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IRRADIATION OF ENRICHED SULFUR-33
IN HIGHLY THERMALIZED FLUXR. E. Lewis
S. A. Reynolds

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PREPARATION OF PHOSPHORUS-33 BY IRRADIATION OF ENRICHED
SULFUR-33 IN HIGHLY THERMALIZED FLUX

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JANUARY 1967

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PREPARATION OF PHOSPHORUS-33 BY IRRADIATION OF ENRICHED
SULFUR-33 IN HIGHLY THERMALIZED FLUX

R. E. Lewis and S. A. Reynolds

ABSTRACT

Millicurie quantities of carrier-free ^{33}P containing <1% ^{32}P were prepared by neutron irradiation of enriched sulfur targets (61.15 and 68.1% ^{33}S) in a highly thermalized neutron flux. Target irradiation conditions and radionuclide yields are given, and the chemical recovery procedure and analytical methods are described.

INTRODUCTION

The preparation of millicurie amounts of ^{33}P by fast neutron irradiation of isotopically enriched ^{33}S and ^{36}Cl by the $^{33}\text{S}(\text{n},\text{p})^{33}\text{P}$ and $^{36}\text{Cl}(\text{n},\alpha)^{33}\text{P}$ reactions has been reported.¹ However, these nuclear reactions on ^{32}S and ^{35}Cl isotopes in the targets result in the production of ^{32}P which contaminates the ^{33}P product. As a result, the ^{32}P must be depleted by aging before the ^{33}P product is suitable for many applications, a procedure which is inconvenient and also results in significant decay losses of ^{33}P . Westermarck² has reported a thermal neutron cross section of 2.3 mb for the $^{33}\text{S}(\text{n},\text{p})^{33}\text{P}$ reaction, but no thermal neutron cross section for the $^{32}\text{S}(\text{n},\text{p})^{32}\text{P}$ reaction has been reported. Therefore, the reactor production of ^{33}P was investigated by irradiating isotopically enriched ^{33}S using a highly thermalized neutron flux to reduce the amount of ^{32}P contamination produced in the target.

Phosphorus-33 has significantly different nuclear properties than ^{32}P -- the half-life is longer (25.0 vs. 14.3 days)³ and the maximum

energy of the beta particle is less (~ 0.25 vs. 1.7 Mev). These properties indicate possible advantageous uses for ^{33}P as a phosphorus tracer or for direct applications, some of which have been reviewed elsewhere.^{4,5}

MATERIALS AND METHODS

Target Material

Sulfur enriched in ^{33}S by electromagnetic separation (using special techniques of calcium pumping and "charge exchange" receivers)⁶ was used as target material. Isotopic composition of the elemental sulfur targets is shown in Table 1.

Table 1. Isotopic Composition of Sulfur Targets

Isotope	Composition, %	
	Sample 1	Sample 2
^{32}S	29.6	36.96
^{33}S	68.1	61.15
^{34}S	2.27	1.89
^{36}S	<0.05	<0.05

Reactor Irradiation

The enriched samples were irradiated in the D_2O column of the CP-5 reactor (Argonne National Laboratory), a position which has a good thermalized neutron flux spectrum. The thermal flux was measured by the $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ reaction (37 barns) using a half-life of 5.24 yr for ^{60}Co . The fission spectrum neutron flux was measured by the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$

reaction (60 mb). The measured flux values for each irradiation experiment are shown in Table 2.

Table 2. Preparation of ^{33}P from Enriched ^{33}S by Thermal Neutron Irradiation

	Sample 1	Sample 2
Sulfur targets ^a , mg	50	275
Irradiation time, days	47	23
Thermal neutron flux, n/cm ² .sec	2.12×10^{13}	2.66×10^{13}
Fission neutron flux, n/cm ² .sec	1.13×10^{10}	4.0×10^9
Phosphorus-33, mc	0.63	2.44
Phosphorus-32, mc	0.005	0.0077

^aIsotopic composition given in Table 1.

Chemical Recovery of ^{33}P and ^{33}S

After irradiation, the sulfur target was dissolved in dichloroethylene and the phosphorus activity was extracted by an equal volume of 1.0 M HCl. Heating the solution to reflux temperature increased the rate and completeness of the extraction. A 95-98% chemical recovery of ^{33}P was achieved.

Since the supply of sulfur at the level of isotopic enrichment used in these irradiations is very limited, the recovery of target material for reuse in subsequent neutron irradiations is desirable. Essentially complete recovery of sulfur was achieved by evaporation of the dichloroethylene solvent.

Product Analysis

The ^{32}P and ^{33}P content of the product was determined by beta liquid scintillation spectrometry using ^{32}P and ^{45}Ca as calibration standards. Half-lives of 14.3 days for ^{32}P and 25.30 ± 0.05 days for ^{33}P were used in these calculations. An aliquot of the phosphorus activity, as phosphomolybdic acid, was extracted into n-butanol and the aqueous phase was assayed for ^{35}S which was determined to be $<0.5\%$. No other radioactive contaminants were detected in the product solution.

RESULTS

The irradiation conditions for the two samples of the highly enriched sulfur and the yields of ^{33}P and ^{32}P are shown in Table 2. From the data in Table 2, the effective cross section for the $^{33}\text{S}(n,p)^{33}\text{P}$ reaction with thermal neutrons is 2.42 mb in the case of Sample 1 and 2.39 mb for Sample 2. The average of these cross-section values, ~ 2.4 mb, is in excellent agreement with the 2.3 mb thermal neutron cross section found by Westermarck.² The production of millicurie quantities of ^{33}P relatively free of ^{32}P activity is shown to be feasible by irradiation of isotopically enriched ^{33}S in a thermal neutron flux if the epithermal flux is sufficiently low.

REFERENCES

1. R. E. Lewis and T. A. Butler, Production of Phosphorus-33 from Chlorine-36 and Sulfur-33 by Fast Neutron Irradiation, Nucl. Appl. 2(2): 102-5 (1966).
2. T. Westermark, Production of ^{33}P with Thermal Neutrons, Phys. Rev. 88: 573-4 (1952).
3. G. Goldstein and S. A. Reynolds, Specific Activities and Half-Lives of Common Radionuclides, Nuclear Data, 1(5): 435-52 (1966).
4. R. H. Sheline, R. B. Holtzman, and C. Y. Fan, The Nuclide ^{33}P and the ^{32}P Spectrum, Phys. Rev. 83: 919-23 (1951).
5. T. Westermark, I. Fogelstrom-Fineman, and S. Forberg, An Approach to the Production of ^{33}P in Millicurie Quantities, in Proc. First (UNESCO) Intern. Conf., Radioisotopes in Scientific Research, Vol. 1, R. C. Extermann (ed.), Pergamon Press, 1958, pp. 19-31.
6. L. O. Love and W. A. Bell, Review of ORNL Electromagnetic Separations Program, 1965, USAEC Rpt. ORNL-4006, Oak Ridge National Laboratory (August 1966), p. 25.

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