CROSS SECTION OF THE $^{90}SR(n,\gamma)^{91}SR$ REACTION

FOR A D20 MODERATED REACTOR FLUX

M.A. Lone, W.J. Edwards and R. Collins
AECL Research, Chalk River Laboratories
Chalk River, Ontario, Canada. KOJ 1J0
(613)-584-3311

ABSTRACT

Thermal neutron capture cross section of the 9%r(n,y)91 Sr reaction was measured in a D20 moderated neutron flux with an activation technique. The sensitivity of the measurement was improved by using chemical separation of ^{90}Y from 91 Sr for reduction of the intense bremsstrahlung background from beta decay of Y. The value obtained for the cross section is 9.7* 0.7 mb.

1. INTRODUCTION

The beta emitter ^{90}Sr, with a half life of 29 yr. would be one of the major heat sources in a High Level spent-fuel Waste (HLW) repository. The ^{90}Sr can in principle be transmuted by neutron radiative-capture to the 9.5 h half-life91 Sr, but the cross section for this reaction is not well known. Prior to our work two vastly different values were reported in the literature viz 0.8+ 0.5 b in reference 1 and 0.014* 0,0024 in reference 2.

In reference 1 the activity of91 Sr was measured with gamma-ray spectroscopy using a NaI detector. The poor resolution of the detector coupled with a large background from beta induced bremsstrahlung radiation resulted in a relatively large uncertainty. The measurement reported in reference 2 was made with a high resolution Ge spectrometer. For reduction of the background from the ^{90}Y beta-induced bremsstrahhrng, lucite and lead absorbers were used between the sample and the detector but no chemical separation was made to reduce the background.

Because of the large discrepancy in the reported values, we decided to measure[3] the cross section by employing chemical techniques to separate Sr from Y for enhancement of the detection sensitivity for 91 Sr.

Neutron capture in ^{90}Sr produces the 9.5 h 91 Sr which decays via ^{91}Y to the stable ^{91}Zr. De-excitation of the states in ^{91}Y produces intense gamma-rays at energies of 556, 750 and 1024 keV that are useful for quantitative measurement of91 Sr. The intensities of these gamma-rays are respectively 61.5’%. , 23.6% and 33%.

2 EXPERIMENTAL PROCEDURES

The experimental procedure was to irradiate purified ^{90}Sr with a measured flux of neutrons and then repurify to remove any miscellaneous activation products except for the desired ^{91}Sr. The 91 Sr in the sample was then measured with a calibrated Ge detector and the 9°Sr in the irradiated sample was determined by the β-counting of its ^{90}Y daughter.

2.1 Sample preparation and chemical processing

Approximately 4 μCi of 9°Sr used for preparation of various samples contained small traces of ^{137}CS and other long-lived or stable fission products. The details of the chemical processing are given in fig. 1. This illustrates the purification of the ^{90}Sr before and after irradiation with neutrons (Stages 1 and 2) and the later growth of the ^{90}Y daughter which is used for the ^{90}Sr determination (Stage 3).

The ion exchange columns used in fig. 1 are highly specific to strontium, retaining it in 3 M HN03 where most other cations, including yttrium are washed out. The strontium can be recovered by an ehrtion using water. The chemical recoveries or yields at stages prior to point “A” in fig 1 need not be quantitative as there is no separation of the ^{90}Sr and 91 Sr, upon whose ratio the cross sections depends. The ehrent from the second column is measured for ^{91}Sr quantitatively by a Ge gamma-ray spectrometer and then requires a measurement of the quantity of ^{90}Sr in the ehrent. But until this point quantitative recoveries are not necessary.

To quantify the measurements of the ^{90}Sr in the mixed strontium solutions from section 2 of fig. 1 after gamma-ray spectrometry for 91 Sr, a small amount of ^{88}Y tracer was added to the column just prior to the start of the ^{90}Y growth period. This ^{88}Y could be measured in the ^{90}Y beta source after decay of the ^{90}Y by means of gamma ray spectrometry. The amount of ^{88}Y used as a tracer was too small to affect the β-count of the ^{90}Y. This measurement, which gives the ^{90}Y recovery from the growth column, is the sole chemical yield required by the experiment.

2.2 Irradiation procedure

The purified ^{90}Sr recovered from the first stage of the chemical processing was dried into a thin quartz tube and irradiated in the NRU reactor at Chalk River in the Hydraulic Capsule Facility. The neutron flux at the location is 2 x 10^{14} n/(cm^2.s) with a Cd ratio of about 30. Three samples were irradiated separately and each was accompanied by a short wire flux monitor of O. 1% Cobalt in aluminum alloy (diameter 0.127 mm).

2.3 Gamma spectroscopy measurements

The ^{9}1 Sr activity in the ^{90}Sr / ^{9}1 Sr solution from column 2 in fig. 1 was measured with a Ge spectrometer. In order to suppress the bremsstrahlung from the β-decay of ^{90}Sr and its ingrowing ^{90}Y daughter a series of absorbers were introduced between the source and the Ge detector.

For calibration of the Ge spectrometer with the absorbers in place a series of absolute standard solutions were obtained from the Chalk River Standards Group[5]. A set of dilutions of these standards was made, each having the same volume as that of the ^{90}Sr / ^{9}1 Sr samples prepared following the irradiations. This set of calibration standards, which contained ^{54}Mn, ^{60}Co, ^{88}Y, ^{133}Ba, and ^{137}Cs was used to calibrate the Ge spectrometer.

2.4 Beta counter measurements

Two duplicate flow -type gas proportional counters with β-end windows were employed to measure ^{90}Sr by means of its ^{90}Y daughter. These counters were calibrated with an aliquot of a standard ^{90}Sr solution that was processed through the column in the same manner as the irradiated samples. After a standard growth period (72 h) aliquots of the newly grown Y were deposited on duplicate “weightless” sources and measured by each of the two counters. After the measurement of the standard ^{90}Y, the β-sources were measured with the Ge gamma-ray spectrometer to obtain the intensity of the small amount of ^{88}Y tracer that had been added prior to the growth period. From this the recovery of the Y from the column was found to be in the range of 97- 100%. The efficiency of the β-counters for detection of ^{90}Y β-rays was determined to be 3.4 ±0.5%.

3 RESULTS

3.1 Gamma-ray spectra

Fig. 2 shows the gamma-ray spectrum from one of the samples. The spectrum is clean with the 91 Sr decay peaks of the daughter ^{9}1Y well isolated and very good peak height to the background ratio illustrated by the log-linear plot in fig. 2. The majority of the background continuum below 530 keV is from ^{90}Sr beta-ray induced bremsstrahlung. The small amount of the continuum in the high energy part of the spectrum is produced by the bremsstrahlung from the build-up of ^{90}Y during the gamma-counting process. The three prominent gamma-rays from ^{9}1 Sr are clearly visible. The weaker background lines from ^{60}Co and ^{137}Cs are due to an earlier contamination of the shielding around the Ge spectrometer.

In fig. 2 the most prominent gamma-ray is the 389 keV transition from the 2.8 h half-life isomer ^{87m}Sr. This may have been produced by neutron capture in ^{86}Sr introduced in the ^{90}Sr stock by an admixture of natural Sr. An alternative possibility is that the ^{87m}Sr isomer is generated by inelastic scattering of gamma-rays and neutrons from the ^{87}Sr that is a fission by-product and would be present in all Sr chemically separated from spent fuel.

The yield of the three major gamma-rays of91 Sr was calculated from the counts accumulated in the 24 h measurement period following irradiation. The necessary corrections for decay during the measurement period and for decay between the end of the irradiation and the start of the 24 h measurements were made. The absolute activity of the ^{9}1 Sr was calculated using the literature values[4] for the yield of each of the three gamma-rays.

3.2 Analysis of the data

The cross section was evaluated individually for the activity determined for each of the three intense gamma-ray transitions in 91 Sr. The results presented in table 1 show good inter-sample agreement. The intra-sample agreement is similar. The difference between the absolute activities calculated from each of the gamma-ray energies appear to be somewhat systematic and results presumably from inconsistencies in the literature data for the gamma-ray yields, and uncertainties in the gamma-ray efficiency data.

Table 1
Cross section (rob) for the 91Sr(n,y)91 Sr reaction

Run	555 keV	749 keV	1024 keV	Internal average
1	9.9	11.1	10.0	10.3
2	8.8	9.8	9.3	9.3
3	8.8	10.0	9.5	9.4
Average		10.3	9.6	9.7
Over all average	= (;:;* 0.7) mb			

3.2 Uncertainties

The uncertainty quoted in the present work is the r.m.s. deviation obtained by calculating the cross section from each of three gamma-rays in each of three samples. The expected accuracy can be established by combining estimates of the various sources of possible errors. The principle sources of uncertainty are[3]:

0.5%	weighing
1.0%	volumetric
2.0%	γ-spectrometry of 91 Sr
2.0%	y-standards
2.2%	β counting
6.9%	y-branching ratios

Combining the squares of the standard deviations we would anticipate an overall statistical and systematic uncertainty of about 7.9°/0. This is reasonably similar to the experimental spread of 70/o obtained from the separate measurements.

A systematic error in the measurements would have been produced had the ^{90}Sr sample contained traces of fissile ^{235}U material. Irradiation of such a sample would produce extra 91 Sr due to the fission process. If this were attributed to the yield from the 90Sr(n,y)91 Sr the cross section would appear to be higher. No traces of fissile material were present in our samples otherwise we would have detected the 2.7 h half-life ^{92}Sr produce in the fission process.

4 SUMMARY

The thermal neutron radiative capture cross section of ^{90}Sr has been measured to be 9.7 ± 0.7 mb. This value overlaps at the two sigma level with the value of 14.0 ± 2.4 mb reported by McVey et al[2]. The low value of the cross section implies that the transmutation of ^{90}Sr in a thermal neutron flux would not be economical.

REFERENCES

1 G. ZEISEL, Acts Phvs. Austr. 23 (1966) 223.

2 L.A. McVEY, R.L. BRODZINSKI and T.M. TANNER, J.Radioanal.Chem. 76(1983) 131

3 M.A. LONE, W.J. EDWARDS and R. COLLINS, "Measurements of the thermal neutron cross section of $^{90}Sr(n,\gamma)$ reaction", Nuclear Instruments and Methods in Phsvics Resaerch, in press (1993)

4 E. BROWNE AND R.B. FIRESTONE, "Table of Radioactive Isotopes," Eds. V.S. Shirley, Wilev, New York (1986)

5 R. MARTIN, Private communication, (1991)

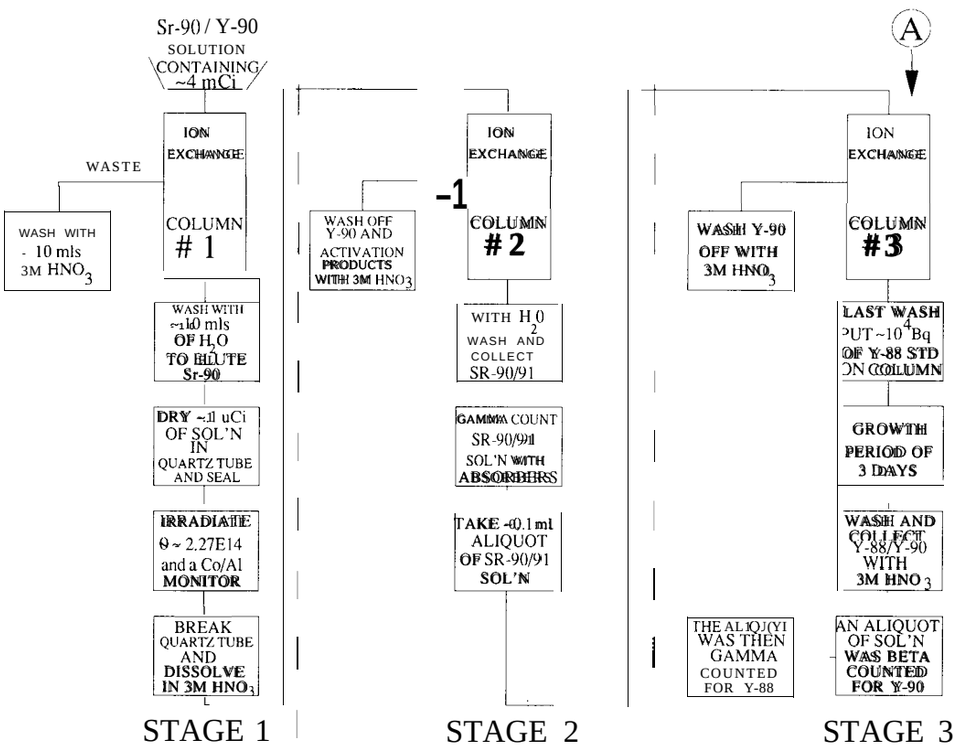

Fig 1 Flowsheet for chemical processing of the ^{90}Sr sample, before and after irradiation.

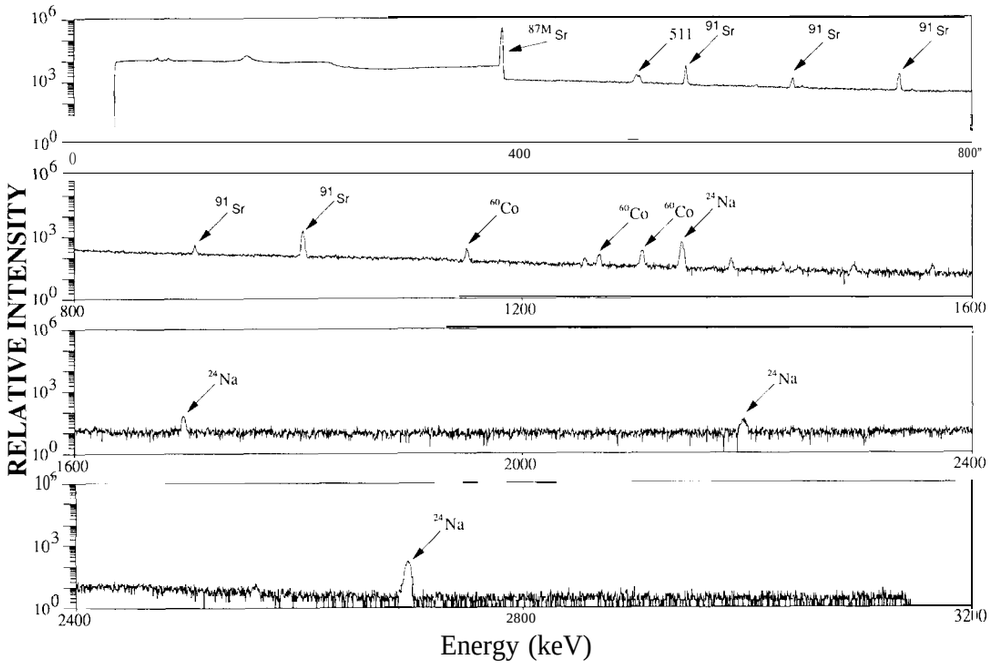

Fig.2 Gamma spectrum measured through absorbers showing transitions in ^{91}Y from the decay of ^{91}Sr. Shown also are background peaks arising from the decay of ^{60}Co, ^{24}Na and ^{87m}Sr.