NEUTRON EXPERIMENTS WITH A SENSITIVE SZILARD–CHALMERS DETECTOR

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Summary

The Szilard-Chalmers reaction has been developed as a sensitive detector for the comparison of neutrons from sources of different primary energy and angular distribution. Using sodium permanganate solution, a detection efficiency of approximately 7 per cent. for all neutrons in the energy range 0–5 MeV was obtained and high accuracy achieved.

The method of detection was employed to make the following measurements: (i) The neutron yield from the spontaneous fission of natural uranium was found to be $(1\cdot56\pm0\cdot12)\times10^{-2}$ neutrons $sec^{-1}\,g^{-1}$. (ii) The cross section for the reaction $^{6}Li(\gamma, n)$ at 6·2 MeV was found to be $(3\pm2)\times10^{-28}$ cm^2. (iii) The cross section for the reaction $^{9}Be(n, 2n)$ induced by Ra-Be and Po-Be sources of neutrons was found to be $(0\cdot37\pm0\cdot06)\times10^{-24}$ cm^2. (iv) A limit was placed on the cross-over transition from the 2·5 MeV state to the ground state in ^{60}Co. The transition probability was found to be less than 1×10^{-6} of that for the cascade consisting of the two γ-rays.

I. INTRODUCTION

The Szilard-Chalmers reaction has been developed in the direction of the use of volumes as large as 300 gallons of aqueous solution, in order to extend the limit of detection of neutrons and allow the comparison of weak neutron sources (Edge 1956). In the case of the largest detector employed, less than 3 per cent. of neutrons having an energy of 10 MeV failed to be moderated and captured. As is well known, the Szilard-Chalmers reaction (1934) involves the decomposition of the molecule in which it occurs, as a result of the recoil following the emission of the prompt γ-rays emitted after neutron capture. The active component is thrown into another state of chemical combination which is capable of extraction from the remainder. The permanganate complex proved most suitable for the work, the activity being precipitated in the form of $^{56}MnO_2$ (Broda 1948).

When intense neutron sources were being measured, sufficient activity was induced in a saturated potassium permanganate solution, but for weak sources a sodium permanganate solution of 670 g/l concentration proved more sensitive.

II. EXPERIMENTAL TECHNIQUE

A standard technique for processing the solution was developed in order to obtain reproducible results.

A proportion of the activity, called the "retentivity", was never precipitated from solution. This proportion had to be kept constant, the two

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factors causing variation being temperature and pH. By maintaining the temperature to within ± 2 °C, and holding the pH at $(7 \pm 0 \cdot 2)$ the variation in retentivity was found to be less than 2 per cent. In general, the pH of the solution remained constant and a buffer proved unnecessary, but it was necessary to keep the solution free of organic matter and to store it in darkness to prevent spontaneous precipitation of manganese dioxide.

Various methods of extracting the activity were investigated, such as centrifuging, electrostatic precipitation, filtration through asbestos wool, sintered glass, or filter paper. Only the last provided the reproducibility required, the most suitable paper tested being Whatman 540. It was found to be unnecessary to add a carrier to the solution to bring down the manganese dioxide ; however, a thin layer (1 g) of pure manganese dioxide powder spread evenly over the filter paper was required. To prevent the precipitate creeping round the edges of the paper in the Büchner funnel, a " Gaco " ring seal was pressed against the circumference of the filter paper by a glass chimney. Using such an arrangement it was found possible to vacuum filter 50 l. of solution in approximately 40 min.

After filtration the container was washed with distilled water and the washings themselves were filtered and collected separately ; less than 0·1 per cent. of the activity was left on the walls.

Before counting the precipitate was washed with distilled water followed by acetone and ether, to dry the paper. Counting was effected by placing the precipitate on the filter paper between two " Cellophane " sheets and wrapping this " sandwich " round a cylindrical thin-walled Geiger counter 3 cm in diameter and 10 cm long, with a wall thickness of only 0·007 in. With appropriate care it was found that the sandwich could be moved and then replaced accurately enough to give a counting rate reproducible to 2 per cent. The counter was fitted into a lead block to increase the backscattering of electrons and provide shielding against cosmic rays. Experiments on the self-absorption of the manganese dioxide indicated that slight variations in thickness of deposit over the area of the filter paper produced a negligible variation in the counting rate, provided the same quantity of manganese dioxide deposit was used each time.

III. Discussion of the Method of Detection

Four factors limited the usefulness of this detecting technique as a means of comparing weak neutron sources. These were the speed and efficiency with which the solution could be processed and the activity counted, the proportion of incident neutrons captured by manganese, as opposed to those that escaped or were captured by hydrogen, the long period activity present in the solution, and finally the cosmic ray background.

To facilitate rapid filtration a large filter paper, 9 cm in diameter, was used and the processing was reduced to the minimum number of steps compatible with maximum accuracy. It was found that, using a 50 l. tank, 90 min were required between the end of an irradiation and the commencement of counting.

Approximately 50 per cent. of the neutrons captured in the sodium permanganate solution were absorbed by ^{55}Mn and it was difficult to increase the

fraction captured by manganese above this value as the solution became too concentrated.

After the decay of the 2·6 hr half-life (Hollander, Perlman, and Seaborg 1953) activity from ^{56}Mn, a considerable proportion of long period activity still remained (Dodson, Goldblatt, and Sullivan 1944) and this did not decay appreciably over the period of a year. In the case of potassium permanganate, it was thought to be caused by ^{40}K adsorbed on the filter paper; the much smaller effect in the case of sodium permanganate suggested that this may have been due to potassium impurities in the compound. In the experiments this long period activity was subtracted from the total activity in order to obtain the ^{56}Mn activity.

Although experiments were made intermittently over a period of 2 years, no significant temporal variation in cosmic ray neutron flux was detected. The mean of 10 determinations of the cosmic ray neutron flux at sea-level yielded a value of $2{\cdot}5 \times 10^{-3}$ neutrons cm^{-2} sec^{-1}, a value in reasonable agreement with other determinations (Kaplan and Yagoda 1952). This background was also taken into account in the experimental work with the detector.

The efficiency of a neutron detector is generally stated to be the number of counts recorded per neutron expressed as a percentage. In the case of a Szilard-Chalmers detector, however, this definition is difficult to interpret, since the decay of activity during and after irradiation requires that the sensitivity is a function not only of the length and variation of intensity during the irradiation but also of the period between the end of the irradiation and commencement of counting—hence, although up to 50 per cent. of the neutrons emitted by the source may be captured by the manganese in the solution, only a small proportion of the resulting activity would be recorded by the Geiger counter, part being lost in the extraction process, part by the decay, and part owing to the efficiency of the counting arrangement itself. A suitable definition of the sensitivity, which has been used by Dodson, Goldblatt, and Sullivan (1944), is the ratio expressed as a percentage of the activity registered by the counter, corrected to the end of the irradiation (long compared with the ^{56}Mn half-life), to the rate of emission of neutrons by the source. Using this definition the detection efficiency of the 50 l. tank of potassium permanganate was found to be 1 per cent., and of sodium permanganate 7 per cent., for a $RaBeF_4$ neutron source placed at the centre of the tank. The method, therefore, is the most sensitive yet applied, and compares with detectors such as " long " BF_3 counters or phosphors (Segre 1953).

The detection efficiency is energy dependent, being a function of the number of neutrons escaping the detector. Theoretical investigations of the fraction escaping were made using approximations such as the Fermi " age " method (Wallace and LeCaine 1947) (curve *C* of Fig. 1) and the two-group diffusion theory of Tait (1954) (curve *B* of Fig. 1). An experimental method was used to determine the number of neutrons of known energy escaping from the 50 l. sphere, and the energy-escape curve calculated using two-group diffusion theory was normalized using it. The resulting curve (*A* of Fig. 1) is considered to be accurate to 5 per cent. over the energy range 0–6 MeV.

The number of neutrons escaping was measured by comparing the relative activity induced by two neutron sources of known strength in two tanks, one of 50 and the other of 1134 l. (300 gal). The two sources used were a ^{24}Na D(γ, n) source, yielding neutrons of energy 0·26 MeV, and a $RaBeF_4$ source which gave neutrons of average energy 4·5 MeV, with a spread extending from low energies up to approximately 13 MeV. It was found that 73 per cent. of the $RaBeF_4$ neutrons and 98 per cent. of the ^{24}Na neutrons were captured in the 50 l. tank, the remainder escaping. At least 99 per cent. of the $RaBeF_4$ neutrons were captured in the large tank.

In the following section four experiments carried out using the technique and only possible with a sensitive neutron detector are described.

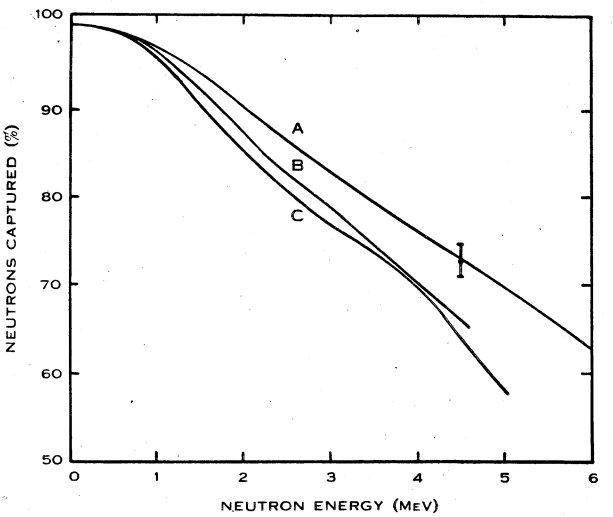

Fig. 1.—The variation of capture efficiency with energy for neutrons emitted from a source at the centre of a 50 l. sphere of potassium permanganate solution.

Curve *A*, the percentage of neutrons captured within the sphere, based on the two-group diffusion theory of Tait (1954) and normalized to pass through the experimental point shown, which was determined as described in the text; curve *B*, the percentage of neutrons captured according to the two-group diffusion theory of Tait (1954); curve *C*, the percentage of neutrons captured based on the diffusion theory given by Wallace and LeCaine (1947).

IV. Experimental Results

(*a*) *The Neutron Yield from the Spontaneous Fission of Natural Uranium*

Until 1952 the best value for the neutron yield from the spontaneous fission of natural uranium was that given by Segre (1952) as $(1{\cdot}5 \pm 0{\cdot}15) \times 10^{-2}$ neutrons sec^{-1} g^{-1} uranium. Littler (1952), however, has employed a subcritical pile to obtain $(1{\cdot}65 \pm 0{\cdot}1) \times 10^{-2}$ neutrons sec^{-1} g^{-1} uranium. In the present experiment an accurate comparison was made of the number of neutrons emitted from a block of uranium of known size with a $RaBeF_4$ neutron source, recently calibrated by Bretscher *et al.* (1949) and Edge (1956).

The cylindrical block of natural uranium metal (9·572 kg) was enclosed in a cadmium shell in order to reduce slow neutron fission, and was sealed in a glass container. The standard $RaBeF_4$ neutron source was similarly encased and also sealed in glass. Each, in turn, was then submerged at the centre of the spherical 50 l. glass tank of sodium permanganate solution (670 g/l), for a known period of several hours. On removal of a source the active $^{56}MnO_2$ deposit formed by the Szilard-Chalmers reaction was filtered off and counted as described above.

A sequence of three runs, alternating the uranium block and the standard source, was performed.

After subtracting the cosmic ray background and correcting for the escape of neutrons from the moderating medium, the ratio of $^{56}MnO_2$ activities gave the ratio of the number of neutrons emitted by the two sources, and from this the spontaneous fission rate of natural uranium was found to be $(1\cdot56\pm0\cdot12)\times10^{-2}$ neutrons sec^{-1} g^{-1}.

(*b*) *The Cross Section for the Reaction* ^6Li (γ, n) *at* 6·2 *MeV*

A cylinder of lithium fluoride, 4 cm in diameter and 4 cm long, was irradiated by the γ-ray flux emitted from a thick calcium fluoride target, with which the cylinder was in contact, under bombardment by protons of 900 keV energy. The specially constructed target tube projected into the spherical 50 l. tank of sodium permanganate solution so that the source was located at the centre. The photoneutrons from the lithium which, from energy considerations, were the only possibilities, were captured in the tank and the active ^{56}Mn precipitated, extracted, and counted as before. For calibration the lithium was replaced by an identically cased cylinder containing heavy water, and in addition background runs were performed using a hollow cylinder. The cross section for D(γ, *n*) at 6·2 MeV was taken from the work of Barnes *et al.* (1952) and from it the cross section for $^6Li(\gamma, n)$ at the same energy was found to be $(3\pm2)\times10^{-28}$ cm^2. The rather large error in this case was due to the small isotopic abundance of ^6Li in natural lithium; separated ^6Li was not available in sufficient quantity for a better measurement. Titterton and Brinkley (1951) find the cross section has increased to $(5\pm2)\times10^{-28}$ cm^2 at 17·6 MeV.

(*c*) *Cross Section for the Reaction* ^9Be(n, 2n)

When beryllium is excited by incident neutrons of energy greater than 1·85 MeV the reaction $^9Be(n, 2n)^8Be$ is energetically possible and has been observed by Grace *et al.* (1951). The early investigations have produced conflicting results for the cross section (Edge 1956) and in order to try to resolve the discrepancies, three independent experimental methods have been employed.

In the first, neutrons produced by a Ra-Be source were used, and a comparison made of the neutrons scattered from beryllium with those from copper. The experiment was carried out as follows:

A collimated beam of neutrons was fired through a brass tube 4 cm in diameter which passed through the centre of a 10 l. spherical glass vessel filled with potassium permanganate solution. This was surrounded by a tank of

borax solution, which acted as a neutron shield. Samples of beryllium or copper, in the form of cylinders 4 cm in diameter, were placed in the centre of this arrangement, so that the detector absorbed the neutrons scattered by these samples. After each exposure the activity was extracted from the solution and counted as before. A series of experiments were carried out and the $(n, 2n)$ cross section in beryllium was determined from the ratio of the relative average activities induced in the two samples.

Taking the values for the total neutron cross sections for beryllium and copper as given in the tables of Hughes and Harvey (1955) and correcting for the escape of neutrons from the detector, for scattered neutrons leading to the $(n, 2n)$ reaction, and for the (γ, n) reaction in the beryllium sample due to γ-radiation from the radium in the Ra-Be source, the cross section for Ra-Be neutrons was found to be $(0 \cdot 40 \pm 0 \cdot 06) \times 10^{-24}$ cm^2.

A second experiment was performed also with Ra-Be neutrons, employing a similar experimental arrangement, except that the tube ran only to the centre of the detector sphere. The total neutron flux was given by the induced activity with the beryllium block absent, the neutrons being stopped by the permanganate solution beyond the centre of the sphere. The additional induced activity with the block inserted was taken to be due to $(n, 2n)$ neutrons. Similar corrections were applied as in the previous experiment and the value $\sigma_{^9\mathrm{Be}(n, 2n)} = (0 \cdot 386 \pm 0 \cdot 1) \times 10^{-24}$ cm^2 obtained.

For the third experiment a small Po-Be source was used, which had essentially the same neutron spectrum as the Ra-Be source. It was held between two beryllium cylinders, 4 cm in diameter and 4 cm long, and the whole was enclosed in a tight-fitting soft glass casing. The beryllium cylinders could be replaced by hollow cylinders of the same dimensions. This source, with and without the beryllium alternately, was placed for a known time at the centre of a spherical 50 l. glass vessel of potassium permanganate solution and the activities induced in the solution with and without the beryllium present determined. Errors and corrections were applied as before, except that in this case no correction for the (γ, n) reaction was necessary.

From the results the cross section for the $(n, 2n)$ reaction was found to be $\sigma_{^9\mathrm{Be}(n, 2n)} = (0 \cdot 354 \pm 0 \cdot 08) \times 10^{-24}$ cm^2. The agreement between the three measurements is good and a combination of the three results allows a most probable value to be stated as $\sigma_{^9\mathrm{Be}(n, 2n)} = (0 \cdot 37 \pm 0 \cdot 06) \times 10^{-24}$ cm^2. This cross section is much larger than is predicted by a theory based on the idea of an orbital neutron circulating around an ^{8}Be nucleus. Schlögl (1948) calculated a cross section of $0 \cdot 05 \times 10^{-24}$ cm^2 at an energy of 4·5 MeV using this model.

Experiments performed using neutrons from the D(d, n) reaction induced using a Cockcroft-Walton generator, gave some indication of a resonance in the neighbourhood of 2·7 MeV and this may be the same as that found by Stafford (1951) in the total cross section.

(*d*) *A Determination of the Cross-over Transition in* ^{60}Co

The γ-ray decay scheme of ^{60}Co is well known and consists of a transition from the 2·50 MeV state (spin and parity 4+) to the 1·33 MeV state (2+) and thence to the ground state (0+). Myers and Wattenberg (1949) were able to place an experimental upper limit of 2×10^{-6} on the ratio of the number of transitions between the 2·50 MeV state and the ground state, to the number going to the 1·33 MeV state. The theoretical value for this transition E_3/E_2, based on a single particle model, according to these authors is 10^{-5} and a discrepancy exists which it is of interest to investigate.

An experiment was, therefore, performed in which two copper cylinders each containing 50 cm^3 of heavy water were supported so as to subtend the maximum possible solid angle at a source consisting of 25 curies of ^{60}Co. The E_2 transition γ-rays from the cobalt lie below the threshold for neutron emission from deuterium, but the E_3 transition from 2·50 MeV to the ground state is above it. Any neutrons from this reaction, therefore, would be detected by a Szilard-Chalmers detector, employing sodium permanganate solution, which was placed in close proximity to the cylinders of heavy water. It was shown in a separate experiment that the detector was completely insensitive to any γ-rays from cobalt. After the experiment the sensitivity of the detector was measured using the standard $RaBeF_4$ neutron source and from the data the ratio of the number of transitions between the 2·50 MeV state and the ground state to the number of transitions to the 1·33 MeV state was determined as less than 1×10^{-6}.

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