Neutron Activation Cross Sections for Br^{79}, Br^{81}, Rh^{103}, In^{115}, I^{127}, and Ta^{181}†

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(Received 26 July 1963)

Neutron activation cross sections for Br^{79}, Br^{81}, Rh^{103}, In^{115}, I^{127}, and Ta^{181} have been measured in the incident neutron energy range from 130 to 1800 keV. The results agree rather well with other activation measurements. The activation cross sections are combined to form the total neutron-capture cross sections of the natural elements and compared with the capture gamma-ray data of Diven *et al.* and Gibbons *et al.* The elemental cross sections agree rather well with the results of Diven *et al.* but do not agree with the lower results of Gibbons *et al.*

INTRODUCTION

THE two most widely used methods for the determination of the energy dependence and absolute value of neutron-capture cross sections are neutron activation measurements[1–14] and capture gamma-ray measurements.[15,16] Two semi-independent techniques have been utilized in neutron activation measurements. One technique consists of a direct measurement of the induced activity by either absolute gamma-ray or beta-ray counting.[1,2,6,14] The other technique consists of a measurement of the activity induced by fast neutron capture relative to the activity induced by thermal neutron capture.[10] The uncertainties introduced into the cross-section measurement in the absolute counting technique derive principally from uncertainties in the determination of the fast neutron flux and uncertainties in the detection efficiency of the absolute gamma- or beta-ray counters. The second technique, that of comparison between fast neutron and thermal neutron induced activity, is independent of absolute gamma-ray or beta-ray detection efficiency. However, this technique does depend on fast neutron flux measurements in the sense that the ratio of the fast neutron and thermal neutron efficiency of the flux monitor must be known. In addition, the thermal neutron activation cross section or the thermal neutron absorption cross section of the nuclide being studied must be known.

The method utilizing measurement of the prompt gamma rays emitted following neutron capture also depends on fast neutron flux measurements or on spherical shell measurements.[7–17] The gamma-ray detector is usually a large liquid scintillator used in conjunction with a pulsed accelerator. The largest uncertainty in this method is probably either the determination of the gamma-ray detection efficiency of the liquid scintillator or some systematic error in the spherical shell measurements. In short, all of the above-mentioned methods depend on either the direct measurement of fast neutron flux, measurement of the ratio of fast to thermal neutron flux, or measurement relative to a previously determined capture cross section.

In the two activation methods, differences should arise mainly from errors in the efficiency of the gamma- or beta-ray detector and errors in the values for the thermal activation cross sections. The thermal activation cross sections, of course, ultimately depend on absolute gamma- or beta-ray counting, except in those cases where there exists thermal neutron absorption cross section measurements which are independent of either neutron flux measurements or absolute gamma-ray or beta counting efficiencies.[18]

Differences in the large liquid scintillator measurements could arise from errors in the gamma-ray efficiency of the scintillator which is sensitive, to some degree, to the spectral distribution of the prompt gamma rays following neutron capture.[15,16,19,20] Rather large

† Work performed under the auspices of the U. S. Atomic Energy Commission.

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differences exist between the liquid scintillator measurements of Diven *et al.*,[15] and those of Gibbons *et al.*[16] Since the liquid scintillator measurements determine the total neutron-capture cross section for any element studied, the results can be compared with activation measurements only when activation cross-section determinations of all of the independent metastable and ground-state decay branches of a given element are available.

In this paper, activation measurements are presented for bromine, rhodium, indium, iodine, and tantalum. The individual activation cross sections are compared with other activation measurements, where they exist. The activation measurements are then combined to form the total neutron absorption cross section, and this total is compared with the liquid scintillator measurements.

EXPERIMENTAL METHOD

The experimental arrangement used for the fast neutron irradiations is shown in Fig. 1. Protons from a Van de Graaff accelerator were bent 25 deg by a steering magnet and struck a vacuum deposited lithium target. The Li(p,n) reaction was used for all irradiations. Samples of an element were prepared for irradiation by forming them into thin disks. The disks had a diameter of 2.54 cm and were between 0.25 mm and 1 mm thick with the exception of the rhodium foils used for absolute beta counting which were 2.5 to 7.5 μ thick. Each sample was positioned 3 cm from the lithium target and centered at zero degrees relative to the incident proton direction. Lithium targets were between 40 and 50 keV thick, measured at the Li(p,n) threshold. The combined effect of the neutron energy spread in the target and the energy change with neutron emission angle yielded a total energy spread for a 40-keV target which varied from approximately 43 keV for the 150-keV irradiation to approximately 62 keV for the 1800-keV irradiations.

A parallel plate fission chamber was used to monitor the neutron flux. For all fast and thermal irradiation, the samples were placed next to the 75-μ thick platinum disk which contained the U^{235} fission source. All fast fission cross sections used were taken from the compilation of Allen and Henkel.[21] Since the fissionable deposit was nearly the same size as the samples, this arrangement insured that both the samples and the fission source were exposed to essentially the same average neutron flux. Corrections for sample transmission were applied where necessary. For the fast neutron irradiations, the samples were close enough to the neutron target so that thermal neutron contamination was negligible.

In some cases the sample decay times were short enough so that it was necessary to monitor the proton

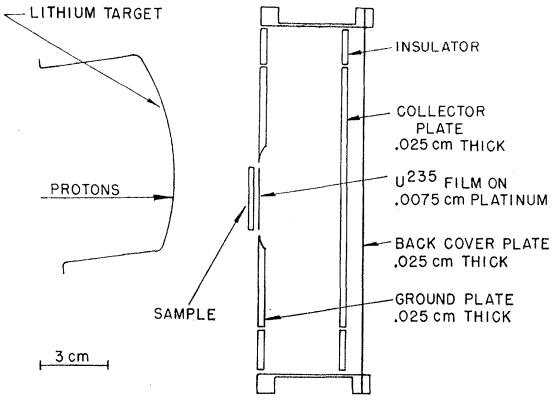

FIG. 1. Experimental arrangement of target, sample disk, and 2π fission chamber neutron flux monitor for neutron irradiations from 150 to 1800 keV.

current with a leaky current integrator.[22] For these cases, the decay constant of the leaky current integrator was set equal to the literature value for the decay constant of the nuclide being irradiated.

For the thermal irradiation, a large block of paraffin, 15 cm thick, was placed between the target and the sample-fission chamber combination. The degree of thermalization was inferred from cadmium ratio measurements. The cadmium ratio was $\sim$100 for all irradiations. Irradiations were made with and without cadmium covering. Sample activities and fission counts were corrected for any activity or fission counts due to epicadmium neutrons. Corrections were also experimentally determined for thermal neutron absorption in the samples, for the presence of the second neutron group from the Li(p,n) reaction,[23] and for average path length in the sample.[24] The fraction of fission counts due to epicadmium neutrons was $\sim$1%. The fraction of sample activities due to epicadmium neutrons varied between 5 and 17%.

The sample activity was counted with either a large NaI scintillation detector for gamma rays, a 0.25-mm

TABLE I. Cross sections used in thermal comparison method.

Target nuclide	Half life	$\sigma_{n,\gamma}$(thermal) barns
Br^{79}	4.5 h	2.9 ±17%
Br^{79}	18.0 min	8.5 ±16%
Br^{81}	36.0 h	3.1 ±16%
Rh^{103}	4.4 min	12.8 ±16%
Rh^{103}	42.0 sec	140.0 ±20%
In^{115}	54.0 min	155.0 ±6%
In^{115}	13.0 sec	52.0 ±11%
I^{127}	25.0 min	6.22 ±25%
Ta^{181}	16.4 min	0.030±33%
Ta^{181}	115.0 day	21.0 ±5%
Br^{79}	4.5 h +18 min	10.4 ±10%
Rh^{103}	4.4 min+42 sec	156.0 ±5%
In^{115}	54.0 min+13 sec	207.0 ±6%

[21] W. D. Allen and R. L. Henkel, *Progress in Nuclear Energy* (Pergamon Press, Inc., New York, 1958), Ser. I., Vol. II.

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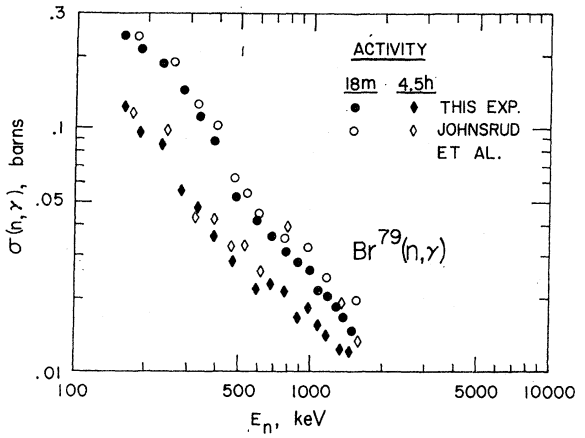

FIG. 2. Activation cross section results for Br^{79} including the data of Johnsrud *et al.* (Ref. 10).

thick plastic scintillator for beta rays, or a 4π gas-flow counter for absolute beta counting. After irradiation, the samples were transported from the target to the appropriate counter. The transfer time was short compared to the decay time of the activity involved. For all activities with decay half lives less than 5 min, the gamma or beta counters were recorded with a time scaling circuit in conjunction with a multichannel analyzer. In this way, the detailed decay curve could be analyzed for background contribution and the contribution of unwanted activities, and an accurate value for initial activity could be obtained.

EXPERIMENTAL RESULTS

Bromine

Samples for bromine irradiations were in the form of thin compressed disks 2.54 cm in diameter with a weight of approximately 3 g. The disks were fabricated from either NaBr or KBr. Irradiations were for 10 or 60 min duration. Immediately after irradiation, the samples were transported to a gamma counting system which contained two independent NaI detectors. Discrimination levels on one detector were set to count all gamma-ray pulses between 400 and 900 keV. A single discrimination level was set on the second detector to count all pulses greater than 660 keV. Each sample was counted five times during the decay period. Thus, the relative contribution of the three periods, with associated half lives of 18 min, 4.5 h, and 36 h, were extracted from the sample irradiations. The absolute normalization of the cross sections was determined from comparison with thermal neutron irradiation. The thermal cross sections used for normalization are given in Table I. The neutron activation cross sections associated with the 18-min and 4.5-h activities of Br^{80} are given in Fig. 2 together with the measurements of Johnsrud *et al.*[10] Figure 3 contains the neutron activation cross section associated with the 36-h activity of Br^{82} together with the measurements of Johnsrud *et al.*[10] Characteristic breaks in the capture cross section in the vicinity of 300 keV are evident in both figures. The breaks may be correlated with the onset of inelastic neutron scattering to the series of levels in Br^{79} starting at 208 keV and the 278 keV level in Br^{81}.[10,12] On the whole, the agreement between our results and those of Johnsrud *et al.*[10] is reasonably good except for the 4.5-h activation for E_n greater than 600 keV where their results are much higher than ours. In this connection, it should be mentioned that the isomer ratio formed from the results of this experiment has an energy dependence which is more reasonable than that which would be derived from

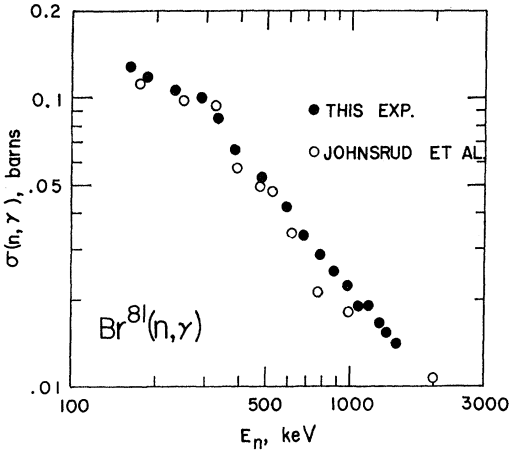

FIG. 3. Activation cross section results for Br^{81} including the data of Johnsrud *et al.* (Ref. 10).

the measurements of Johnsrud *et al.*[10] The cross section measured by Johnsrud *et al.* shows an abrupt increase between 600 and 800 keV. The theory of Huizenga and Vandenbosch[25] suggests a weaker energy dependence.

Rhodium

The rhodium measurements were made with a 2.54-cm-diam disk 0.025-cm thick, fabricated from metal. The beta activity was measured with a thin (0.025-cm thick) plastic scintillator. This arrangement was used for all energy dependence measurements and for all absolute normalization measurements utilizing the thermal neutron comparison method described above. For absolute normalization measurements utilizing absolute beta counting, very thin rhodium foils 2.5 μ to 7.5 μ thick were used. Immediately after irradiation, the foils were transferred to a 4π gas-flow counter of the Chalk River design. In both of the methods described above, the beta activity was recorded as a function of decay time with the aid of a multichannel time analyzer. The decay was followed for a sufficient time, so that both the 42-sec and the 4.4-min activities could be determined. The cross section as determined from the 4π beta counter

[25] J. R. Huizenga and R. Vandenbosch, Phys. Rev. **120**, 1305 (1960).

measurements and the cross section as determined from the thermal neutron comparison method agreed within 3%. The capture cross sections associated with the metastable state and ground state of Rh^{104} are given in Fig. 4. Again the break in the cross section near 300 keV may be correlated with inelastic neutron scattering to the 297 keV level in Rh^{103}. Cross section measurements of Hughes *et al.*[26] are included for comparison. Hughes measurements were made with fission neutrons with an effective energy of 1 MeV. Since Hughes cross sections are appropriate only for a fission neutron spectrum they cannot be compared directly with our monoenergetic neutron results. In order to compare Hughes data with ours, it was necessary to calculate the displacement of the capture cross section resulting from the experimental average of the capture cross section over the fission neutron spectrum.[27] The arrows in Fig. 4 indicate the position of Hughes points after correction for this displacement. The agreement between our data and that of Hughes *et al.* is good.

Indium

The indium measurements were made with either compressed disks of indium sulfate or metal foils 2.54 cm

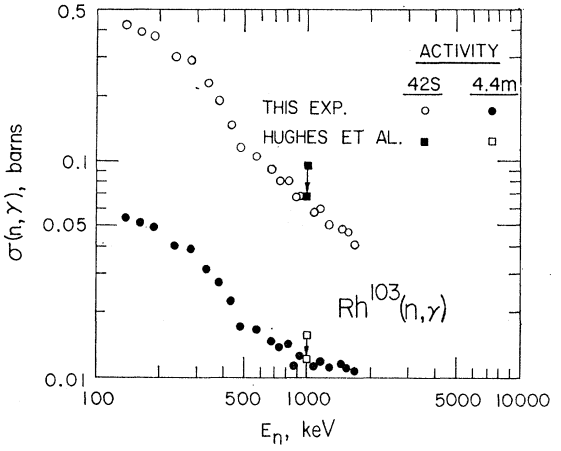

FIG. 4. Activation results for Rh^{103} including the data of Hughes *et al.* (Ref. 26).

in diameter. The indium sulfate disks weighed approximately 2 g and the foils weighed approximately 0.5 g. Measurements of the 13-sec and 54-min activities were made at different times. The 13-sec beta activity was counted with a thin plastic scintillator used as beta detector in conjunction with a multichannel time analyzer. Since the activity decayed quickly, it was necessary to invert the fission chamber and place the beta detector approximately 1 cm from the indium foil, with the fission chamber and the indium foil in position for irradiation. With this arrangement the counting of the beta activity was begun within 1 sec of the end of the irradiation. Tests were made to insure that the proximity of the beta detector to the indium foil and the fission chamber, during the irradiation, had no appreciable effect on the indium foil activity or the fission counting rate. The decay of the activity, for all runs, was recorded with a multichannel time analyzer, in increments of 1 sec, and followed for 400 sec. For the 54-min activity measurements, the gamma rays following the beta decay of the In^{116} 70-keV level were counted with a NaI scintillation detector. The results for both of the activation measurements are given in Fig. 5 together with the measurements of Johnsrud *et al.*[10] for the 54-min activity. For neutron energies below 900 keV, the measurements of the 54-min activation cross section of Johnsrud *et al.* are in good agreement with ours. Above 900 keV, the measurements of Johnsrud *et al.* are significantly higher than ours. Part, and perhaps most, of the difference may be accounted for by the presence of inelastic neutron scattering in In^{115} which directly populates the 4.5-h metastable state. The γ ray associated with the decay of the 4.5-h metastable state was present in all of the gamma-ray spectra associated with bombarding energies above 900 keV. For this reason, the gamma-ray bias in our measurements was set at 600 keV. Johnsrud *et al.*[10] state that their gamma-ray bias was set at 100 keV, thus, they would have included some of the gamma rays from inelastic neutron scattering in their measurements. Without detailed information concerning their experiment, it is difficult to estimate the degree to which the cross-section measurement is affected; however, the difference indicated in Fig. 5 is probably not unreasonable.[28,29]

Iodine

Iodine measurements were made with KI disks 2.54-cm diam and approximately 3 g in weight. Cross-

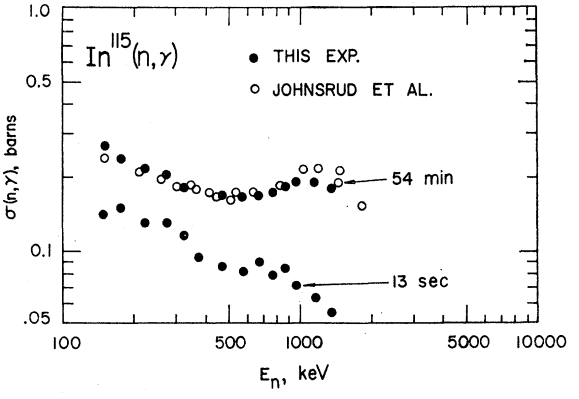

FIG. 5. Activation results for In^{115} including the data of Johnsrud *et al.* (Ref. 10).

[26] D. J. Hughes, R. C. Garth, and J. S. Levin, Phys. Rev. **91**, 1423 (1953).
[27] L. Cranberg, G. Frye, N. Nereson, and L. Rosen, Phys. Rev. **103**, 662 (1956).
[28] A. A. Ebel and Clark Goodman, Phys. Rev. **93**, 197 (1954).
[29] H. C. Martin, B C. Diven, and R. F. Taschek, Phys. Rev. **93**, 199 (1954).

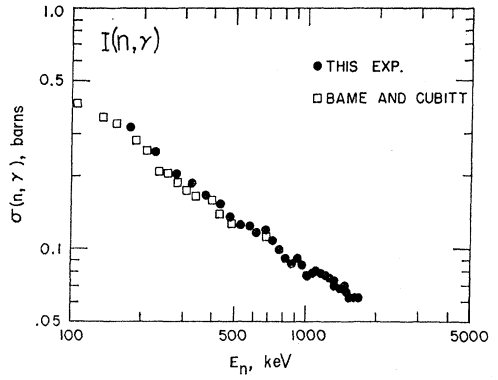

FIG. 6. Activation results for I^{127} including the data of Bame and Cubitt. (Ref. 9).

sections determinations were made by comparing fast and thermal neutron irradiations and by comparing the gamma activity from iodine and gold disks after simultaneous irradiations at 400, 600, and 1000 keV. Previous gold capture cross-sections measurements,[1] which were obtained from absolute gamma-ray counting, were used to obtain the absolute capture cross section for iodine. For the measurements which compared the iodine activity to the gold activity, the gamma spectrum was recorded with a multichannel pulse-height analyzer and the area under each photo peak was determined graphically. The NaI scintillator efficiency measurements of Lazar *et al.*[30] were used to obtain the small efficiency correction between the 412-keV gamma ray from gold and the 450-keV gamma ray from iodine. The fractional contribution of the 450-keV iodine gamma ray (17.2%) was taken from the measurements of Benczer *et al.*[31] The two methods for absolute normalization agreed within 5% with the thermal normalization giving the lower value. The capture cross section for iodine is given in Fig. 6 together with results of other investigators.

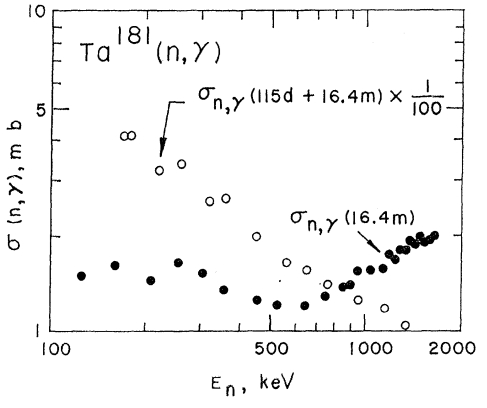

FIG. 7. Activation results for Ta^{181}.

[30] N. H. Lazar, R. C. Davis, and P. R. Bell, Nucleonics 14, No. 4, 52 (1956).
[31] N. Benczer, B. Farrelly, L. Koerts, and C. S. Wu, Phys. Rev. 101, 1027 (1956).

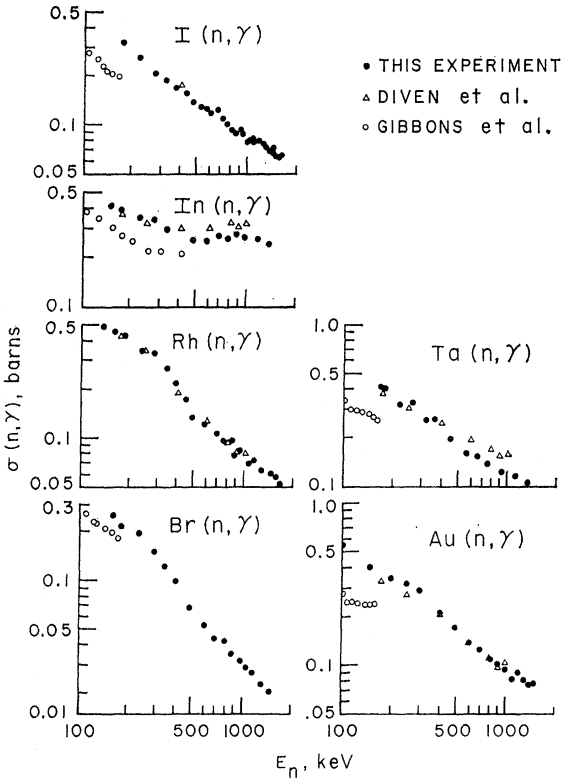

FIG. 8. Total neutron capture cross section for the natural elements compared with the data of Diven *et al.* (Ref. 15) and Gibbons *et al.* (Ref. 16). For indium, our measurements do not include neutron capture in the 4.3% abundant isotope In^{113}.

Tantalum

Samples for the tantalum irradiations were metal disks 2.54 cm in diameter and 0.051-cm thick. Measurements of the 115-day and 16.4-min activity were made at different times. In both cases, the gamma-ray activity was measured with a NaI scintillation detector. The absolute activation cross section was determined by comparison with thermal irradiations. The results are given in Fig. 7.

The most striking feature of the two curves is the different energy dependence. The isomer ratio changes by approximately a factor of five over the measured energy range. The change is probably indicative of the increasing importance of higher neutron orbital angular momenta in the capture reaction as the energy is raised.[25]

COMPARISON OF CAPTURE GAMMA-RAY MEASUREMENTS

Basically, activation measurements determine partial absorption cross sections. The measured cross section refers specifically to capture leading to the ground-state decay of the active nuclide or a metastable state decay of the same nucleus. At most, an activation cross section is the neutron absorption cross section of a single isotope. The measurements derived by the large liquid scintillator pulsed source technique have generally de-

termined the neutron capture cross section of the natural element. Thus, activation measurements and large liquid scintillator measurements can be compared directly only if the activation cross sections are combined to form the neutron absorption cross section of all of the naturally occurring isotopes of a given element. This has been done for the measurements reported here. The comparisons are direct except for indium for which the activation cross section of 4.3% abundant In^{113} is not available. The results are shown in Fig. 8 together with the large liquid scintillator results of Diven *et al.*[15] and Gibbons *et al.*[16] The absolute normalization of the measurements of Diven *et al.* is based ultimately on the U^{235} fission cross section as is the data here. Gibbons *et al.* derive their absolute normalization from the spherical shell transmission measurements of Schmitt and Cook[17] and from the measured shape of the $B^{10}(n,a,\gamma)$ cross section which is used to determine the energy-dependent shape of all of their cross sections.

It is evident from Fig. 8 that the results obtained here are in better agreement with the results of Diven *et al.*[15] than with the results of Gibbons *et al.*[16] Even in the case of indium, the difference which exists between the present results and those of Diven *et al.*[15] at higher energies may be explained by the previously mentioned fact that the activation measurements reported here do not include the cross section for capture in the 4.3% abundant In^{113}. The absolute counting measurements obtained here indicate that the cross sections derived from the thermal neutron comparison measurements do not contain any systematic errors related to the thermal irradiations. The disagreement between these results and those of Gibbons *et al.*[16] could arise from some unknown factor in the spherical shell transmission measurements, or from an inaccurate determination of the shape of the $B^{10}(n,a,\gamma)$ cross section which was used by Gibbons *et al.*[16] for the determination of the energy dependence of all their capture cross sections. It is unlikely that any errors in the U^{235} fission cross section could sufficiently change the results of either the activation measurements or the large liquid scintillator measurements to account for such a large difference.

DISCUSSION OF ERRORS

Errors in the experimental measurements reported here arise from uncertainties in a number of factors: flux attenuation, epithermal neutrons, and thermal neutron capture cross sections in the thermal neutron irradiations; single scattering corrections, the second group of neutrons from the $Li(p,n)$ reaction and the U^{235} fission cross section for all of the fast neutron measurements. The largest sources of error are the reported values for the thermal neutron capture cross sections and the U^{235} fission cross section. The uncertainty in the U^{235} fission cross section varies from $\sim 10\%$ for the lowest neutron energies used to $\sim 5\%$ for the highest neutron energies. Reported errors in the thermal neutron capture cross sections are given in Table I.

Uncertainties in the shape of the capture cross sections reported here are much smaller. Measurements in the vicinity of the sharp "breaks" in the capture cross section of bromine and rhodium were reproducible to $\sim 5\%$.

Finally the author apologizes for not including much of the previously reported capture work in the figures. Omissions were made entirely for clarity of presentation. Inclusion of all previous data would have made some of the figures unreadable.