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FAST NEUTRON CROSS SECTION REQUIREMENTS FOR NUCLEAR ENERGY APPLICATIONS

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INTRODUCTION

In the early days of the Manhattan Project when an unknown cross section was needed, the procedure for obtaining a value for it was simple. You went and asked Fermi. Invariably he would refuse to hazard a guess. The next step, so the story goes, was to recite slowly a long string of numbers, and if one of the numbers produced a gleam in Fermi's eye - that was the value to use!

Even in the lifetime of the Manhattan Project this was not considered to be an entirely adequate procedure, and considerable effort in machines and men was devoted to measuring the needed neutron cross sections. Since then, the scale of programs for cross sections measurements has expanded continuously, resulting in the development of many ingenious measuring techniques and bearing more than a few rich by-products for the growth of nuclear physics. (The birth of the optical model of the nucleus resulted directly from measurements of fast total and elastic scattering cross sections that were requested by reactor physicists.) But there has been a more than equal growth in the amount of cross section data required for the further development of nuclear energy. Today, the need for further measurements of microscopic neutron cross sections is greater than ever.

Various countries and organizations have issued lists of the presently unknown cross section data felt to be necessary for their nuclear energy programs. Out of these "request lists" one can form a comprehensive picture of what microscopic neutron cross sections still need measuring in order that nuclear energy reach successful and economic development. It is intended to present here something of this picture, particularly the part that concerns fast neutron cross sections. By "fast" is meant incident neutron energies from the order of 50-100 KeV at the lower end to about 20 MeV at the upper limit. This is the region where by-and-large electrostatic generators, in their single or multiple versions, are still the accelerators of choice for producing the neutron sources for cross section measurements. The particular energy regions to be emphasized within this span will first be described in a general way, along with the kinds of cross sections and nuclei of interest (Sect. I). A few specific cross section problems will then be discussed (Sects. 2-4) in greater detail as illustrations of the broader picture.

1. Survey of the Fast Neutron Cross Section Needs

The needs for fast neutron cross sections in the peaceful applications of nuclear energy arise from four general sources:

1. Design of predominantly thermal reactors
2. Design of "fast" reactors
3. Protection against reactor radiation
4. Techniques for measuring neutron flux and spectra

Each of these sources emphasizes a different portion of the energy region. In the design of thermal reactors the paramount interest is obviously in cross sections in the eV range, and the high energy cross section enters in only peripherally - leakage, fast effect, activation of coolants and structural materials. For most of these applications the cross sections are important only at energies where there are appreciable numbers of fission neutrons, i.e. mainly up to 3 MeV. But where the cross sections involved have thresholds, the energy region of interest can be considerably higher. Thus, for computing fast fission contributions, only data above the U^{238} fission threshold, about 1.6 MeV, is significant. The hard gamma rays emitted in the decay of N^{16} formed by the $O^{16}(n,p)$ reaction make the cooling water of many reactors a potent source of radiation, albeit a short-lived source. Here, however, the reaction has a threshold of about

10 MeV and the excitation curve requires measurements in the 11-18 MeV region.

Even in small, so-called "hard spectrum," fast reactors the spectrum is no more energetic than a virgin fission spectrum. Hence, most of the cross sections directly influencing reactivity and breeding ratio are of critical importance only up to 3 or 5 MeV. Most current fast reactor designs tend to a much more intermediate spectrum, and measurements to 2 MeV probably cover the majority of the sensitive cross section values. There are also of course many of the same phenomena with high-energy thresholds as in thermal reactors.

Turning to the field of shielding - protection against neutron or gamma ray radiation - the situation is markedly altered. To explain why, a short digression on some of the basic principles of neutron shielding is necessary. Absorption reactions in the MeV region usually have small cross sections and cannot be depended on as sinks to eliminate fast neutrons. Instead it is necessary first to slow down the fast neutrons at least into the eV range and then to dispose of them by radiative capture or other absorption reactions. The most efficient element for slowing down is of course hydrogen and almost every neutron shield contains hydrogen in one or more of its layers. Now, the hydrogen cross section increases quite rapidly as the neutron energy decreases, ranging from 0.9b at 10 MeV to 3b at 2 MeV to 19b by 10 KeV. As a result a fast neutron making a collision in a hydrogenous medium will have lost so much energy that it is unlikely to stray much further in its lifetime from the position of its first collision. The neutron dose at a point deep in a hydrogenous shield is therefore the result mainly of source neutrons that have penetrated to the region of the point without making any collision which results in an energy loss. The low hydrogen cross section at high energies insures that such penetrating unscattered neutrons will be of quite high energy even though there may be very few of them in the source distribution. For example, the penetration of fission neutrons through 60 cm of water is dominated by the source neutrons of about 6.5 MeV in energy, even though only about 2% of fission neutrons have energies of 6 MeV or higher. In LiH, an exotic material which is the favorite of the shielders for high efficiency neutron shields, the situation is even worse; here attention focusses on the 8-10 MeV range. For most shielding needs, therefore, the demands are for information on scattering cross sections, elastic and inelastic, particularly in the energy range from 5 to 14 MeV. In addition, the neutron in slowing down can produce secondary gamma rays, by e.g. inelastic *scattering or radiative* capture, which are often more difficult to get rid of than the neutrons themselves. There is thus considerable interest in the energy distribution of any gamma ray products of the neutron reactions.

Most of the reactions which are used to measure fast fluxes have thresholds ranging from 1 to 10 MeV. They involve chiefly (n,p) (n,α) reactions and inelastic excitations of isomeric levels. Here too the energy regions of interest are obviously considerably higher than for the purely reactor design requirements.

What nuclei are of interest? The question can be answered by a sort of qualitative histogram of the "importance" of a nucleus vs. say, atomic number. Some measure of the "importance" of a given nucleus is given by the product of the number of cross section requests involving the nucleus and the priority assigned to the request. As one might expect in such a plot the predominant peak, towering far above all other features, is in the heavy element region from $Z = 90$ through $Z = 94$ (Th - Pu). Here are the fuels and fertile materials upon which all nuclear chain reactions depend. The next largest peak is at the other end of the scale from $Z = 1$ to $Z = 14$. For the nuclei from deuterium through silicon probably only helium, fluorine and neon are of negligible importance for nuclear technology. Within this group are the moderators such as D, Be and C, coolants such as Na and Li, structural material such as Al and Mg, and shielding constituents including Li, C, N, O, and Si.

Between these two peaks one can spot smaller ones for the structural materials from chromium through copper, Z = 24 to 29, and for the "exotic" high temperature materials clustered around the magic nucleus of zirconium - yitrium through molybdenum (Z = 39 to 42). Outside of these peaks only a few other nuclei are important enough to call for specific mention - Ca (in some concretes) and the heavy nuclei for gamma shielding W and Pb. In addition we should provide a nearly continuous and low "background" representing the random nuclei of interest scattered through the periodic table - fission products, control and poison materials, threshold activants. But the major interest is confined to the relatively narrow regions of atomic number described above.

What are the kinds of fast neutron cross sections that are called for? Here again some semi-quantitative answers can be given. A little less than half of such requests are for cross sections of non-elastic, neutron emitting processes - inelastic scattering, (n, 2n) reactions, (n, pn) etc. Both total cross sections and distributions of the products in energy and angle are involved. Three other groups of cross sections form the bulk of the rest of the requests and are roughly of equal magnitude. One concerns the angular distribution of elastically scattered neutrons. Another is for the properties of fast fission, chiefly cross section and number of neutrons emitted. The last of the three is for excitation functions of activation cross sections. Somewhat smaller in volume is the group of questions on, fast radiative capture cross sections above 50 keV. Finally, a very small proportion of the requests ($<5\%$) are for total cross sections. The experimenter may note with some bitterness that the volume of requests is almost directly proportional to the difficulty of measurement. There is indeed some causal connection between the two aspects. The easy measurements are usually (but not always) made early in the game. The hard ones have tended to discourage the nuclear physicist, sending him off elsewhere to seek greener pastures.

2. Fast Neutron Cross Sections of Beryllium

These are some general characteristics of the cross section needs of nuclear technology. To give flesh and blood to the bare bones I should like to discuss in some detail a few particular examples, mainly those in which I have been personally concerned. The first of these involving reactions in Be stems mainly from reactor design requirements and proves to be somewhat of an exception to the general features we have mentioned. The Be^9 nucleus has the lowest magnitude Q value, 1.67 MeV, for the (n, 2n) reaction of any stable nucleus. In fact there is the singular oddity that the threshold for the (n, 2n) reaction is below that for inelastic scattering since the lowest excited level in Be^9 appears to be at 2.43 MeV. Almost half the neutrons in the fission spectrum are above the (n, 2n) threshold. It is therefore conceivable that the reaction could produce enough additional neutrons in the reactor to affect the neutron economy significantly. With presently available data it may be estimated[1] that in an infinite block of Be for every 11 fission source neutrons the net effect of the fast neutron reactions is to produce 1 extra neutron. This may not seem very large, but it means that in a thermal reactor for every neutron absorbed about 0.2 neutrons will result from the "fast effect" in Be. Those 0.2 of a neutron may spell all the difference between being able to breed or not. It clearly could be of considerable importance to know the (n, 2n) cross section in Be as a function of energy.

For many years the only clues to the magnitude of the cross section came from integral experiments.[2] The results were contradictory, ranging from millibarns to 4 barns! In recent years there have been a number of measurements with mono-energetic neutrons below 14 MeV[3-7]. It has been established that from 5 to 18 MeV the cross section slowly decreases from about 550 mb to 400 mb. But many of the features of the reaction, some of them the most significant for reactor applications, are still unclear. There are many possible ways in which the (n, pn) reaction can proceed, e.g.:

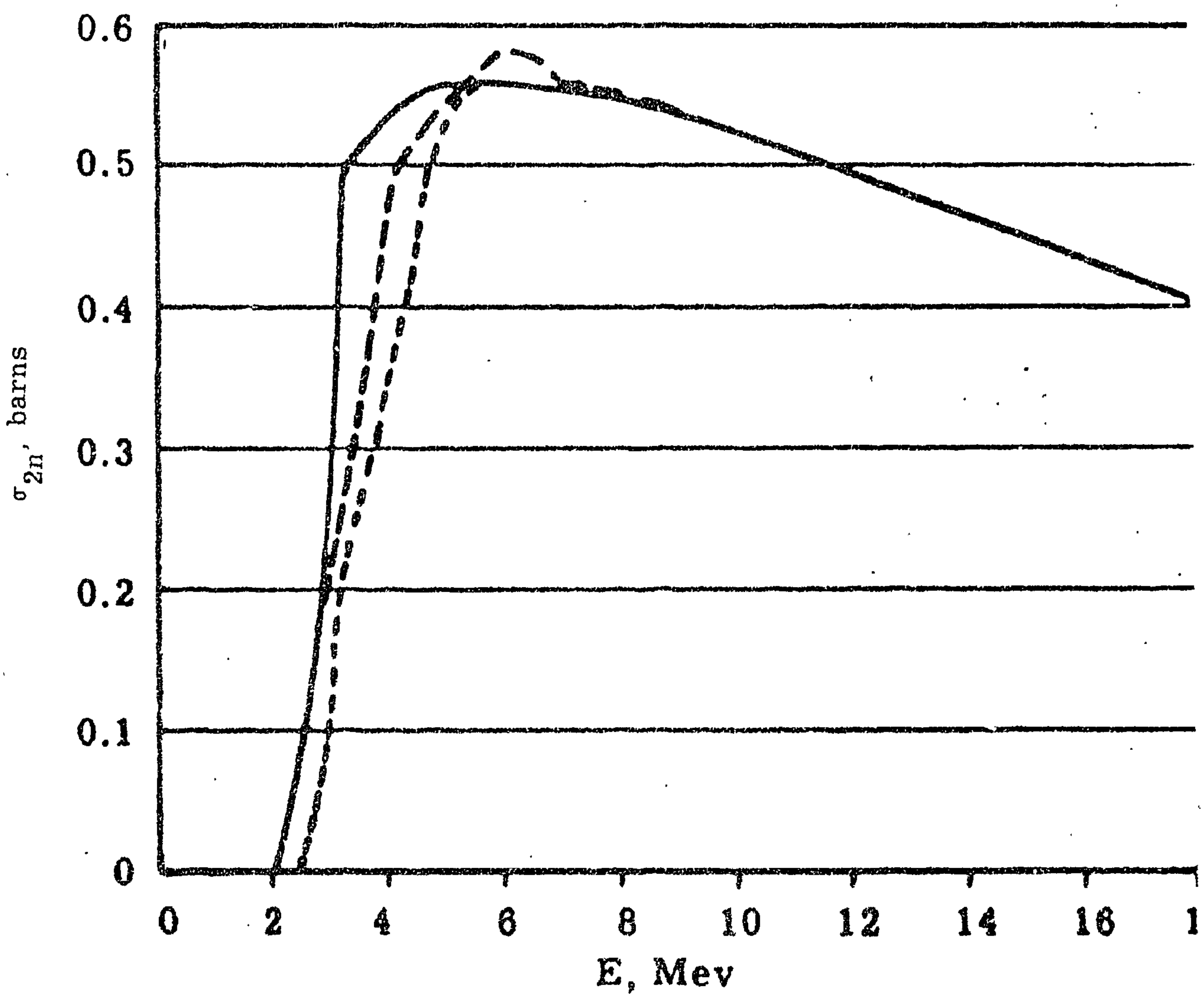
$$Be^9 + n \rightarrow Be^8 + n' + n'' \rightarrow 2He^4 + n' + n''$$

$$\rightarrow He^5 + He^4 + n' \rightarrow 2He^4 + n' + n''$$

$$\rightarrow Be^{9''} + n' \rightarrow He^5 + He^4 + n' \rightarrow 2He^4 + n' + n''$$

and so on. A large fraction of the time it seems that the reaction proceeds via an initial inelastic scattering, which can only occur above a threshold of 2.7 MeV. Indeed, there is no experimental evidence for the (n, 2n) reaction below this energy. The most recent experimenters, Cranberg and Levin,[5,6] merely give an upper limit of 100 mb for the value of the cross section between 1.7 to 2.8 MeV. Unfortunately the mean energy of the fission neutrons is just in this region. A cross section of 100 mb from 1.7 to 2.8 MeV would contribute more than one third of all the extra neutrons resulting from the (n, 2n) reaction.

In 1960, Dr. A. Krumbein and I calculated the so-called infinite medium fast effect - how many extra neutrons slow down below 1 eV in an infinite Be medium



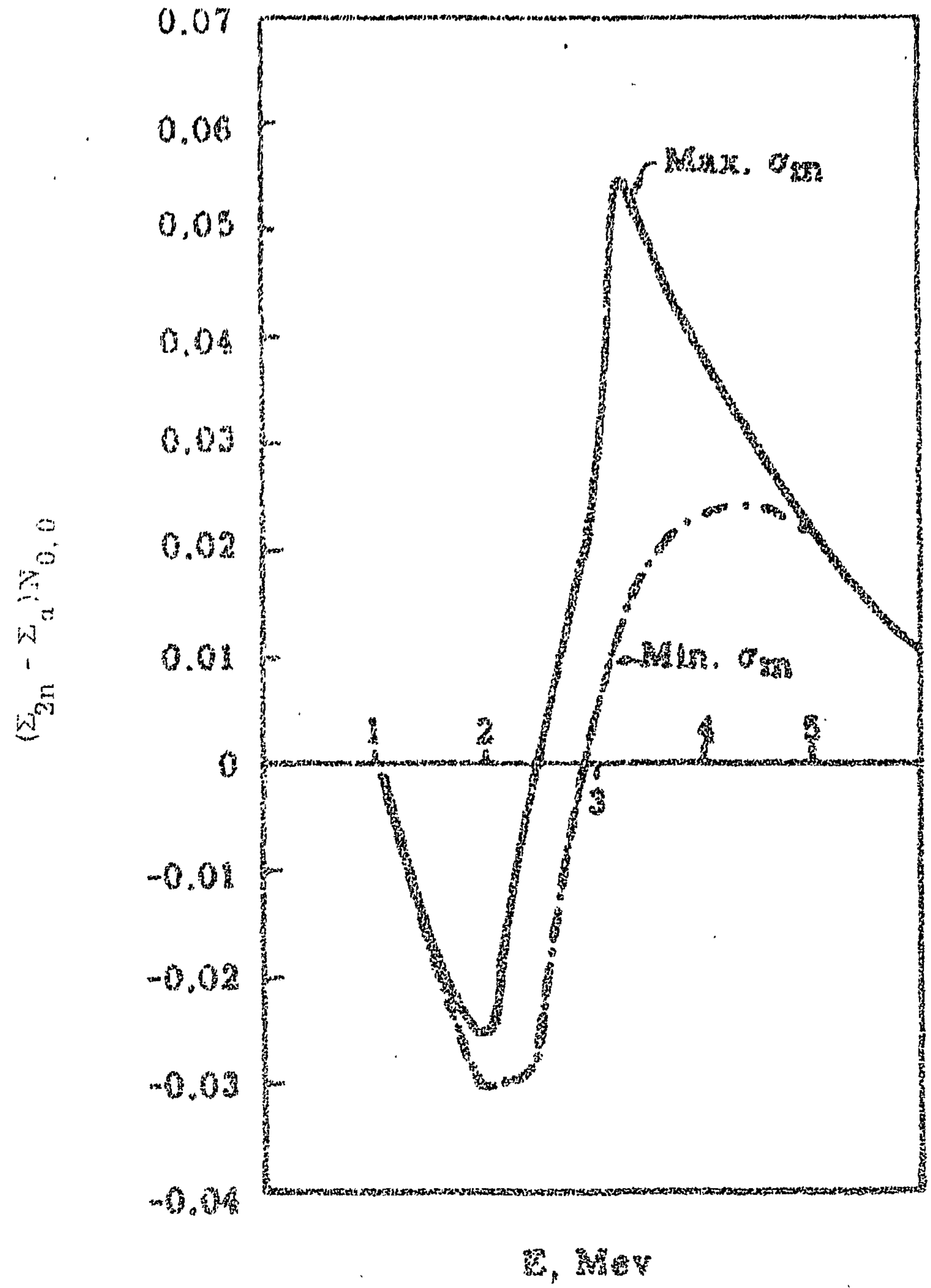
Three σ_{2n} curves for Be

FIGURE 1

containing fission sources[1]. This type of calculation obviously gives an upper limit to the gain one could expect from the reaction. Three possible (n, 2n) cross section curves were used in the calculation as shown in Fig. 1. At the time the solid curve was considered to be an upper limit, but we now consider it the closest of the three to actuality. The differences between the three curves may not seem large, but they are put in proper perspective by plotting the results of the calculation as the number of extra neutrons produced per MeV interval of the total flux spectrum in the medium. Fig. 2 shows the results for the two extreme curves. The negative values below 3 MeV are the result of absorptions by the (n, α) cross section which reaches a flat peak at about 2 MeV. That a slight shift in the cross sections in the 2 to 3 MeV region has a marked effect on the final answer is also shown in the following table:

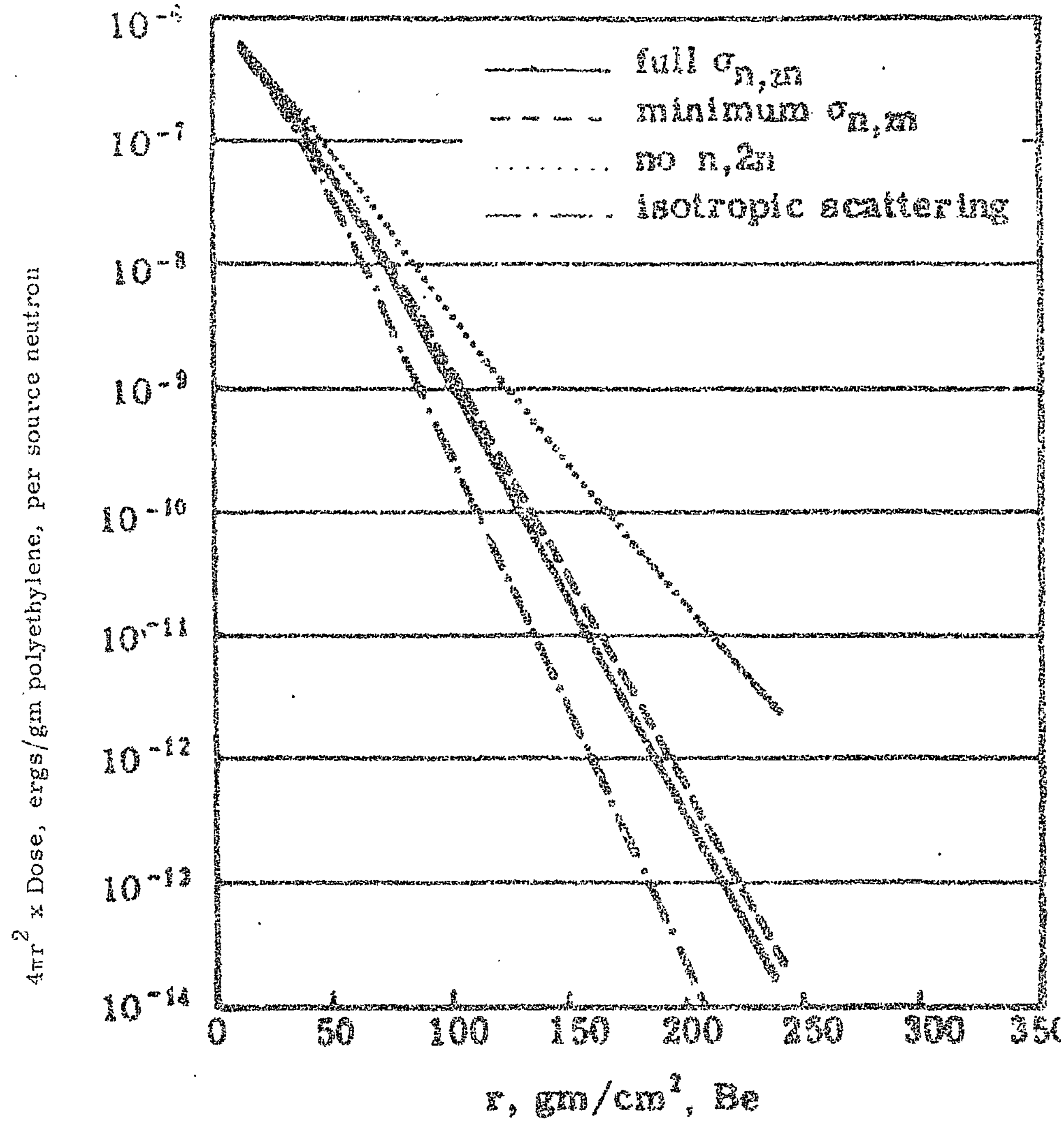
Cross Section Data	Net neutrons produces, as per cent of source neutrons
Maximum $\sigma_{n,\,2n}$	9.0%
Intermediate $\sigma_{n,\,2n}$	7.0%
Minimum $\sigma_{n,\,2n}$	3.4%
Maximum $\sigma_{n,\,2n}$, Isotropic elastic scattering	6.8%

The last entry shows the effect of making a very drastic change in the angular distribution of elastically scattered neutrons - assuming isotropy in the center-of-mass system. The change is clearly in the right direction - isotropic scattering means the neutron loses more energy per collision and therefore stays less time in the energy region where the (n, 2n) reaction is significant. But it is not a very large effect considering the radical change assumed in the angular distribution. Our present knowledge of the angular distribution in Be is probably adequate for calculation of the fast effect. This conclusion may need modification in practical reactor designs where the Be would be used in finite geometries and the fast leakage would be more seriously affected by how the neutrons scatter elastically. The spectrum of neutrons produced in the (n, 2n) reaction likewise has little influence on the value of the fast effect in an infinite medium but is of somewhat more importance in an actual reactor lattice. To evaluate the Be fast effect what is primarily needed therefore is more information on the threshold behavior of the total (n, 2n) cross section; how the neutrons scatter, and the distributions of the reaction products, are of secondary significance only.



Be Fast Effect Response Curves
Problems 1 and 3 May 1961

FIGURE 2



Dose-Distance Curves, Fission Source in Be

FIGURE 3

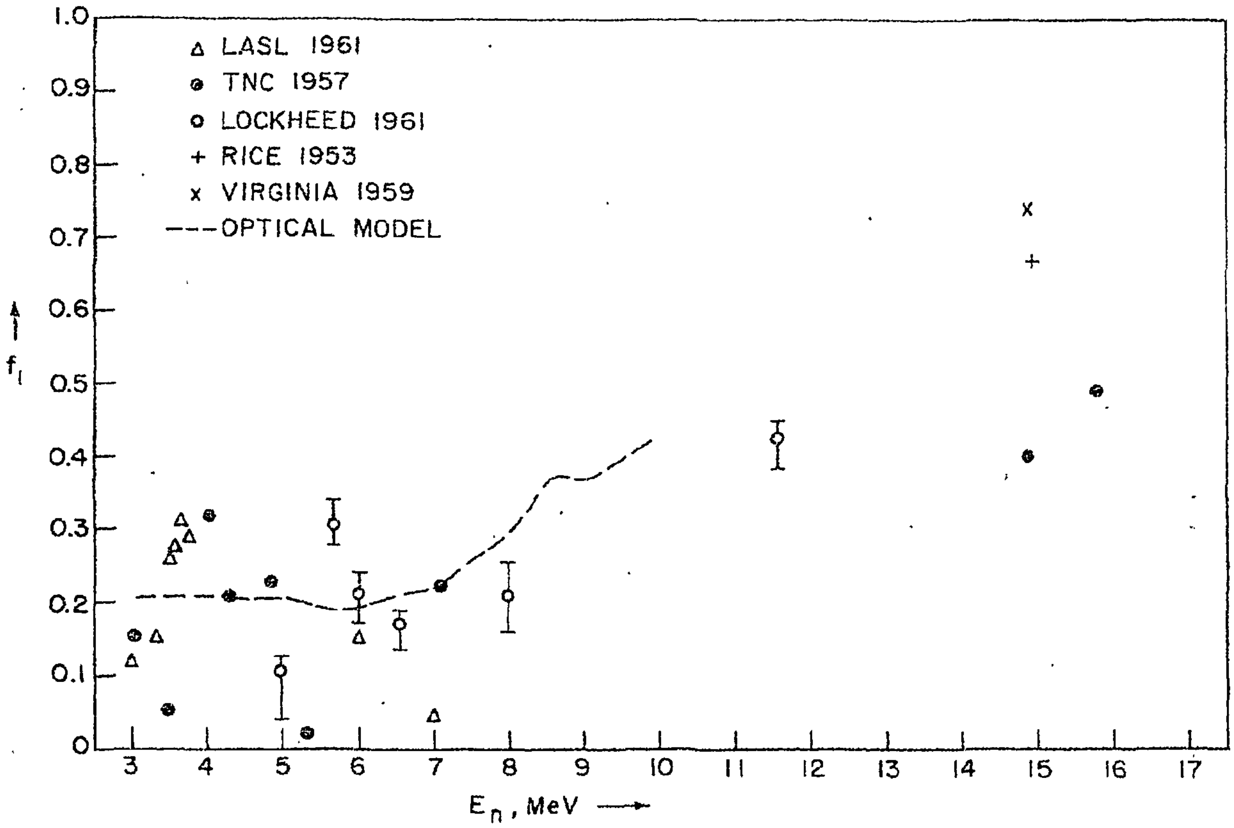

1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
f_l
△ LASL 1961
⊗ TNC 1957
○ LOCKHEED 1961
+ RICE 1953
x VIRGINIA 1959
---OPTICAL MODEL
3 4 5 6 7 8 9 10 11 12 13 14 15 16 17
E_n, MeV

FIGURE 4

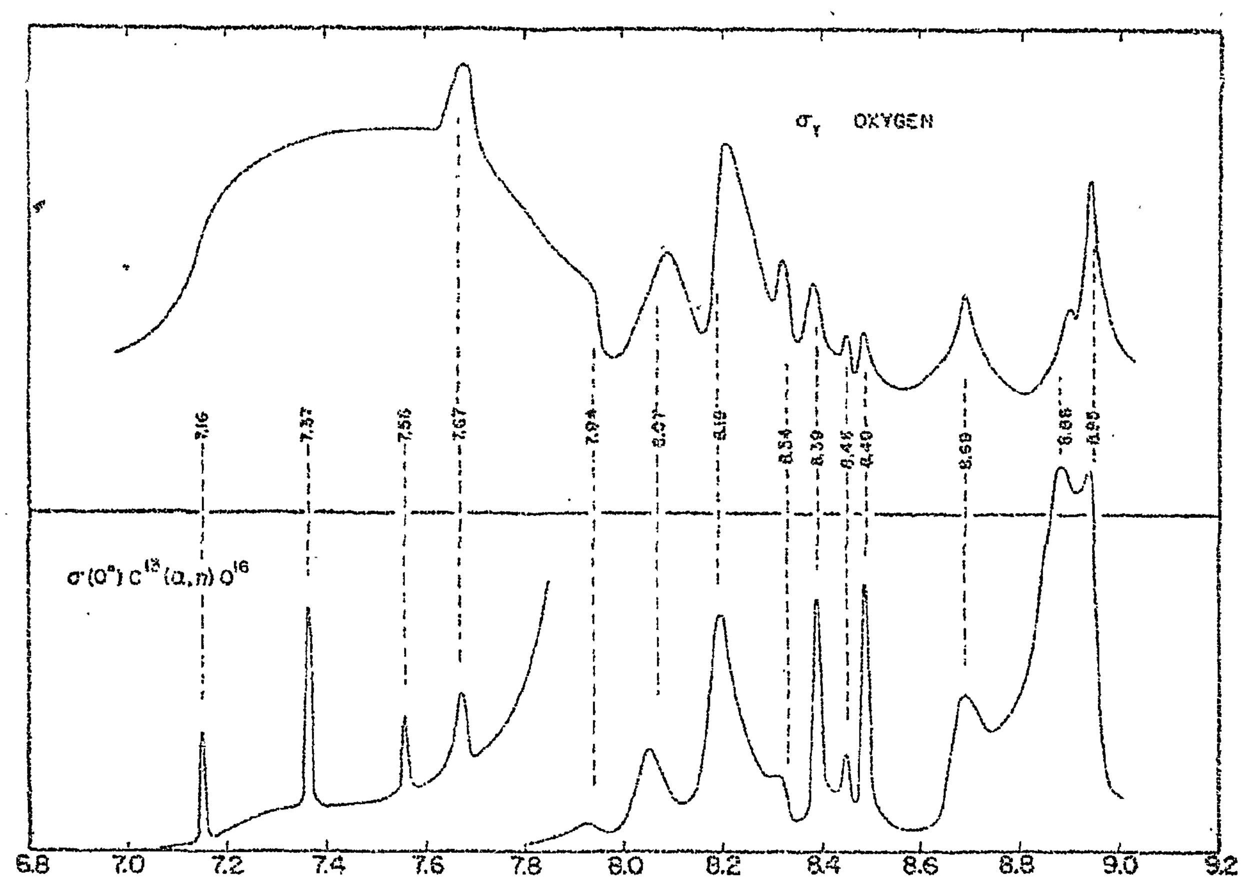
The situation is quite different if we consider the use of Be as part of a neutron shield. Fig. 3 shows the calculated fast neutron doses from a point fission source in Be for various cross section assumptions. Note that at a thickness of about 150 gm/cm^2 (about 1 meter), the maximum variation in the (n, 2n) cross section affects the dose by less than 10%. On the other hand at the same distance the assumption of isotropic elastic scattering changes the dose by a factor of 10. While such thick Be (or even BeO) reflectors are not likely designs, the extreme sensitivity of the penetrated flux to the nature of the scattered angular distribution is found in other, more practicable materials.

3. The Angular Distribution of Elastic Scattering by Oxygen

Oxygen is a good case in point. It is an almost invariable component, in one compound or another, of any shield (and of many reactor cores). Up to 3 MeV the angular distribution for elastic scattering has been very well measured at ORNL, ANL, and other laboratories[8]. The information here is more than adequate, but above 3 MeV there is mostly the darkness of ignorance, and the shadows are only slowly being pushed back. For many applications it is convenient to represent the shape of the angular distribution by an expansion in Legendre polynomials:

$$\sigma_{n,n}(E, \theta) = \frac{\sigma_{n,n}(E)}{4\pi} \left[1 + \sum_{1}^{\infty} (2\ell + 1) f_{\ell} (E) P_{\ell}(\cos\theta) \right]$$

The first coefficient in the series, f_1, is identical with the average angle of scattering (in the C.M. system). Some idea of the task confronting the user of cross sections may be gleaned from Fig. 4 which shows values of f_1, obtained from most of the recent measurements of angular distributions on O above 3 MeV. In our earliest attempts at providing an evaluation of the data, effort was concentrated especially in extending the lower energy results up from 3 to 4 MeV. In this region there were some very old and dubious Swiss data[9] (not shown) and three newer (but probably equally dubious) points from Texas Nuclear[10]. These flatly contradicted each other. Devious ways were tried to make sense out of the data, but nothing even reasonably adequate was obtained until Philips at LASL measured the angular distribution for some 6 energies between 3 and 4 MeV. His work gives a minimally adequate picture of the angular distributions in this region, at least from the point of view of the applications.



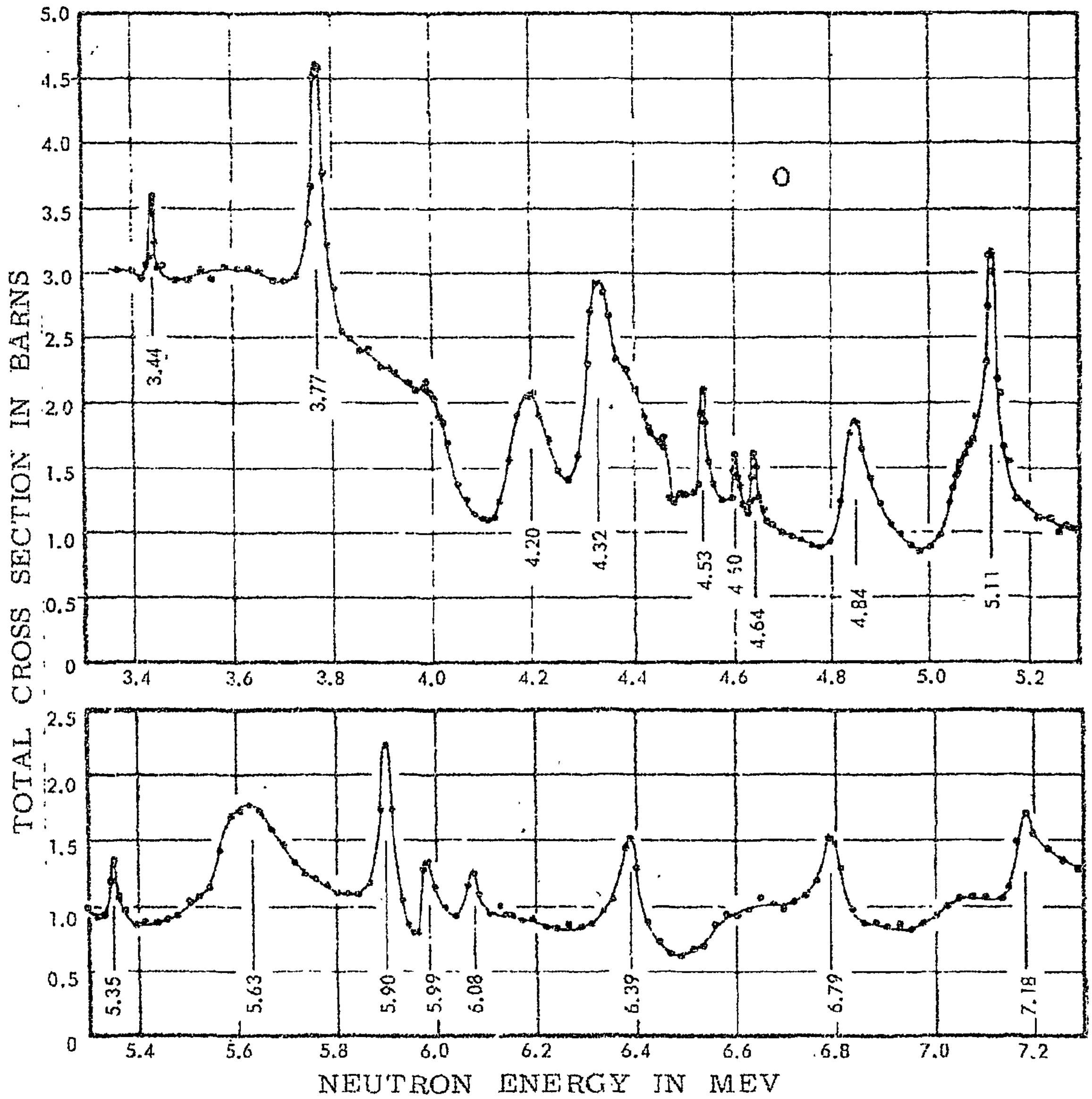
EXCITATION ENERGY OF O^{17} IN MEV

The total neutron cross section of oxygen and the $C^{13}(\alpha, n)O^{16}$ 0^{o}-yield plotted as a function of excitation energy of the compound nucleus O^{17}. The oxygen total cross section shown includes data taken from references 19 and 20. The energy scale of the data from reference 20 was corrected for target contamination.

FIGURE 5

But our earlier "devious" investigations turned up some interesting and unresolved questions in nuclear physics. We tried to see if we couldn't evolve a set of scattering phase shifts consistent with the total cross section. Fig. 5 (taken from a paper by Walton et. al.[11]) shows the total cross section as it had been measured in 1957. Underneath it is an excitation curve for the (α, n) reaction in C^{13} plotted against the corresponding excitation energy of compound nucleus O^{17}. The very broad "hill" in the total cross section has no counterpart in the (α, n) reaction except possibly in the low background between resonances. On the other hand, of the 4 narrow resonances in the (α, n) cross section only one, at 3.77 MeV, showed up in the total cross section measurement. Now, the resonances are too narrow ($\leq$ 25 keV) to be of any significance in reactor of shielding calculations - the interest rather is in the round hump. What produces it? It's of the wrong shape and much too high to be a single level. J. L. Fowler and H. O. Cohn [12] had evolved a single particle model which fit the scattering data (including resonances) below 3 MeV, but no reasonable modification of it provided anything like the hump above 3 MeV (Kalos, unpublished 1959). H. Lustig in 1959 made a valiant effort to analyse the data on the basis of a model with spin-orbit coupling and using some of the information from the (α, n) reaction. He concluded (NDA 2111-3, Vol. A., 1959) that the hump conceals as many as 5 levels in it, but the results are not convincing or satisfying. As he himself notes, five levels gives one a large number of empirical parameters to play around with. Two of the resonances have unreduced widths of about 500 KeV, with angular momenta of $\ell = 1$ and 2. It is very hard to fit such peculiar levels into any of our present nuclear models.

Since that time the total cross section has been remeasured by Fossan et. al.[13] who have found one more of the narrow resonances, as shown in Fig. 6. It is quite likely that careful high resolution measurements would turn up the remaining two. Hunzinger and Huber[14] in Switzerland have made angular distribution measurements at 100 KeV intervals in this region. Unfortunately, the technique used did not permit measurement to small angles, so they completely missed the prominent "diffraction" peak in the forward directions. They also did not see any of the narrow resonances, but here their energy interval was just too large. A. L. Sayres[15] at Columbia, using a similar technique, did find one of the resonances - that at 3.77 MeV. The angular distribution shows some complicated interference



The total cross section of O. Each dot represents one data point. Statistical errors are less than 3%. The neutron energy spread varied between 15 and 20 kev.

FIGURE 6

effects. He plans to repeat the measurements with 25 keV steps, decreasing to 8 KeV at the resonances. The narrow resonances themselves are not of much interest. But the interference they exhibit with the broader background should be very helpful in pinning down its nature. It is hoped in this way to determine definitively the nature of the levels producing the hump and how they fit in with nuclear models. From the point of view of the practical application this is a side diversion. But it's one that may have very interesting consequences for nuclear physics, and one which wouldn't have been uncovered except for the original requests of the reactor and shielding people.

Above 4 MeV the total cross section of oxygen shows partially resolved resonance structure which is not yet blurred into a continuum even at 10 MeV. It would be hopeless experimentally to measure the angular distribution with all the detail of the resonance structure, nor would it be useful for the applications of the data. Clearly what is wanted is that the measurements be made with sufficiently poor energy resolution as to average over the fine resonance structure; I am not sure all experimenters realize this requirement. Actually, there are many more high energy measurements available for oxygen than for most other materials, but as the scatter of points shows this is not an unmixed blessing. There are many instances of widely discrepant measurements as at 7 MeV and 14 MeV. In deciding between them one must often use subjective criteria. The distributions from X were obtained when the outfit was just starting and they were short on both equipment and experience, while those from Y were done at a laboratory where the people know all the pit-falls and had (at the time) the world's best setup. And rightly or wrongly such factors sway one's judgment.

How one's evaluation of the situation changes with time is shown by Fig. 7 which reproduces all the points of Fig. 4 and adds some smooth curves drawn at different times for sets of cross section data to be used in reactor and shielding calculations. The solid line comes from a set published in 1958 at a time when the Texas Nuclear data were still tentative. The next set, published a little over a year later leaned heavily on the Texas Nuclear work as the most extensive available. These data led to angular distributions much less anisotropic than the 1958 list, especially from 5 MeV on. Within the next few years more measurements were made at Los Alamos, Lockheed and the University of Virginia. They don't add up to any consistent picture, but they did seem to indicate greater anisotropy at 14 MeV. The dotted curve shows our latest (1962) guesses.

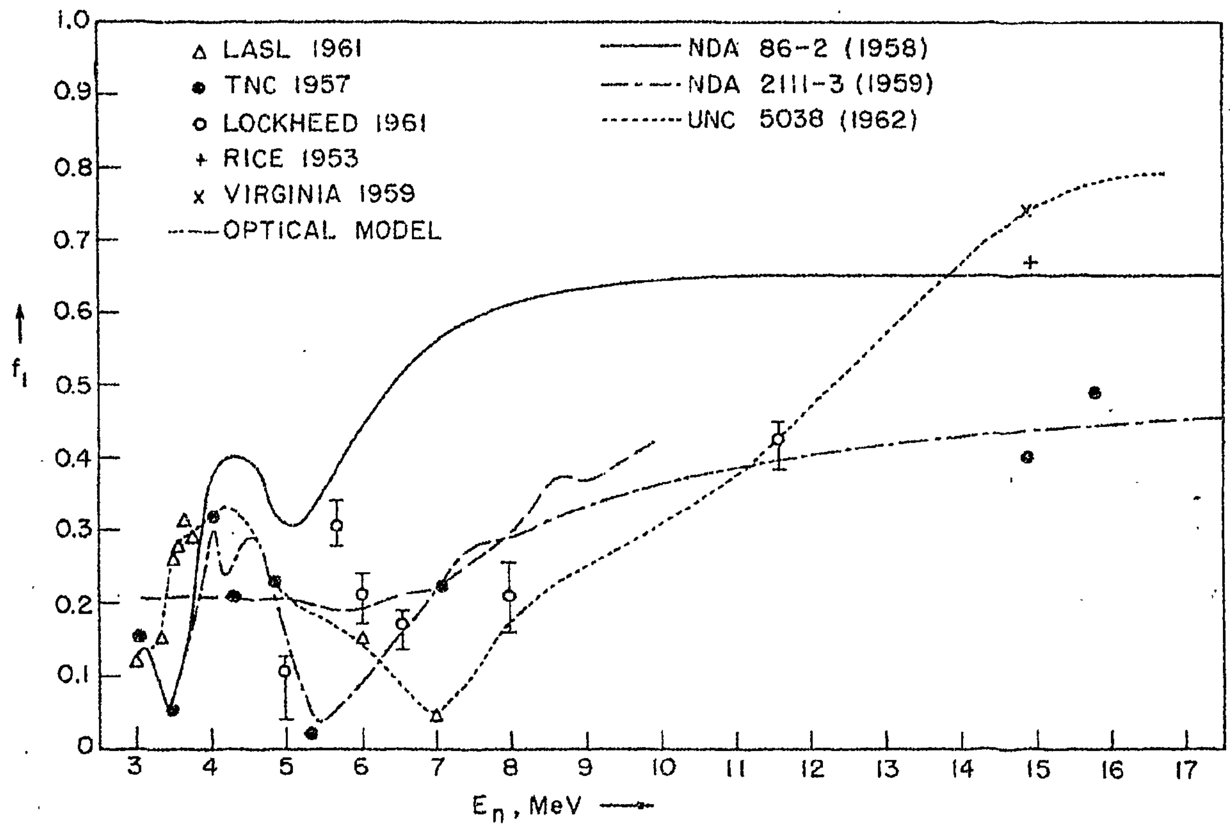

1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
△ LASL 1961
● TNC 1957
○ LOCKHEED 1961
+ RICE 1953
x VIRGINIA 1959
----OPTICAL MODEL
NDA 86-2 (1958)
NDA 2111-3 (1959)
UNC 5038 (1962)
f_1
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
E_n, MeV

FIGURE 7

What difference do these changes make? As an indication, calculations have been made of the fast neutron dose from a point fission source in water, using both the 1958 and 1959 data. At 30 cm there is no significant difference. At 60 cm the 1959 set gives an 18.5% lower dose than the 1958 set. At 90 cm the difference is 28%, increasing to almost 40% at 150 cm. Considering that the main brunt of the attenuation in water is due to hydrogen, and that the two sets differ significantly only above 4 MeV, the effect of the change in oxygen angular distribution must be considered quite impressive. The correct set of angular distribution data is still not known and many more measurements must be made before the question is resolved.

4. The Significance of Inelastic Scattering for Fast Reactors

It has been mentioned that the bulk of the fast requests are for nonelastic data, mainly inelastic scattering. Some of these come from shielding requirements, both for neutron attenuation and for determining the sources of secondary gamma radiation. But a large fraction of the interest stems from fast reactor design, and here fortunately the reasoning behind the requests and the accuracies desired is much better documented than in most other reactor areas. In 1961 the IAEA ran a conference on the physics of fast and intermediate reactors(16). A number of papers and much of the discussion was devoted to the sensitivity of various calculated aspects of fast reactors to the input microscopic data. By and large, the picture presented at that conference is still valid today, two years later. All participants agreed that the quantities needed most accurately were the data directly entering into the critically -$\nu(E)$ and the fission cross sections of the fuel. Because of the large size of presently designed cores an uncertainty of 1% in reactivity could correspond to an error of 50-100 kg of U^{235} in the critical mass. And for such accuracy in k, 1/2%-1% accuracies in ν and σ_f are needed. I think such conclusions are qualitatively understandable, and the accuracies, at least for ν, are even within reach.

But it may come as a greater surprise to learn that such an accuracy in k required 2 - 3% accuracy in the inelastic cross sections for the fuel and fertile material, and 5% uncertainties for the same cross section in structural material(17) - accuracies almost impossible to obtain with present techniques. To understand why these cross sections are so important it must be remembered that most present fast reactors are so large and so dilute in fuel that a good deal of slowing down from the virgin fission spectrum occurs. In a typical design[18] the relative nuclear concentrations might be

	n
Fuel	1.0
Fe	1.4
Na	.6
C	1.0

This is still far from the situation in thermal reactors where there may be hundreds of moderator nuclei for every fuel nucleus. But there is still enough extraneous material in the core to ensure that a fission neutron will make at least a few scattering collisions before ending its life by absorption. For such a core, for example, about half the fissions are caused by neutrons with energies of less than 250 KeV, whereas the average energy of virgin fission neutrons is 2 MeV.

One can see physically that most of this softening of the spectrum comes not from the conventional moderation by elastic scattering, but from inelastic collisions. A useful gauge of the moderating power of a constituent is $n\xi\sigma_s$ where n is the relative concentration, ξ the mean logarithmic ratio of energy loss and σ_s the scattering cross section. For the core described above these moderating powers for a 1.9 MeV neutron are roughly:

	$n\xi\sigma_s$
Fuel	1.04
Fe	.67
Na	.21
C	.69

The figure for C arises solely from elastic scattering; that for Na about equally from elastic and inelastic scattering, while for the others only inelastic scattering contributes significantly. Thus, during most of the neutron collision history in such a core the uranium and iron are as good or better moderators than carbon! To be able to calculate this moderation properly it is not surprising therefore that the inelastic scattering properties must be carefully and accurately measured.

These examples of how neutron cross section measurements are vitally necessary for the development of nuclear energy could be multiplied manifold. What is obviously more difficult to prove is the benefit towards our knowledge of the nucleus. One can but point to past contributions - resonance theory, optical model, statistical properties of levels - where neutron experiments provided the crucial ideas. The patient tilling of the vineyards of nuclear spectroscopy, however unglamourous compared to the more fashionable physics pursuits of the hour, will surely still bring a rich reward of new physical knowledge.

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