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$Fe^{54}(n,p)$, (n,α), and $(n,2n)$ Cross Sections*

S. R. SALISBURY AND R. A. CHALMERS
Lockheed Missiles and Space Company, Palo Alto, California

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Absolute cross sections for the $Fe^{54}(n,p)Mn^{54}$, and $Fe^{54}(n,\alpha)Cr^{51}$ reactions have been measured for the neutron energy intervals 2.2 to 6.2 MeV and 13.1 to 16.8 MeV. The cross section for the $Fe^{54}(n,2n)Fe^{53}$ reaction has been measured at 14.0 and 16.8 MeV. Ten samples 1 by 2 by 0.45 cm enriched to 97% in Fe^{54} were irradiated. Neutrons were produced with the $D(d,n)$ and $T(d,n)$ reactions, using gas targets and a Van de Graaff accelerator. Samples were placed 9 cm from the gas cell at angles from 0° to 122°. The absolute neutron flux at 11° was measured with a proton-recoil telescope. Relative neutron angular distributions were obtained from published $D(d,n)$ and $T(d,n)$ cross sections by integrating over source and target dimensions; the effect of scattering in the gas-cell window foil was also included. These angular distributions were normalized to the telescope values. The induced gamma activity following the decay of Mn^{54}, Cr^{51}, and Fe^{53} was counted with a large calibrated NaI scintillation spectrometer. The (n,p), (n,α), and $(n,2n)$ results at 14 MeV agree with previous experimental work. Previous measurements of the (n,p) cross section from 2 to 6 MeV give values somewhat lower than the present work. All experimental results are somewhat higher than cross sections obtained by a statistical-model calculation based on optical-model potentials, while a more empirical calculation gives good agreement with the $(n,2n)$ data.

INTRODUCTION

KNOWLEDGE of the $Fe^{54}(n,p)Mn^{54}$ reaction cross section is of considerable importance because it permits the use of natural iron as an integrator of neutron flux for energies above 2 MeV. The 314-day half-life of the residual nucleus Mn^{54} permits integration periods of many months. The simple decay scheme facilitates use of the reaction, since Mn^{54} decays by electron capture solely to the first excited state of Cr^{54}. The observed radiation is the 835-keV de-excitation gamma ray.

Despite its practical significance, knowledge of the cross section of this reaction was sparse at the outset of this work.[1–5] It was determined that an activation measurement over a broad neutron energy range could best be made using relatively large samples of Fe^{54}. The experiment was performed with 50 g of 97% enriched Fe^{54} obtained on loan from the U. S. Atomic Energy Commission.

Measurements of the $Fe^{54}(n,\alpha)$ and $Fe^{54}(n,2n)$ reaction cross sections were also made by counting the gamma rays from the decay of Cr^{51} and Fe^{53}. Previous measurements[5–10] of the (n,α) cross section exist only for energies from 13 to 15 MeV although the Q value of the reaction is positive. Previous measurements of the $(n,2n)$ cross section exist only within 1 MeV of threshold[4,5,7,8,10] and are insufficient to permit extrapolation to higher energies.

Fe^{54} samples were irradiated with both d-D and d-T neutrons to cover neutron energy intervals from 2.2 to 6.2 MeV, and from 13.1 to 16.7 MeV. Absolute values of the neutron flux were obtained with a proton-recoil telescope. Several samples were exposed simultaneously at various angles to eliminate telescope uncertainties as a source of relative errors. The long Mn^{54} half-life with the resulting low activity and the relatively low neutron fluxes at angles away from zero led to the choice of pairs of 5-g samples at each angle. All samples were the same size to eliminate relative errors in gamma-ray counting efficiencies.

EXPERIMENTAL PROCEDURE

1. Irradiation

Three irradiations of the Fe^{54} samples were made. In the first, all five pairs were arranged in a circle 9 cm from the neutron source as shown in Fig. 1 at mean angles of 0°, 38°, 63°, 88°, and 122°. The deuterium gas target was bombarded with 2.93-MeV deuterons. Neutrons emitted at the above angles had energies of 6.18, 5.38, 4.26, 3.22, and 2.23 MeV. In the second irradiation a pair of samples was placed at the 0° position. A deuteron energy of 1.00 MeV was used to produce neutrons of 4.69 MeV. In the third irradiation three pairs of samples at 0°, 100°, and 130° were irradiated with neutrons of 16.75, 14.05, and 13.10 MeV obtained from the bombardment of tritium gas by 1.49-MeV deuterons.

To ensure freedom from effects of thermal neutrons, the 1- by 2- by 0.45-cm thick iron samples were surrounded by a layer of 0.010-in. cadmium foil. As a check that the effect of stray high-energy neutrons was negligible, several additional exposures of indium samples

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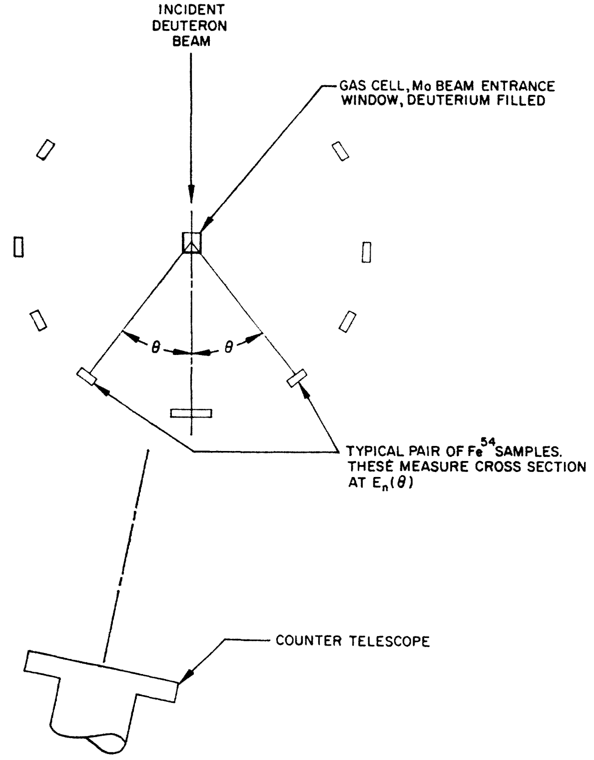

FIG. 1. Schematic view of apparatus for the first irradiation.

were made which demonstrated that the neutron flux obeyed the inverse-square law when measured from the gas target cell.

The deuterium gas target was 9 mm long with a 3.556 mg/cm^2 Mo window and was filled to 2 atm pressure. Neutron-flux calculations using the number of incident deuterons and the target thickness with an experimentally determined 30% correction for beam heating yielded reasonable agreement with the flux measured by the proton-recoil telescope. The tritium target was 3 cm long and filled to 0.9 atm. Beam currents of about 3 μA were used.

The proton-recoil telescope was a duplicate of one built at Los Alamos that provides coincident signals from two proportional counters and a CsI(Tl) scintillation detector.[11] The output of the scintillator was pulse-height analyzed, gating on the output of the coincidence circuit. Satisfactory operation of the telescope was determined by a variety of tests varying such parameters as thickness of radiator foil used, distance from gas cell, and angle of incidence. A count made with no radiator in the telescope yielded a 4% background count rate for which subsequent correction was made. A typical pulse-height spectrum is shown in Fig. 2.

For the first D(d,n) irradiation the relative angular distribution of the neutrons produced has been obtained by folding the published D(d,n) cross sections[12] into the multiple-scattering distribution of the incident beam in the entrance foil. The mean angle of this latter distribution was 3°. Numerical integrations were then performed over the cell length and sample size. The resulting relative angular distribution was then normalized to the telescope value.

The second D(d,n) irradiation was treated in the same way. A similar technique was used for the T(d,n) reaction, except that since the angular distribution[12] was slowly varying, the multiple-scattering contribution was not used.

The effect of elastic scattering in the Fe is to slightly increase the path length for neutrons scattered in the center region of the sample, while decreasing it for those scattered near the edges so as to escape through the sides. To calculate the effect, cross sections and angular distributions for elastic scattering in Fe^{54} must be used. These results are not in general available, but estimates of the numbers were gotten from other nuclei in the same mass region. The calculation using these estimates showed the net correction for elastic scattering to be less than 1%. No correction was made to the flux; the effect was reflected in an added uncertainty.

Inelastic scattering will decrease the neutron energy by at least 1.4 MeV, and the average inelastically scattered neutron is expected to lose over half its energy. The decrease in the (n,p) cross section at this lower neutron energy will tend to compensate for the increased path length due to scattering. In addition, the number of inelastic scatterings should be less than the elastic. Again no correction was made but allowance for the effect was made in the uncertainty.

As a test of the entire procedure, exposures were made of sandwiches of indium, aluminum, and natural iron foils and cross sections were measured. The activity observed in the indium was the 4.5-h 335-keV isomeric transition resulting from inelastic scattering. Aluminum

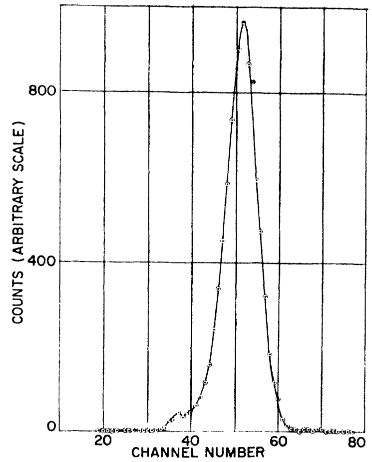

FIG. 2. Pulse-height spectrum of the recoil-proton telescope.

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[12] J. B. Marion and J. L. Fowler, *Fast Neutron Physics* (Interscience Publishers, Inc., New York, 1960), pp. 80, 81, 84.

TABLE I. Decay modes, half-lives, and gamma-ray energies.

Reaction	Mode	Half-life (Nuclear Data Sheets)[a]	Present work	Measured gamma ray Emitter	Energy (keV)
$Fe^{54}(n,p)Mn^{54}$	ϵ	314 day	315 day	Cr^{54}	835
$Fe^{54}(n,\alpha)Cr^{51}$	ϵ	27.8 day	27.5 day	V^{51}	320
$Fe^{54}(n,2n)Fe^{53}$	β^+	8.9 min	8.65 min	annihilation radiation	511
$Fe^{56}(n,p)Mn^{56}$	β^-	2.58 h	2.57 h	Fe^{56}	845
$Al^{27}(n,p)Mg^{27}$	β^-	9.5 min	9.56 min	Al^{27}	834 1015
$In^{115}(n,n')In^{115m}$	I.T.	4.5 h	4.48 h	In^{115}	335

[a] Reference 17.

and iron (n,p) reactions led to 9.5-min Mg^{27} and 2.58-h Mn^{56} activities, respectively. The measured cross section for the $Fe^{56}(n,p)$ reaction at 6 MeV was in excellent agreement with the value of Terrell and Holm.[13] The concurrently irradiated Al foil, however, led to a value 33% lower than that of Los Alamos[14] while the $In^{115}(n,n')In^{115m}$ cross section was higher than would be implied by the trend of the data of Martin *et al.*[15] Another exposure at 4.7 MeV, however, gave excellent agreement for the $Al(n,p)$ result while the $In^{115}(n,n')In^{115m}$ result was 30% high. A similar test of the $Fe^{56}(n,p)Mn^{56}$ reaction at 16.75 MeV gave a value within experimental uncertainty of that of Terrell and Holm.[13]

2. Counting

Induced activities were all measured with 4-in. by 4-in. NaI detectors; counting was performed on a TMC 400-channel analyzer. The absolute efficiencies of the scintillation detectors were determined with a variety of calibrated point sources. These were obtained in part from the National Bureau of Standards and in part from 4π beta calibration of sources of our own manufacture. Good agreement was obtained from the two types of sources. The 314-day half-life of the Mn^{54} allowed counting to be postponed for a day after the irradiation to permit decay of the 845-keV gamma ray from $Fe^{56}(n,p)Mn^{56}$ which had a 2.58-h half-life.

The corrections for self absorption of the gamma rays by the Fe^{54} samples were fairly large for all three activities observed. In the case of the Mn^{54} activity it was experimentally obtained by constructing a similar sized block of natural iron with a known amount of Mn^{54} distributed evenly through it. This was done by mixing a known volume of liquid containing the Mn^{54} with a few grams of iron powder and a binder, and molding it into the shape of the Fe^{54} samples. A second known volume of the same Mn^{54} solution was made into a point source for calibration. The ratio of the counts of these two Mn^{54} sources then gave the required correction. An additional correction was made to compensate for the difference in density between the Fe^{54} and the artificial samples which were 5.5 g/cm^3 and 3.2 g/cm^3, respectively.

The 320-keV gamma ray of Cr^{51} is not much higher in energy than the backscatter peak of the 835-keV Mn^{54} gamma ray. Its intensity varied from scarcely observable to comparable to the backscatter peak. The TMC analyzer was equipped with an accessory for subtracting one stored spectrum from another and it was decided to use this to remove the Mn^{54} activity experimentally. A Mn^{54} source was counted with a small iron block placed over it to reproduce the backscatter of the Fe^{54} sample. An arbitrary fraction of this spectrum was then subtracted from each Fe^{54} spectrum in order to obtain a result looking as much as possible like that of a pure Cr^{51} source. The correction for self-absorption of these gamma rays in the iron was obtained by multiplying the self-absorption correction for 835-keV gamma rays by the ratio of the transmissions of iron for these two energies.

After the bombardment by 13–17-MeV neutrons, spectra were obtained rapidly to look for both the 9-min Fe^{53} activity and 20-min Mn^{52m} activity. The latter would have been made by the reaction $Fe^{54}(n,t)Mn^{52}$. No other activities were expected to be observed because the product nuclei of (n,d), (n,He^3), (n,np), and $(n,2p)$ reactions are all essentially stable. Present in the spectra in addition to the Mn^{54}, Mn^{56}, and Cr^{51} gamma rays were annihilation radiation, a 383-keV gamma ray and a 910-keV gamma ray interpreted as a sum peak of the first two, all attributed to Fe^{53} activity, and a 1.37-MeV gamma ray. This last was attributed to Na^{24} from any or all of the following: $Mg^{24}(n,p)Na^{24}$, $Al^{27}(n,\alpha)Na^{24}$, and $Na^{23}(n,\gamma)Na^{24}$. All three initial nuclei are among the known impurities of the Fe^{54}. No trace was seen of the 1.43-MeV gamma ray which is most prolific in the decay of Mn^{52}. No attempt was made to calculate an upper limit to the cross section of the (n,t) reaction due to the uncertainty of the decay mode. The 1.43-MeV gamma ray of Cr^{52} follows the beta decay of both the Mn^{52} metastable and ground states. However, their half-lives differ by a factor of 400. Since there is no way of knowing in what ratio these states are formed,

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[15] H. C. Martin, B. C. Diven, and R. F. Taschek, Phys. Rev. **93**, 199 (1954).

TABLE II. Fe^{54} reaction cross sections (mb).

E_n (MeV)	ΔE_n (MeV)	(n,p)	(n,α)	$(n,2n)$
2.23	±0.075	61.8±5	6.53± 3.33	
3.22	±0.125	216.0±17	6.42± 5.16	
4.26	±0.145	385.0±32	4.91± 2.19	
4.69	±0.085	395.0±33	2.71± 1.24	
5.38	±0.130	481.0±39	10.04± 3.68	
6.18	±0.080	565.0±46	16.29± 1.75	
13.10	±0.080	462.0±37	72.1 ±14.5	
14.05	±0.105	368.0±29	91.6 ±37.1	2.83±0.5
16.75	±0.055	204.0±16	96.2 ±15.7	50.4 ±5.0

this same factor would be an uncertainty in the meaning of such an upper limit.

The Fe^{53} activity was analyzed by counting the annihilation radiation because the 383-keV gamma ray, although clearly present, is only about one-fifth as intense and would have been considerably less reliable statistically. The sum peak was not usable as it was not well resolved from the 840-keV Mn peak. Although no other source of annihilation radiation was expected (in the absence of the Mn^{52} gamma ray), further proof that it originated with Fe^{53} was sought. A half-life measurement was made which gave a value within 3% of the accepted Fe^{53} value. The self-absorption correction for the 511-keV radiation was obtained as for the Cr^{51} gamma ray.

The Mn^{56} activity from the natural iron samples was analyzed only through the 845-keV gamma ray. The other characteristic gamma rays of 1.81 and 2.12 MeV are weaker in intensity by factors of 4 and 6, respectively. The samples were only 0.005 in. thick so that self-absorption was not significant.

In measuring the $Al^{27}(n,p)Mg^{27}$ cross section, both the 0.834- and 1.015-MeV gamma rays were used with an abundance ratio of 70/30 as reported by Monaro *et al.*[16] Again the sample was thin enough to neglect self-absorption.

The indium samples were all counted several hours after exposure to reduce the effect of capture-gamma radiation which has a 54-min half-life. For the isomeric transition, a conversion coefficient $\alpha=0.98$ was used in accordance with Martin *et al.*[15] Self-absorption in the indium samples was directly measured by counting various thicknesses of indium which had all been exposed to the same neutron flux. Counts were plotted as a function of thickness of indium, a smooth curve drawn and extrapolated to zero thickness.

Each activity in each sample was counted from four to eight times. At least one sample of each activity was counted for one half-life. The maximum statistical uncertainty for each Fe^{54} cross-section measurement was 0.5% for the (n,p) reaction, 25% for the (n,α) reaction, and 5% for the $(n,2n)$ reaction.

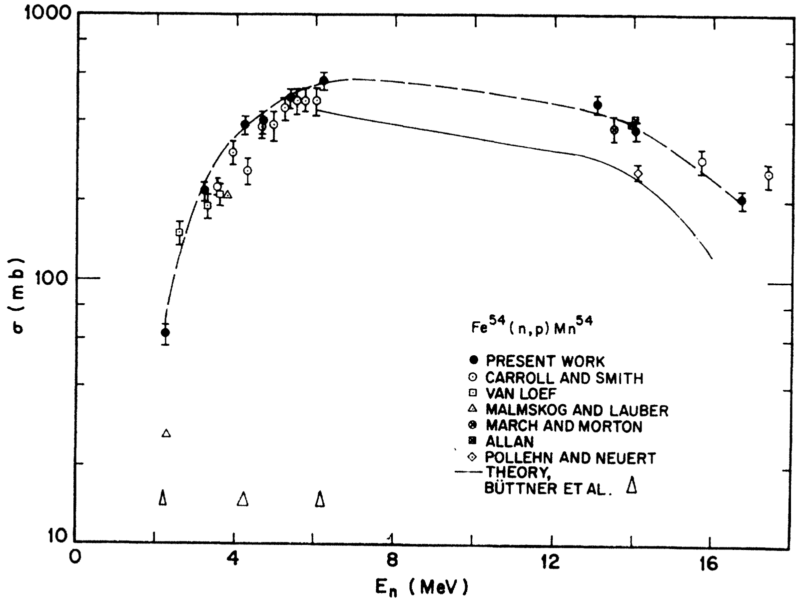

FIG. 3. The $Fe^{54}(n,p)Mn^{54}$ cross section. The solid line shows the result of a statistical-model calculation. The dashed line connects the results of the present work.

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[16] S. Monaro, G. B. Vingiani, and R. Van Lieshout, Physica 27, 985 (1961).

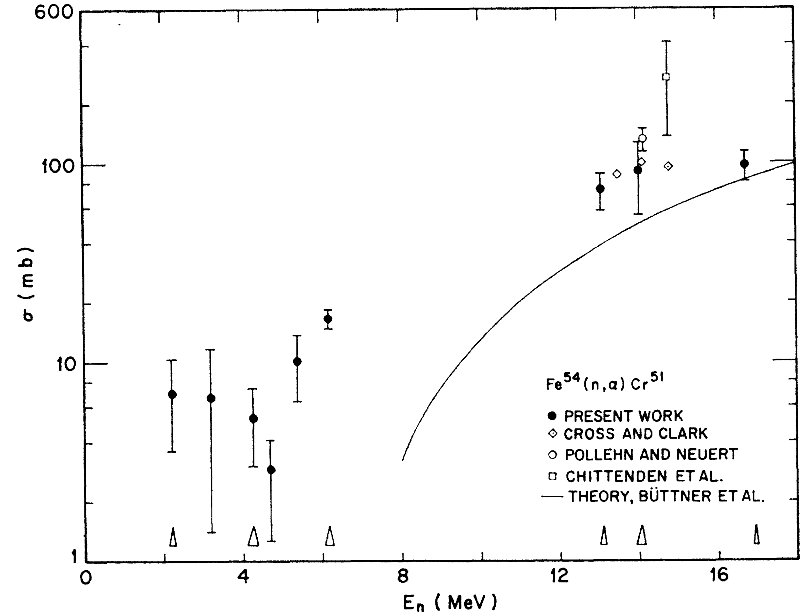

FIG. 4. The $Fe^{54}(n,\alpha)Cr^{51}$ cross section. The solid line shows the results of a statistical-model calculation.

Individual photopeaks were fitted to a Gaussian distribution by a least-squares computer code and their areas calculated from the Gaussian parameters. The half-width, peak value, and mean of the distribution were considered variable parameters. For different spectra of the same activity taken at constant gain, the half-width and mean values were determined from statistically good data and fixed at these optimum values to determine peak counts for cases with poor statistics. All such counts for a given gamma ray in a given sample were then twice fitted to an exponential decay curve by another least-squares computer code. The half-life was first considered unknown (yielding the measured values in Table I) and then fixed at the

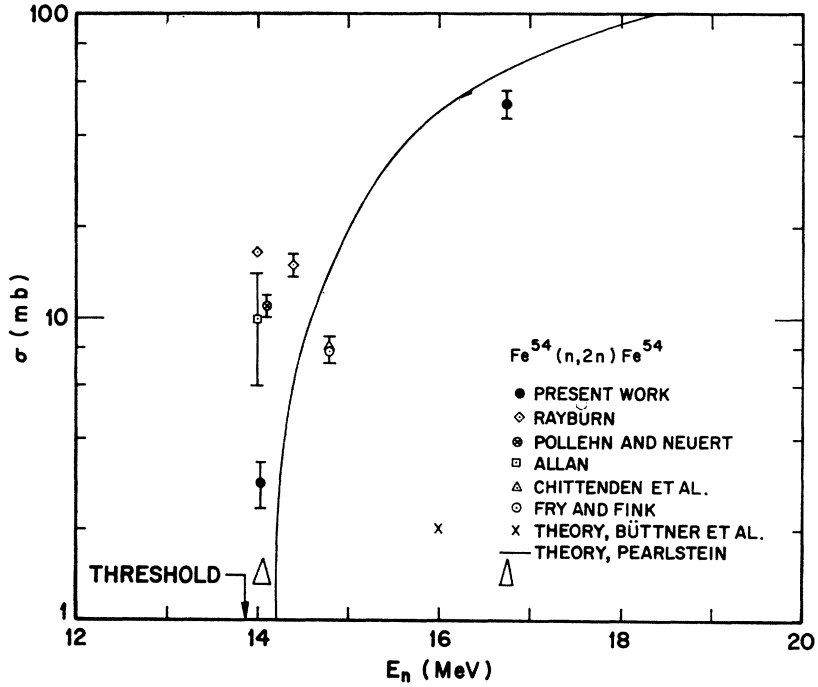

FIG. 5. The $Fe^{54}(n,2n)Fe^{53}$ cross section. The calculation by Büttner *et al.* gave zero values at 14 and 15 MeV in addition to the point shown. The curve shows the results Pearlstein (Ref. 25) obtained using a hybrid theoretical and empirical procedure in a statistical-model framework.

literature value.[17] The latter decay curve was extrapolated to zero time to give the disintegration rate at the end of the bombardment which was used to calculate the cross section.

RESULTS AND CONCLUSIONS

The results for the $Fe^{54}(n,p)Mn^{54}$, $Fe^{54}(n,\alpha)Cr^{51}$, and $Fe^{54}(n,2n)Fe^{53}$ reactions are given in Table II. The neutron-energy spread shown is the interval within which 90% of the neutrons lie. The cross-section uncertainty is the rms sum of all known uncertainties.

Since the completion of the present work, Carroll and Smith[18] have reported an additional experimental result. Their experiment used an activation technique exposing natural Fe samples one at a time. Absolute neutron flux was determined with a proton-recoil telescope placed in line with the sample. Only activity from the (n,p) reaction was reported.

Experimental results may be compared to a statistical-model calculation which has been made by Büttner, Lindner, and Meldner[19] from 6 to 18 MeV. The energy-level density in this region is sufficiently high to permit the use of a statistical theory.

This calculation, which neglects direct interactions included the (n,p), (n,α), $(n,2n)$, (n,n'), (n,np), and (n,pn) reactions. They used the nonlocal optical potential of Perey and Buck[20] for the calculation of neutron cross sections. For protons the potential of Perey[21] was used and for alpha particles that of Huizenga and Igo.[22] Level densities were gotten from Lang.[23] The effect of gamma emission was gotten from the formula of Axel.[24] In addition, the other two-body emission reactions $(n,2p)$, $(n,n\alpha)$, $(n,\alpha n)$, $(n,p\alpha)$, $(n,\alpha p)$, and $(n,2\alpha)$ were calculated but found to have cross sections less than 1 mb. The consideration of the two-body emission reactions shows the important effect they have in decreasing the (n,p) and (n,n') cross sections.

Our results for the (n,p) reaction are shown in Fig. 3 with the general trend indicate by the dashed line. It agrees in shape with, but lies about 50% higher than, the statistical-model calculation which is shown as a solid line. Triangles indicate the neutron-energy spread. The present work gives somewhat higher results than earlier work for the 2–6-MeV region.[1,2] It agrees, however, within experimental uncertainties, with much of the concurrent work of Carroll and Smith[18]. The Van Loef activation measurements[1] were normalized to the Ni^{58} and $Zn^{64}(n,p)$ cross sections which had themselves been normalized to the $P^{31}(n,p)$ cross section. The Malmskog and Lauber data are preliminary results obtained by counting the emitted protons.[2]

The present work agrees well with the mean of the various 14-MeV points, as well as with the higher energy points of Carroll and Smith.

Figure 4 shows the cross sections found in this and other experiments[5,6,7] for the $Fe^{54}(n,\alpha)Cr^{51}$ reaction. The curve indicates the theoretical results which are an order of magnitude low at 8 MeV but approach our results at 17 MeV. There is an apparent minimum at 4.2 MeV; however, the large statistical uncertainties at the lower energies make interpretation difficult. Additional measurements in this region would be of interest. Other work has been done only around 14 MeV and the present work agrees well with the other measurements.

The present work and other measurements[4,5,7,8,10] made for the $Fe^{54}(n,2n)Fe^{53}$ reaction are shown in Fig. 5. The threshold at 13.9 MeV allowed only two measurements of this cross section. These indicate that the cross section rises rapidly from threshold in agreement with earlier results, and also suggest the shape of the curve to higher energies.

The calculation by Büttner *et al.* which was made at 1-MeV intervals gave a nonzero value only at 16 MeV. This value was about an order of magnitude lower than experiment and is shown as a single point.

An additional predicted curve for the $(n,2n)$ cross section is shown in Fig. 5 based on the work of Pearlstein on the systematics of $(n,2n)$ reactions on a wide variety of nuclei.[25] The empirical curve of Flerov and Talyzin[26] is used to obtain an estimated total nonelastic cross section at 14 MeV. A second empirical curve is used to obtain the fraction of decays through channels emitting only neutrons and gamma rays. Finally, the energy dependence of the $(n,2n)$ reaction from threshold to the opening of the $(n,3n)$ channel is found by a statistical-model calculation. Assumptions in the calculation include use of a simplified level-density expression omitting pairing energy and the energy independence of the sum of all neutron-emitting cross sections. The cross sections obtained by this calculation are in reasonable agreement with the 14.05- and 16.75-MeV measurements of this experiment.

For the reactions considered in this experiment the theoretical calculations of Büttner *et al.* are in general low, disagreeing by as much as an order of magnitude. These authors have compared similar calculations with experiment for a number of reactions in medium-weight nuclei and have found the same order of agreement.

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[17] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C., 1964).

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