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EFFECT OF IRRADIATION TEMPERATURE ON THE DISTRIBUTION OF ^{32}P-ACTIVITY IN NEUTRON IRRADIATED ADENOSINE TRIPHOSPHATE

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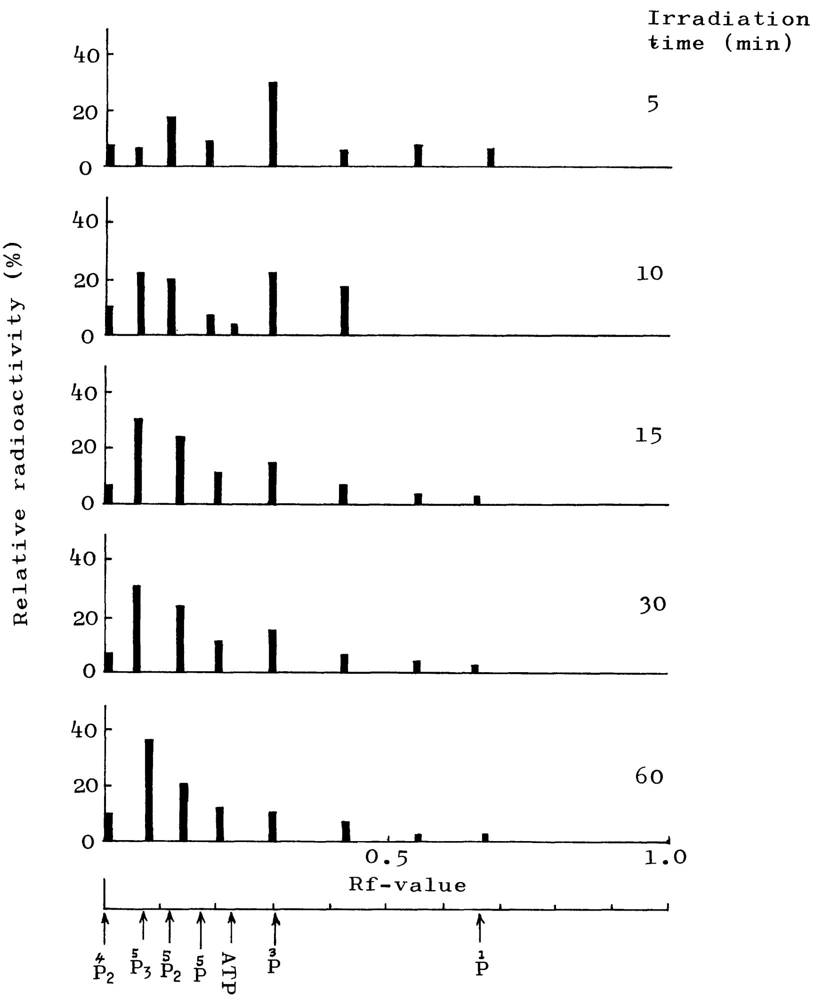
The radioactivity distribution in reactor-irradiated adenosine triphosphate changed with irradiation time ($5\sim60$min). This was mainly due to differences in the irradiation temperature, which was estimated by melting points of carboxylic acids and amides.

The radioactivity distribution in neutron-irradiated materials are influenced by many factors such as irradiation conditions (neutron flux, γ-dose and irradiation temperature), thermal annealing after the irradiation and chemical treatments at the analysis. Then, on discussing results obtained by various workers, we have to know details on the factors. Sellers et al.[1] irradiated phosphorus oxyacids with the neutrons of 10^{13} n/cm$^2\cdot$sec at reactor temperature for 48 hr. The principal recoil product of sodium phosphate was phosphate accompanied by traces of phosphorus and several unidentified products while those of sodium phosphite and hypophosphite were phosphite and hypophosphite with a little phosphate and phosphorus. Lindner and Harbottle[2] separated ten or more recoil products from the neutron irradiated phosphate at reactor, dry-ice and liquid nitrogen temperatures by the paper electrophoresis. Nakamura et al.[3] separated fifteen recoil products including known and unknown compounds when the neutron irradiated alkali and ammonium phosphates were separated by the anion exchange chromatography. Anselmo reported the high percentages of radioactivity due to polyphosphate in irradiated disodium hydrogen phosphate[4] and found the formation of a P-N bond in the neutron irradiated ammonium hypophosphite.[5] Fenger[6] suggested that the discrepancies among results of various workers described above could in most cases be explained by differences in the irradiation temperature from results of the initial distribution of the activity of phosphorus-32 by irradiating ammonium dihydrogen phosphate at liquid nitrogen temperature. However, he did not take care for the effect of room or more higher temperature on the recoil products. Generally, so little attention has been payed to the temperature during irradiation, especially in the case of a so-called reactor temperature.

At the short time irradiation, the temperature of the inside of the irradiation capsule is not in equilibrium with the temperature of the irradiation facilities. On the other hand, at the long time irradiation, the temperature of the inside of the capsule is higher than that of the facilities. To elucidate the factors affecting the activity distribution, we have to know more details on the temperature surrounding the target.

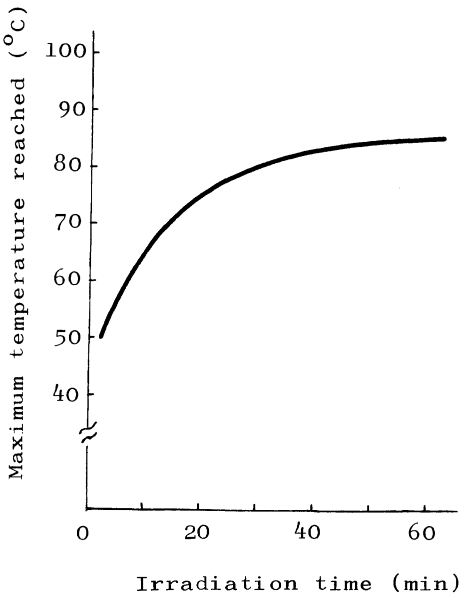
Taking an example from the recoil products of adenosine triphosphate (ATP),



$\overset{1}{P}$:hypophosphite, $\overset{3}{P}$:phosphite, $\overset{5}{P}$:phosphate, $\overset{5}{P}_2$:pyrophosphate, $\overset{5}{P}_3$:triphosphate, $\overset{4}{P}_2$:hypophosphate, ATP:adenosine triphosphate.

Fig.1. Relationship between radioactivity distribution and irradiation time Solvent: n-propanol:ammonia:water (6:3:1 v/v). At the bottom, Rf-values of standard compounds are shown with Blaser and Worm's notation.

we would like to visualize the importance of the temperature surrounding the target during the irradiation. As usual, sodium salt of ATP sealed in a polyethylene capsule was irradiated at the pneumatic tube 3 of the KUR (Kyoto University Reactor) at 5 Mw operation with thermal neutrons of 2.34×10^{13} n/cm^2·sec at reactor temperature for difinite time. After irradiation, all the irradiated samples were chemically treated under the same condition by paper chromatography.[7)] Rf-values of known compounds are shown at the bottom of Fig.1. The positions of ATP or bases were determined by absorption band of 260 mμ. The distribution of radioactivity was examined by autoradiography using Fuji medical X-ray film. Quantitative determination of the radioactivity on the chromatographed strip was made by an autoradioscanner. As shown in Fig. 1, the fractions of lower Rf values in paper chromatograms remarkably increased with increasing irradiation time. This means that the yields of oxyacids in higher oxidation state and of polymers increased as a function of irradiation time. These facts are not explained only by the total neutrons bombarded and γ-dose because such changes with irradiation time have never been observed at 1 Mw operation (4.4×10^{12} n/cm^2 sec).[7)] Thus, we tried to estimate the temperature of the inside of the polyethylene capsule, in which the sample was sealed. As materials with high cross section are not suitable to measurement of the temperature, carboxylic acids and amides were selected and their melting points were used as an indicator of the maximum temperature reached during the irradiation. About



Compounds used	m.p. °C
Lauric acid	44
Myristic acid	54
Palmitic acid	64
Stearic acid	70
Acetamide	81
Capronamide	100

Measurements carried out at 5,10,15,20,30 and 60min.

Fig.2. Relationship between temperature reached and irradiation time

20mg of each compound listed in Fig.2 was sealed in a polyethylene tube, put into a larger polyethylene capsule and irradiated for definite time at the position where ATP was irradiated. From the state of each sample observed under a microscope, we estimated the maximum temperature of the inside of the polyethylene capsule. For example , at 20min irradiation, capronamide and acetamide did not melt but stearic and other acids completely melted. This shows that the temperature is between 70° and 81°C. At 60min irradiation, half amount of acetamide melted showing that the temperature is near 81°C. In this way, we estimated the maximum temperature of the inside of the polyethylene capsule during the irradiation. As shown in Fig.2, the temperature changed from ca. 55°to 85°C at 5 Mw operation as a function of irradiation time. According to Fenger's report,[8] the irradiation at 0°C and at the temperatures of dry-ice and liquid nitrogen gave similar results in recoil products of ammonium dihydrogen phosphate while the irradiation at "reactor temperature" gave the results much different from those of lower temperature irradiations. This shows that the higher the temperature becomes, the more the activity distribution is influenced. Thus, the difference of the temperature observed in this experiment should not be neglected. From these observations, we concluded that the change in the activity distribution with irradiation time was mainly due to different temperature of environment during irradiation.

References

1) P.Sellers, T.Sato and H.Strain, J. inorg. nucl. Chem., 5, 31 (1957).
2) L.Lindner and G.Harbottle "Chemical Effects of Nuclear Transformations" IAEA, Vol. 1, p 485, Vienna, 1961.
3) T.Nakamura, K.Ujimoto, N.Yoza and S.Ohashi, J. inorg. nucl. Chem., 32, 3197 (1970).
4) V.Anselmo, Radiochim. Acta, 4, 203 (1965).
5) V.Anselmo, ibid, 12, 17 (1969).
6) J.Fenger, ibid, 12, 186 (1969).
7) Y.Kiso, M.Kobayashi, Y.Kitaoka, K.Kawamoto and J.Takada, Bull. Chem. Soc., Japan, 40, 2779 (1967).
8) J.Fenger, Risö Report (Denmark), No. 95 (1965).

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