bach.[3] Data were taken at ten different detector biases. Corrections were applied to the data for elastic energy loss, multiple scattering, finite detector size, variation of the beam energy and intensity with angle, finite-source detector spacing, and fission effects. The fission correction was determined by first calibrating the detection efficiency for fission neutrons at each detector bias by using the spontaneous fissions from Pu^{240}, and then measuring the induced fission rate by means of a detector bias set above the initial neutron energy but below the upper energy limit of the fission spectrum.

Table II summarizes the results of the measurements. Fission effects on U^{235} and U^{238} at 14 MeV were negligible.

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Search for Delayed-Neutron Emission in Br^{86} Decay*

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A search for delayed-neutron activity in the decay of 54-sec Br^{86} was carried out. No statistically significant activity was observed, but an upper limit for the neutron branch of ~0.25% was set. This limit corresponds to a maximum of ~10% for the possible contribution of Br^{86} to the 55-sec delayed-neutron period observed in the thermal neutron fission of U^{235} and ascribed to Br^{87}.

INTRODUCTION

THE discovery of[1] 54-sec Br^{86} presents several problems in the interpretation of observations on short-lived bromine isotopes in fission arising from the similarity in properties of Br^{86} and Br^{87}. Experiments are being carried out in this laboratory and elsewhere[2] in an attempt to characterize the decay of these nuclides sufficiently to distinguish their individual contributions to the fission product observations.

One of the prominent features of Br^{87} is its decay to an excited state of Kr^{87} above the binding energy of the fifty-first neutron in Kr^{87}, and the subsequent neutron emission from this state. This decay gives rise to the well-known 55-sec delayed-neutron period in nuclear fission. Although Br^{86} decays to the closed-shell nucleus Kr^{86} and branching to a state above the neutron binding energy (expected to be about 9.5 MeV) may not be very likely, it would be of importance to establish whether Br^{86} may be contributing to the 55-sec delayed-neutron period observed in fission. The present investigation was undertaken to establish the extent of any such contribution.

EXPERIMENTAL

The apparatus and bombardment procedure used for the previously described experiments on the discovery of[1] Br^{86} were utilized in the present investigation. About 40 cc (STP) of enriched Kr^{86} was bombarded for 40 sec with neutrons produced at the Argonne National Laboratory 60-in. cyclotron. Conditions for the bombardments were essentially identical to those for the previous Br^{86} experiments. Several runs were made to ensure that Br^{86} was produced. The decay curves observed were superposable on those of the previous experiment, and indicated the presence of Br^{86} in approximately the same intensity.

Neutrons were detected with a ring of nine BF_3 counters immersed in deuterated paraffin oil and surrounded with a shield of borated paraffin.[3] Samples were placed in the center of the ring for counting. Calibrations of the neutron counter assembly, carried out with a Cf^{252} spontaneous fission source and with the delayed neutrons from thermal fission of U^{235}, indicate an efficiency of 1.2%. In the latter case, U^{235} was irradiated in the pneumatic tube assembly ("rabbit" facility) of the Argonne CP-5 reactor in an amount calculated to give about the same disintegration rate of Br^{87} as that of the Br^{86} expected from the $Kr^{86}(n, p)$ reaction at the cyclotron (i. e., 10^5–10^6 beta disintegration/min). Counting of the delayed neutrons from fission was begun about 0.2 min after the end of irradiation. The decay curves obtained clearly indicate the 55-sec period and shorter composite periods of ~22 sec and ~7 sec which were not further resolved. Figure 1 represents a composite of 5 runs combined to obtain greater statistical accuracy. The extrapolated initial counting rate of the 55-sec period was about 700 counts/min. Within the errors of irradiation timing and uncertainty of the neutron flux, this value is consistent with expectations and indicates

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

[1] A. F. Stehney and E. P. Steinberg, Phys. Rev. **127**, 563 (1962).
[2] E. T. Williams and C. D. Coryell, American Nuclear Society Meeting, Boston, June, 1962 and (private communication).
[3] A. F. Stehney and G. J. Perlow, Phys. Rev. **113**, 1269 (1959).

that any delayed neutron emission from Br^{86} produced at the cyclotron in approximately this intensity should easily be observable.

The quartz bombardment tube was freed from the vacuum system by a remotely activated cutter after recovery of the Kr^{86}, pulled through about 40 ft of flexible tubing to a position outside of the cyclotron chamber wall, and inserted in a reproducible position inside the neutron counter assembly. Counting was begun about 30 sec after the end of bombardment, and the data were recorded on punched tape.

After about 6 min of neutron counting, the quartz tubes were removed from the neutron counter and rinsed with a hot solution containing 2 ml $1M\,(NH_2OH)_2\cdot H_2SO_4$, 2 ml H_2O, 1.5 ml concentrated HNO_3, and 5 mg Se "hold-back" carrier. Bromine carrier and $AgNO_3$ solution were then added to precipitate AgBr which was filtered and mounted for beta counting in an end-window, flow-type, proportional counter. The decay curves obtained were superposable on those obtained in the previous Br^{86} experiments.[1] Although the 54-sec Br^{86} period was essentially gone by the time the samples were counted (about 8 min after the end of bombardment), the data could be normalized to those of the previous experiments through the longer lived periods (mainly the 3.0-min Br^{85}) to obtain the initial Br^{86} activity.

Although this technique permitted relatively quick observation of the neutron activity, the high background of induced neutron activity in the quartz bombardment vessels (probably from 4.2-sec N^{17} and secondary neutrons produced in the deuterated paraffin shielding of the neutron counter from 7.4-sec N^{16} γ rays) made observation of the presence of a low-intensity 54-sec period difficult. Moreover, an uncertainty in relating the neutron and beta counts was introduced by the chemical isolation of bromine in an unknown radiochemical yield.

In another series of runs, bromine was chemically isolated from the quartz bombardment vessels as described above immediately after bombardment, and the AgBr was counted in the neutron counter within 1.5 min after the end of the bombardment. After about 6 min of neutron counting, the samples were transferred to the beta counter to obtain the normalized 54-sec Br^{86} abundance associated with each delayed neutron count.

Background data were obtained on blank samples carried through the typical bombardment and counting cycle.

RESULTS AND DISCUSSION

In both types of experiment (neutron counting before and after chemical separation of bromine) the neutron counting data from the Kr^{86} bombardments were essentially identical with the background, and no significant 54-sec neutron activity attributable to Br^{86} was observed.

A composite decay curve of the neutron activity observed from Kr^{86} bombardments before chemical isolation of the Br^{86} is shown in Fig. 2 along with the data

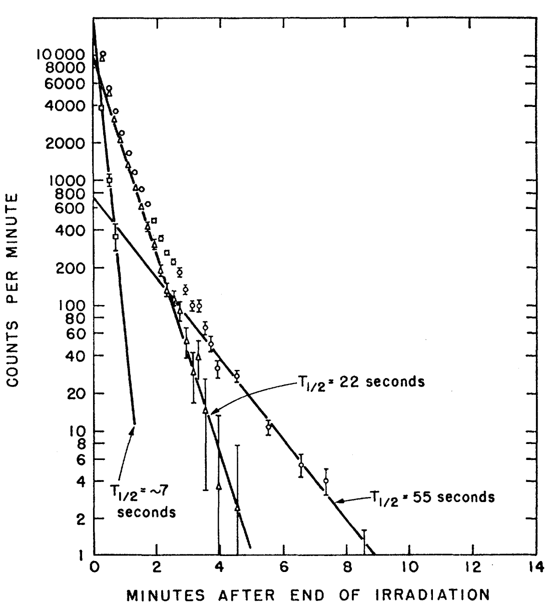

FIG. 1. Decay curve of delayed neutrons from thermal neutron fission of U^{235}. (○=original data with background of 2.4 ± 0.3 counts/min subtracted; △=55-sec component subtracted; □=22-sec component subtracted.)

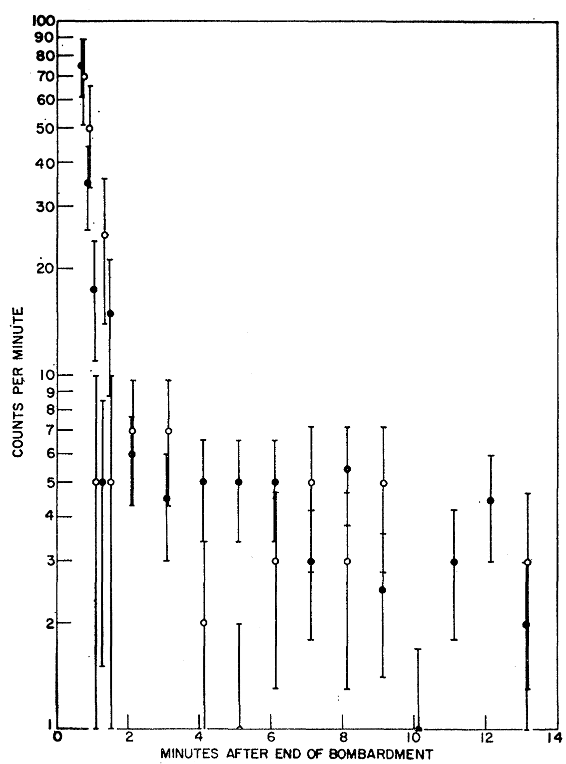

FIG. 2. Delayed neutron data from Br^{86} samples prior to chemical isolation compared with background blanks. (●=Br^{86}; ○=blank sample.)

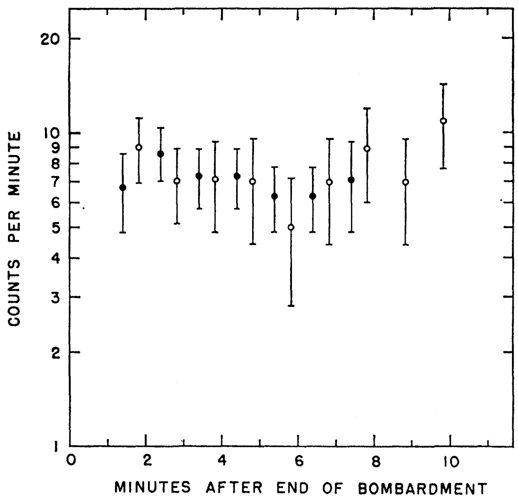

FIG. 3. Delayed neutron data from Br^{86} samples following chemical isolation compared with background blanks. (●$=Br^{86}$; ○=blank sample).

for blank background runs. Although such data are difficult to evaluate quantitatively, it is estimated that an initial intensity of about 10 counts/min of a 54-sec component may have been obscured in these decay curves.

The beta counting data on samples of AgBr isolated after 6 min of neutron counting indicated a production of about 2×10^5 disintegrations per minute of Br^{86}, uncorrected for chemical yield. If a reasonable correction is made for an estimated 50% loss in the yield of bromine activity in simply rinsing out the quartz tubes before chemical isolation, an upper limit for the neutron branching ratio of $(\lesssim 10 \text{ counts/min})/(1.2\times10^{-2})\ (4\times10^5)$, or 0.21% is obtained.

The composite data of 4 runs in which the neutron activity was observed following chemical isolation of the bromine are shown in Fig. 3. The background data (composite of 3 runs) are also shown. A least-squares analysis of these data gives a slope of -0.000164 ± 0.000154 for the Br^{86} samples and one of -0.000472 ± 0.000276 for the background data with an initial net neutron intensity (at the end of bombardment) of -0.023 ± 7.29 counts/min.

The beta counting data indicated an average production of $(1.95\pm0.42)\times10^5$ disintegrations per minute of Br^{86}, uncorrected for chemical yield.

The poor statistics associated with the neutron counting data result from the low level of activity, the short counting intervals, and the low counter efficiency. If the standard deviation is taken as a measure of an upper limit for the neutron counting rate (i.e., $\sim$7.0 counts/min) associated with a beta disintegration rate of $\sim2.0\times10^5$ dis/min, a value of 0.29% is obtained as an upper limit for the neutron branch. An average neutron branching ratio of $\lesssim$0.25% is obtained from the two types of experiment.

An upper limit to the possible contribution of Br^{86} to the 55-sec delayed-neutron period observed in fission may be calculated from the fission yield of Br^{86}, its neutron branching ratio, and the observed fission yield of the 55-sec delayed-neutron period. The latter value is 0.052% from the work of Keepin,[4] the fission yield of Br^{86} is estimated as 2.0%, using the observed yield[5] at Kr^{86} (2.02%) and the calculated independent yields of the chain members beyond Br^{86} (0.02%), and the neutron branching ratio is taken as $\lesssim$0.25% from this experiment. The fractional contribution of Br^{86} to the 55-sec delayed-neutron period is, then, given by

$$(2\times10^{-2})(\lesssim 0.25\times10^{-2})/(5.2\times10^{-4})\lesssim 0.1.$$

Thus, although no positive evidence for delayed neutron emission accompanying Br^{86} decay was obtained in the present work, an upper limit of about 10% may be set for the possible contribution of Br^{86} to the observed 55-sec delayed-neutron period in U^{235} fission, and the interpretation already given to the delayed-neutron data need not be significantly modified.

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[4] G. R. Keepin, T. F. Wimett, and R. K. Zeigler, J. Nucl. Energy **6**, 1 (1957); Phys. Rev. **107**, 1044 (1957).

[5] See, for example, S. Katcoff, Nucleonics **18**, No. 11, 201 (1960).