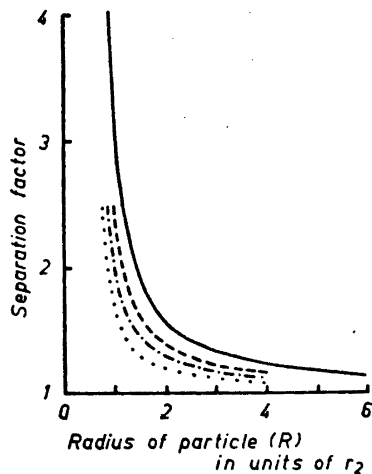


Fig. 5. The recoil separation factor for different values of r_2/r_1 (the ratio between the recoil ranges) plotted as a function of particle radius. The particle radius is given in units of r_2 .

—	R — axis	r_2/r_1	= 1.0
⋯	⋯	»	= 1.5
- · - · -		»	= 2.0
- - -		»	= 3.0 (our case)
—		»	= ∞ ($r_1 = 0, r_2 \neq 0$)

The assumptions underlying (3) may be realized when ^{32}P and ^{33}P are produced for example by neutron irradiation of a sulphur sol. To determine s in this case the recoil ranges of ^{33}P and ^{32}P (r_1 and r_2) must be calculated. This can be done from the following equation²² which is valid for $A_1 > A_2$ and thus probably only gives the right order of magnitude in this case:

$$r = 60 \frac{A_1 + A_2}{A_1} \sqrt{\frac{Z_1^{2/3} + Z_2^{2/3}}{Z_1 Z_2}} A_2 E_r 1/p \quad (4),$$

where r is the recoil range in Å, E_r is the recoil energy in keV and p is the density of the absorber. (1 refers to recoil atom and 2 to absorber atom). Using the recoil energies calculated above (45 and 15 keV respectively), the most probable recoil ranges of ^{32}P and ^{33}P are 1 230 and 400 Å, respectively, *i. e.* $r_2/r_1 \sim 3$. The separation factor for this case is shown by the dashed curve in Fig. 5. Since sulphur sols with a particle radius of about 2 000 Å can be prepared⁸, it would be possible to get a s -value of about 1.6. Trouble may, however, arise from the fact that the size of the particles in a sol increases with time and nothing is known about the behaviour of a sulphur sol in a strong radiation field. A disadvantage is that large volumes must be irradiated since the concentration of sulphur in such a sol is only a few percent. Also a large amount of ^{33}P recoils away from the sulphur particles, the loss being 48 % when $r_2 = 3r_1$ and $R = r_2/2$.

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APPENDIX 1

Assume that R aggregates are formed and kFN are destroyed in the time interval from t to $t + dt$, then

$$dN = (R - kFN) dt = (R - KN) dt \quad (1)$$

where N is the number of aggregates at the time t and F the neutron flux.

Integration gives:

$$N = N_0 e^{-Kt} + R/K (1 - e^{-Kt}) \quad (2)$$

where N_0 is the initial number of S_μ aggregates.

Assuming that the second term in (2) can be neglected compared with the first, then

$$e^{-Kt_1} = 1/6 \quad (3)$$

using the value for the decrease of S_μ in the sublimated samples in a time t_1 (Table 1).

The fraction of radiophosphorus set free in the same time is

$$f = \frac{Rt_1 - R/K(1 - e^{-Kt_1})}{Rt_1} = 1 - 1/Kt_1 (1 - e^{-Kt_1}) \quad (4)$$

Combining with (3) gives $f = 0.54$.

APPENDIX 2

From a spherical particle will escape

$$n = \int C_1 w dV \quad \text{recoil atoms/sec} \quad (1)$$

where C_1 is the rate of production of recoil atoms (atoms/cm³ sec), dV the volume element and w the solid angle within which recoil atoms produced in dV escape from the particle.

Now $dV = r^2 \sin \theta d\theta d\phi dr$ and it can be shown that

$$w = \frac{(r_1+r)^2 - R^2}{4r_1 r}, \quad (0 < r_1 < 2R) \quad (2)$$

where r_1 is the recoil range, r the distance from the centre of the sphere to dV and R the radius of the particle. Then (1) transforms to

$$n = C_1 \int_0^R \int_0^{\pi} \int_0^{2\pi} \frac{\pi 2\pi R}{4r_1 r} \frac{(r_1+r)^2 + R^2}{4r_1 r} r^2 \sin \theta d\theta d\phi dr = \pi C_1 r_1 (R^2 - r_1^2/12) \quad (3)$$

when $0 < r_1 < R$ and the same result is obtained when $R < r_1 < 2R$.

If two different atoms with recoil ranges r_1 and r_2 recoil from the particle then the ratio between their concentrations in the particle after a certain time would be

$$(N_1/N_2)_I = C_1/C_2 \frac{4/3R^3 - r_1(R^2 - r_1^2/12)}{4/3R^3 - r_2(R^2 - r_2^2/12)}, \quad (0 < r_1 < r_2 < 2R) \quad (4)$$

If $r_1 = r_2$ or $R = \infty$, then $(N_1/N_2)_{II} = C_1/C_2$, *i. e.* no separation would take place and a suitable definition of the separation factor would be

$$s = \frac{(N_1/N_2)_I}{(N_1/N_2)_{II}} = \frac{4/3R^3 - r_1(R^2 - r_1^2/12)}{4/3R^3 - r_2(R^2 - r_2^2/12)} = \frac{(R+r_1/4)(R-r_1/2)^2}{(R+r_2/4)(R-r_2/2)^2} \quad (5)$$

(It is assumed that the recoil atoms are stable but the same result is obtained for radioactive atoms).

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