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ron Capture and Gamma-Ray Production Cross Sections

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I. Introduction

The radiative capture of neutrons plays a decisive role in the neutron balance of nuclear reactors. The accurate knowledge of these cross sections is vital to the economic design of reactor systems, not only from the standpoint of neutron economy, but also from the shielding requirements, in which capture γ-ray production must be carefully evaluated.

In the present report, the authors have attempted to summarize, and evaluate, the experimental techniques available for neutron capture cross section and spectrum measurements. The adequacy of present methods in relation to the requirements of the technical user is assessed. The requests are analyzed in terms of motivation and nature of the needs, and the practicality of the requests is discussed.

For the purposes of this report, the gamma-ray production cross-section requests are also included, although in many cases they may result from processes other than radiative capture; e.g., neutron inelastic scattering. These production cross sections are needed in shielding calculations for nuclear power technology.

The above considerations lead to a natural division of the report into three parts: (1) an analysis of the nature of requests for capture and γ-ray production cross sections, (2) the techniques for measurement of capture cross sections, and (3) the techniques for measurement of gamma-ray production cross sections. Because of the broad range of the desired information, many techniques are required. This report therefore adopts a comprehensive point of view in the main, using illustrative

specific examples to amplify the conclusions drawn. A detailed examination of each problem posed by the many requests is not feasible. It is hoped instead that the general guidelines on the ability of present day technology to meet our nuclear cross section needs will be made clear.

II. General Comments on Requests

An examination of Wash 1078 - the compilation of requests for nuclear cross section measurements, June 1967 - shows that of 411 numbered requests, 176, or about 43%, fall into the categories of neutron capture and γ-ray production. (Recent requests increase this number to 183 as of March 1968.) In the latter area fall 45 requests, mostly concerned with differential cross sections involving the gross γ-ray spectrum shape with a low resolution of 0.25 to 1.0 MeV in γ-ray energy. These requests are mainly in two subgroups: (1) low Z elements abundantly present as constituents of the earth's crust and atmosphere, and (2) the rare earth region. The capture cross-section requests fall primarily into the following categories: (1) medium weight elements comprising reactor structural materials, such as zirconium and iron, (2) fission product poisons, (3) fissionable isotopes, and (4) transuranium elements, such as those important to isotopic fuel sources. While the precisions required for γ-ray production requests are relatively modest, the capture cross-section precision required, especially for capture in the fissionable isotopes, is quite high. The problem of obtaining samples is critical in the case of fission product poisons (which may have short half-lives), and in the transplutonium elements, which are

just now becoming available through irradiations at the Savannah River and HFIR reactors. In many requests, as will be indicated below, it is the sample problem which is at present the critical factor in limiting the feasibility of the measurement. The sample problem may simply be availability or it may be in some cases the limitations imposed by sample radioactivity.

III. The Nature of the Requests for Radiative Capture and Gamma-ray Production Cross Sections

The needs for which radiative capture and γ-ray production spectra are requested may be classified in various ways: reactor or shielding, biological dose or heating effects, effects on media or upon electronic instruments. In each the requirement on accuracy, resolution and need for detailed angular information is somewhat different. It is useful to review these so as to acquaint experimentalists with the kind of information needed in each application.

1. Reactor calculations. Usually, the need for capture cross sections lies in its role in removing neutrons. Thus, it is needed as a single valued function of neutron energy. Resonance parameters for capture are a convenient parametrization and are frequently used directly in reactivity studies. In any case, narrow (e.g. p wave) and wide resonances behave somewhat differently in capturing resonances and must be distinguished, but the effect of very narrow resonances is often much more pronounced than appears in the total cross section. Thus, direct measurement of radiative capture and observation of spectrum as an aid to assigning resonance parameters are important.

Fast reactor studies require some knowledge of inelastic neutron

processes and, to the extent that measurements of the gammas produced elucidate the level structure and partial cross sections, they are helpful.

2. Reactor shielding. Gamma radiation produced in neutron reactions play a significant role in determining biological dose and similar effects outside a reactor shield. This is especially true for high efficiency designs as in space applications. Many different reaction modes can be accompanied by photon emission, but radiative capture and neutron inelastic scattering are most important. The sources in the shield itself are particularly significant, especially in or beyond heavy materials used for structure or for shielding against core γ rays.

These materials are often adjacent to core moderators or hydrogenous neutron shielding material in which a strong epithermal spectrum may be present. The resulting capture gammas are an important source. To predict resulting effects, some knowledge of the resonant capture cross section and spectrum in different resonances, or at least classes of resonances is needed. Since the penetration of the radiation in the heavy materials in which it is produced depends strongly upon energy, the variation from level to level of ground state or other energetic transitions is very important.[1] The effect in shielding of the correlation of ℓ value (which determines, to some extent, self-shielding effects and hence the probability of capture) and spectrum has not been investigated in any detail, so it is not clear what kind of detail is needed. It is likely that a distinction among several broad classes of resonances is sufficient. These might be grouped by ℓ value or in wide energy ranges.

As indicated above, gamma radiation from inelastic scattering is another important source. The uses of these data are essentially the

same as for capture, except that anisotropy of the spectrum is more likely to be significant. In the past, the angular distribution has been taken to be isotropic on the assumption that variations of $\sigma(n)$ of less than about 50% were unimportant in calculations. Goldstein and Ozer[2] find that for calculations of total neutron flux in homogeneous geometry specification of angular distributions of elastic scattering through P_3 gives errors of only 5% in relatively deep penetrations. It is not an implausible inference that measurements which define the γ-ray angular distribution through P_2 are likely to be sufficient for most applications. In applications of ordinary reactor shielding, gammas from neutrons above 8-10 MeV are not likely to be critical, but several requests are intended to assess the trend of spectra in this range. Presumably the spectrum can be predicted in the continuum range but there is inadequate data as yet.

3. Reactor and shield heating. Radiation heating, especially in shields is an important consideration in reactor engineering design. The need for data is entirely similar to that mentioned above except that low energy photons may be of greater interest if they represent a significant fraction of the energy release. Their effect is nearly the same even when internally converted. Thus, they can often be accounted for by considerations of total energy release.

The rest of this section is devoted to a discussion of specific requests or classes of requests in the light of the remarks of the preceding paragraphs.

Requirements for γ production in O, Fe, C, Ni, Cr, Mn, and Zr is motivated by reactor shielding investigations. As has been

emphasized above, the necessity for data above about 8 MeV has not been established in this application. Thus, it would appear that the need for new C (#408) or O (#409) data is marginal. One requestor commented that a reassurance that the γ production goes as expected would be welcome. Specifically, he would like to check that the total cross section for gamma production in C and O at (about) 1 MeV is less than 10 mb and that the cross section near a few volts can be extrapolated as 1/v from thermal.

Need for data on Fe should be satisfied by measurements up to 7.5 MeV. Some indication of angular distribution would be useful, but unless variations of more than about 50% from average values it need not be very detailed.

Data for Cr, Ni, and Zr can be inferred from neutron measurements and also from theory. No doubt some up-to-date measurements, especially at about 7 MeV, would be most helpful in buttressing such results.

The applications to reactor shielding extend down to about a volt in neutron energies. The structural materials Cr, Mn, and Ni offer with Fe the problem of finding all resonances that account for the capture integral. Thus, there is some problem in predicting gamma production and direct measurements of it are needed. The same is true of Zr particularly because of the large p-wave contribution to the capture and consequent expected variation of capture spectrum.

IV. The Techniques for Measurement of Radiative Capture Cross Sections.

In general the ability to measure an absolute capture cross section involves a knowledge of the (1) incident neutron flux, (2) the number of sample nuclei, and (3) the detection efficiency. With the exception of the spherical shell transmission technique, which is essentially a transmission measurement in which scattering is canceled out because of geometrical considerations, all three of the above factors must be known and will contribute to experimental uncertainties. In most practical cases, however, only cross section ratios are measured, and the reduction to absolute values involves only the use of certain standard cross sections. The latter assume paramount importance, and the available cross section precision is limited by a knowledge of these standard cross sections.

Before entering into a discussion of the precision presently available, we summarize briefly the commonly used neutron sources and detection methods.

Sources

(a) Broad Range or "White Spectrum Sources" include nuclear reactors and accelerator-based photoneutron and charged particle induced evaporation neutron sources. The latter includes linac and cyclotron sources in which the high energy neutrons are moderated to produce a slowing-down spectrum. The energy dependence of capture cross sections is studied by time-of-flight experiments. The thick target pulsed Van de Graaff source may also be included in this category.

The unique advantage of the reactor source lies in the high neutron dose available for internal target irradiation. For measurements where

thermal or energy integral cross sections are sufficient, reactor irradiation and subsequent analysis by mass spectrographic or radiation counting techniques provide the only feasible means of measurement on highly radioactive or low abundance samples. In addition the reactor-chopper combination appears attractive for total cross section determinations on small samples, for energies up to a few hundred eV, according to an analysis by Michaudon.[3] In most applications, however, higher intensities and resolutions are available for the accelerator-based sources, and the bulk of present day experiments are being carried out with these. The electron linear accelerator is especially effective in the "resonance region" 10 eV to $\sim$ 100 keV, but is limited by the gamma flash at energies above that range.

At energies of a few tens of keV to several hundred keV, the pulsed Van de Graaff offers lower backgrounds. Most of the present day capture measurements are being carried out with these two accelerator sources.[4]

One important category which can be included with the above sources is nuclear explosions. Offering an intensity advantage over conventional sources of 10^{10}, the nuclear explosion is adaptable to measurements on small, highly radioactive samples.[5]

(b) Discrete energy sources from nuclear reactions and their respective energy are summarized in the following table, taken from reference 4.

Table I

Source		Energy	Energy Spread
Photoneutron	Sb-Be (γ,n)	24 keV	$\sim$ 2 keV
	Th-D_2O (γ,n)	195 keV	$\sim$ 2 keV
Threshold	$^{7}Li(p,n)^{7}Be$	30 keV	$\sim$ 15 keV
	$^{3}H(p,n)^{3}He$	65 keV	$\sim$ 15 keV
Discrete	$^{7}Li(p,n)^{7}Be$	5 - 100 keV	$\sim$ 2 keV
	$^{51}V(p,n)^{51}Cr$	5 - 100 keV	$\sim$ 2 keV
	$^{3}H(d,n)^{4}He$	14 MeV	

A significant advantage of the photoneutron sources is their accurately-known strengths as determined by the highly accurate $MnSO_4$ bath activation technique. A careful application of the latter method can yield a source calibration accurate to $\pm$ ½%.[6] Thus the Sb-Be photoneutron source provides an accurate 24 keV calibration point.

The strength of the lithium, tritium, and vanadium (p,n) reaction neutrons can be determined by the methods of associated particle detection, or associated radioactivity detection. The associated radioactivity method is capable of $\pm$ 2% accuracy, while with the 14 MeV neutrons from $^{3}H(d,n)^{4}He$ reaction, 1% has been achieved by measuring the associated particle.[6]

It appears, therefore, that highly accurate source calibration points for capture cross section measurements exist throughout the energy of interest. These complement the more accurately known 0.2% to 0.3% standard thermal cross sections for capture in ^{3}He, ^{10}B, and ^{197}Au.

Detectors

There are several modes of detection of capture processes. Perhaps the simplest is the detection of the radioactivity of the residual nucleus. Although capable of high intrinsic accuracy, the method is limited to discrete energy sources and to those nuclides which possess residual radioactivity. Its chief virtue then is in the measurement of highly precise source calibrations, as in the $MnSO_4$ bath technique.

Methods suitable for use in time-of-flight capture measurements with "white" spectrum sources are those which detect the prompt gamma-ray emission following radiative capture. The two commonly-used detectors are the large liquid scintillator capture tank and the so-called "Moxon-Rae" detector and its variants. Both detectors have suitably fast response

times in the nanosecond range, and both can be made relatively independent of the specific nature of the gamma-ray cascade to the residual nucleus ground state.

(a) Capture tanks are generally made large in volume, typically 1000 liters or more, in order that quanta with energies of up to 8 MeV may be converted in the tank. The efficiency of such a tank, for example, as developed by the General Atomic group,[7] is typically 95 ± 5% when a correction for low energy quanta below ~ 3 MeV is made. Using a boron-poisoned scintillator, the error in the extrapolation procedure used to include low energy events may be held to approximately 3%, and the overall error in the detection efficiency of about 6%. Because of the large scintillator volume needed, the ambient background in the tank presents difficulties for samples with cross sections below ~ 100 mb. Furthermore, accelerator-induced fast neutron and gamma-ray backgrounds become important with electron linac time-of-flight measurements using the capture tank above 100 keV. With the pulsed Van de Graaff techniques, such as employed at Karlsruhe, the background problems are less severe, and cross sections of 5% accuracy have been reported at 30 keV.[8]

(b) Moxon-Rae detectors[9] depend on the conversion of gamma quanta to electrons in a graphite or plastic converter and subsequent detection of these electrons in a plastic scintillator. If the converter is thicker than the range of the most energetic electron it can be shown that the efficiency for detection of a single quantum is proportional to the quantum energy. Thus the efficiency for detection of a neutron capture γ-ray cascade is dependent only on the total energy of the cascade, which is essentially the neutron binding energy. The low efficiency, ~ 2.5%, of this detector is compensated by its low background, fast response, and

insensitivity to neutrons. The efficiency dependence on the neutron binding energy results in additional error when cross section ratios measured with respect to a standard are made.

Recently the efficiency of the Moxon-Rae detector has been increased to about 10% by the Geel group,[10] who use a sequence of Moxon-Rae detectors in series . Furthermore, it has been suggested by Maier-Leibnitz and verified by Macklin and Gibbons,[11] that application of a suitable weighting function to the pulse output of any detector can convert that detector to the Moxon-Rae type. Thus, even the high efficiency of a NaI crystal can be utilized in this manner.

(c) Special detection methods are applied in the instance where fission competes with capture. These methods consist of placing the sample in a fission chamber inside the scintillator tank. The fission events are put in anti-coincidence with detected events in the tank, so that α, the ratio of capture to fission may be inferred. Unfortunately, α-particle pile-up is significant, especially for example in the case of U^{233}, so that extremely thin samples must be used. Diven[12] has used a method which is applicable where fast timing is not important. In this method the fast fission neutrons are moderated and captured by a cadmium or gadolinium poison placed in the liquid scintillator. Delayed coincidences between the resulting capture gamma rays, and the prompt fission gamma rays signal a fission event. The α pile-up is eliminated and thicker samples may be used.

(d) The slowing down time spectrometer has been applied for broad resolution measurements in the range $\sim$eV to $\sim$ 10 keV and is effective for low cross section materials.

To estimate the precision available with these methods, we recall that the relevant quantities are (a) incident flux, (b) number of sample nuclei, and (c) detector efficiency. In most measurements, however, only the energy dependence of the incident flux is measured and knowledge of the detector efficiency is bypassed by using a standard cross section, or using the "saturated resonance" technique. The latter is practical only at low neutron energies where $\Gamma_\gamma \gg \Gamma_n$, and thus the attainable precision is limited by a knowledge of the incident flux energy dependence, the cross section standard, and various self-shielding and multiple scattering effects associated with the physical sample being measured.

For measurements up to $\sim$ 100 keV, the $^{10}B(n,\alpha\gamma)$ reaction shows no evidence of departure from 1/v, and is commonly used as a flux monitor. Above 100 keV, however, uncertainty exists as to the behavior of the $n\alpha_0/n\alpha_1$ branching ratio, and this uncertainty limits the accuracy of a boron gamma counter to $\pm$ 20% above 100 keV.[6]

Thus the limiting accuracies below 100 keV are set by the knowledge of the standard cross sections and by sample effects. The most commonly used and most carefully measured capture cross section is $^{197}Au(n,\gamma)^{198}Au$. Some idea of the difficulties in obtaining agreement between different measurements of this standard may be gained by examining the measurements recorded on the SCISRS I tape for the 30 keV capture cross section of gold (Table II).

Table II

Cross Section	Method	Date
0.515 b	Scintillator tank, referred to indium	April 1961
1.129	Activation, referred to boron	May 1961
0.809	Activation, referred to boron	May 1961
0.947	Activation, referred to U235	December 1962
0.513	TOF, Moxon-Rae	1963
0.565	TOF, scintillator tank	1964
0.598	Activation, referred to $MnSO_4$ bath	October 1966

The consistent discrepancies between the shell transmission measurements at 24 keV and the capture tank measurements at 30 keV have been explained in terms of large multiple scattering corrections in the shell measurements.[13] After such corrections are made, the results are consistent with recent pulsed Van de Graaff measurements of Pönitz, [8] et al. Pönitz has summarized the recent measurements and finds for the best value:

$$\sigma_{n,\gamma}(E_n = 30 \text{ keV}) = 0.596 \pm 0.012 \text{ b}$$

with reasonable agreement between shell transmission, capture tank, and Sb-Be activity measurements. Good agreement in the absolute gold capture measurements of Pönitz and Harris[14] is found up to 100 keV, but there is a 15% discrepancy above 100 keV with measurements of Barry[15] and Grench,[16] which are based on ^{235}U fission cross sections. There appears to be, therefore, some question about either the accuracy of the $^{197}Au(n,\gamma)$ cross sections (or the ^{235}U fission cross sections) above 100 keV, while below 100 keV, the gold capture cross section is known to 5% or better.

The question of sample corrections due to multiple scattering and self shielding effects is in an unsatisfactory state at the present time. These corrections are aggravated by the large cross section fluctuations due to the neutron resonance structure. The lack of agreement between multiple scattering codes has been underscored by S. Friesenhahn,[17] in the lack of agreement between various laboratories on the sodium 2.85 keV resonance radiation width. In this resonance multiple scattering is very important, since $\Gamma_\gamma/\Gamma_n \approx 10^{-3}$. A significant difference between the GA and RPI multiple scattering corrections exists, leading to a deduced radiation width of 0.47 ± 0.05 eV (GA calculation) vs 0.60 ± 0.06 eV (RPI calculation) for the same set of data. Clearly

it would be valuable to have an inter-laboratory exchange and evaluation of multiple scattering correction codes.

To emphasize this point, we quote some recent pairs of values from recent Γ_γ determinations at the RPI Linac[18]. In Table III are selected values of radiation widths for resonances of widely-differing neutron widths.

TABLE III

Isotope	E_o(keV)	Γ_n (eV)	Γ_γ (eV)	
23 Na	2.85	410	0.61 ± .06	(g = 3/8)
	52.2	700	2.60	(g = 3/8)
27 Al	5.88	20	0.41	(g = 7/12)
	35.0	1700	2.8	(g = 7/12)
56 Fe	1.15	0.088	0.6	
	27.7	1670	1.44 ± 0.14	
57 Fe	3.96	177	1.14 ± 0.10	
	6.21	396	1.32 ± 0.12	
	29	3000	4 ± 1	

The comparison in every instance shows a high radiation width correlated with high neutron width, a result which is suggestive of the presence of systematic error when $\Gamma_n \gg \Gamma_\gamma$.

In the lower energy region < 10 keV, where the neutron widths are not excessively high, an examination of recently published radiation widths in ^{91}Zr[19] and ^{nat}Mo[20] suggests that typical errors are in the order of 10% at best, where $\Gamma_n > \Gamma_\gamma$. The details of resonance analysis using capture data are too complicated to discuss here. It suffices to

say that a combination of transmission, capture, and scattering measurements are necessary to deduce radiation widths in the general case, for an arbitrary rate of capture to scattering. Precisions of better than 10% on radiation widths have seldom been achieved.

In the light of the above discussion, several comments can be made concerning the general problems in fulfilling the requests of Wash 1078. In general, requests demanding precisions of better than 5% in the 10-100 keV region are presently incapable of fulfillment, since the best known standard, gold, is known no better than 5%. These requests include, for the light to medium A, (#57), F and (#108), ^{59}Co (a one per cent radiation width measurement is quite beyond present capabilities). The case of the zirconium requests present special problems because of the large ratio of scattering to capture and the low capture cross sections. Recent measurements on ^{91}Zr (n,γ) performed up to energies of 4 keV claim accuracies of $\sim$ 25% in radiation widths, and from 7 to $\sim$ 40% in neutron widths[19]. These are illustrative of presently available accuracies and indicate that the precisions requested in (#90)Mn, (#118)^{63}Cu, (#119)^{65}Cu, (#124)Zr, and (#152) Nb cannot be satisfied. The ^{51}V thermal cross section presently known to 6% should be capable of being measured to 3% as requested in #85.

In the heavier nuclei, in fission product poisons, several requests call for 2-5% in the low energy range 0-1 eV, e.g. (#197)^{151}Eu and (#198)^{153}Eu. These accuracies are consistent with known thermal cross section standards (e.g. ^{197}Au) and are therefore attainable. On the other hand, (#216)^{169}Tm; (#225)^{177}Hf; (#226)^{178}Hf and (#227)^{179}Hf require accuracies of 3-5% in resonance parameters and capture cross sections up to 5 keV; these are not attainable. The resonance integral request

of 5% accuracy for thorium (#243) in the range up to 2 keV is within present capabilities, since precise knowledge of the thorium radiation width is not needed here (however, there are significant and unexplainable discrepancies in the neutron widths for the low energy resonances). For the range 2 keV to 1 MeV, the 5% request (#244) seems beyond attainment due to the present $\sim$ 10% knowledge of the thorium radiation widths. This situation results from the observation that when $\Gamma_n < \Gamma_\gamma$, then the neutron width is the controlling factor, whereas at higher energies, where $\Gamma_n > \Gamma_\gamma$, the radiation width uncertainty becomes dominant. In the case of ^{197}Au, request #235 for 1% in the capture integral, however, seems to be outside present capabilities of differential cross-section measurements, since it would require measurements of the neutron width of the 5 eV level to an accuracy of 1%.

Requests calling for measurements on fission product poisons in the A range from $\sim$ 90 to $\sim$ 150 require samples not presently available to the experimenter. Several of these have short half-lives; e.g. 36 hr ^{105}Rh (#163), 78 hr ^{132}Te (#167), 21 hr ^{133}I (#168), 16 m ^{135}Xe (#171) and 9.2 hr ^{135}Xe (#172). Thermal cross sections for radioactive nuclei may be obtained by the source depletion method, i.e. measurement of activity changes after irradiation in a thermal column. The cross section variation up to $\sim$ 1 eV for these requests, however, requires external methods which need sample amounts which are not presently available. If the capture cross sections are large compared to scattering cross sections, $\sigma_{n\gamma} \gg \sigma_s$, total cross section measurements on these small, highly active, samples may be best satisfied with reactor-based transmission

measuring devices, either a diffraction spectrometer or a chopper.

Recently Pönitz, using the gray-detector, liquid scintillator technique, has performed a careful absolute measurement of the ^{238}U capture cross section (request #310), 20 keV up to 500 keV, with reported accuracy of 5 to 9%[8]. The discrepancies with older measurements, however, are somewhat larger, and it is not yet clear whether the request is satisfied. The call for 3% to 10 MeV, request #309, is clearly unrealistic.

The fissile and transplutonium materials present slightly different problems. The scope of the present report limits detailed discussion of fission measurements involved in the determinations of α and η. In general, it can be observed that (n,γ) experiments carried out in a background of high material activity can best be handled with the use of nuclear explosions, in which a high dose of neutrons is absorbed by the sample. The rate at which such experiments can be carried out is somewhat uncertain, depending both on sample availability and experimental test frequency.

Only total cross sections are feasible for such samples as ^{242}Cm and ^{244}Cm. Request (#322) ^{238}Np (2.1 days half-life); (#402) ^{251}Cf; and (#404) ^{252}Cf do not appear feasible because of sample unavailability[21].

V. The Techniques for Measurement of Differential Gamma-ray Production Cross Sections

Information on the characteristics of gamma-ray production by neutron bombardment of various elements is a necessity for shielding considerations in the proper design of reactors and other devices which product high-neutron fluxes. Of particular interest are energy spectra, angular distributions and, when available, cross sections differential in angle and γ-ray energy for a range of neutron energies. In the range of incident energies of interest to most nuclear technologists, gamma-ray production cross sections contain contributions from neutron capture and inelastic neutron scattering. Although the capture cross section of a typical nucleus decreases from about 100 mb at 1 MeV to 10 mb at 10 MeV, the inelastic neutron scattering cross section rises quite rapidly above threshold so that typically total gamma-ray production cross sections are in the order of barns. From considerations given below it will be evident that differential γ-ray spectra associated with capture of this magnitude can be readily observed with existing experimental techniques over the full range of energies of interest. Indeed, measurements of the differential γ-ray spectra both in laboratory angle and energy have been in progress for some time for neutron energies below 7 MeV. Bergqvist and Starfelt[22] in Sweden, I. L. Morgan and collaborators at Texas Nuclear Corporation[23] and Conde, Drake and Hopkins[24] at Los Alamos have been engaged in extensive programs of such measurements in the MeV region. In addition, many studies of inelastic neutron scattering have been reported in which the scattering is detected by observation of the γ-ray spectrum. Such experiments[25] frequently furnish γ-ray cross-section information which

is very useful in fulfilling cross-section requests. In the resonance region (0-100 keV) the study of capture spectra is being actively pursued in many laboratories using reactors or accelerators as sources. In the discussion which follows we will assess the capability for fulfilling the requests for gamma-ray production cross sections in Wash 1078 in light of this current activity and the general knowledge of the state of the art in such measurements. In particular we will be concerned with extending measurements to cover the full range of incident neutron energies appearing in the cross-section requests.

The cross section of interest here is the laboratory cross section for the production of γ-rays which is differential both in γ-ray energy and emission angle. Frequently, however, the request calls for the angular dependence only if the distribution is significantly anisotropic. In addition many requests require only the measurement of the spectrum at one specified angle. Nevertheless all measurements involve basically measurement of the double differential cross section as a function of neutron energy with specified resolution. We have summarized in Table IV some of the basic requirements of all priority-I requests for differential gamma-ray data which are tabulated in Wash 1078. Frequently where there are several requests concerning the same target we have summarized the overall demands. The information displayed includes target (natural element unless indicated), incident neutron energy region for which cross sections are desired, incident neutron energy and γ-ray resolutions, and overall precision with which the cross section is to be determined. It should be noted that although only priority-I requests are considered, the features are typical of requests of any priority. We shall attempt

to determine experimental approaches to fulfilling these requests which are (1) feasible in principle and (2) realizable with existing facilities. We shall consider these requests from the point of view of neutron sources, sample material for targets, and γ-ray detection system.

Measurements are requested essentially from the low-energy resonance region to 16 MeV. From point of view of neutron sources, any effort to fulfill requests would probably involve the use of 4 modes of neutron production:

(1)	0-100 keV	Reactor or accelerator produced neutrons
(2)	30 keV - 1 MeV	Neutrons produced by $^{7}Li(p,n)Be^{7}$ reaction
(3)	1-10 MeV	Neutrons produced by $T(p,n)^{3}He$ reaction
(4)	5-16 MeV	Neutrons produced by $D(d,n)^{3}H$ reaction

The first two modes of production have been exploited widely over the last decade and exploit well-known techniques. The $T(p,n)^{3}He$ is currently being used by many Van de Graaff laboratories and in particular at LASL, work is being carried out on γ-ray production measurements in the MeV range using this reaction. By contrast the effective use of the $D(d,n)^{3}H$ will require a considerable developmental effort before it will be useful over the full 5-16 MeV neutron range. The reaction is presently being successfully applied at lower energies ($\lesssim$7.5 MeV), but at higher energies tertiary reactions will compete with the two-body reactions. Consequently in the usual mode of operation, in which a thin target of deuterium is bombarded with deuterons of known energy to produce neutrons whose energy is uniquely related to angle of emission, a background of neutrons produced by tertiary reactions will be present. Present evidence is that these reactions, $D + d \rightarrow 2p + 2n$ or $D + d \rightarrow p + n + {}^{2}H$, produce a

neutron continuum whose maximum energy lies considerably below the energy corresponding to two-body production. Thus, the use of time-of-flight techniques in conjunction with pulsed beam operation of a tandem Van de Graaff will permit discrimination against the continuum. The point to be emphasized is that a successful program of measurements for cross-section requests will require a considerable effort to determine precisely the characteristics and effects of tertiary reactions and development of techniques to discriminate against them.

The resolutions in neutron energy specified in the requests and summarized in Table IV are not at all demanding in terms of the state of the art. In this respect, no problem should arise in adopting sources to these measurements. As we shall see below, the relatively poor resolution specified also permits the use of considerably more intense neutron beams than is usual.

In regard to sample materials for all the priority-I requests with the exception of ^{235}U, ^{238}U, and ^{239}Pu, targets of elements in their natural abundance are requested. Thus, no serious problems are anticipated in obtaining suitable targets.

An inspection of Table IV indicates that γ-ray detector resolution of approximately 250 keV at high energies and approximately 10% at low energies is needed to satisfy all cross-section requests. Although a major part of neutron γ-ray spectroscopy is presently being performed with Ge(Li) detectors, the older Raboy-Trail NaI(Tl) spectrometer in which a moderate-size central crystal is surrounded by a concentric annulus still appears suitable for these measurements. The simple Ge-diode detectors can be very useful particularly in cases in which the spectrum

is dominated by a few low-energy lines. However, spectra are, in general, complex with contributions over a broad range of energies. Under these circumstances a useful detector must have a well-defined and precisely known response function, i.e., both the total efficiency and the form of the response function must be known. The response should also be free of broad tails or continua which complicate the unfolding of contributions of various γ rays. This type of difficulty has produced considerably uncertainty in attempts to extract a γ-ray spectrum from a pulse height distribution. Thus, it appears the problem of the Compton continuum will limit the usefulness of simple Ge(Li) detectors in the near future to studies of inelastic neutron scattering and other special cases in which the excitation of a few specific states is of primary interest.

A version of the Ge(Li) detector which produces a superior line shape, consisting of a single very sharp peak on a very weak continuum, is the pair-spectrometer configuration in which the crystal is placed inside a four-sector NaI annulus. Recent measurements made at Texas Nuclear indicate that this arrangement has close to the ideal response function for γ-ray production measurements. At the present time, the very low detection efficiency limits its usefulness to measurements of moderately high cross sections.

Thus, for three reasons, the best overall detector appears at present to be the Raboy-Trail type. First, with a NaI central crystal, the absolute detection efficiency can be readily determined to within 5% over the full range of energies of interest in γ-ray production measurements. Second, for measurements at higher incident neutron energies, where background

problems are particularly severe, the system has the added advantage that the annulus shields the central crystal from background radiation as well as suppressing events in which radiation escapes from the central peak. Finally, the system can readily be converted for high-resolution work by replacing the central crystal with a Ge(Li) detector.

In regard to this last point, a useful compromise between Ge(Li) and NaI detectors is frequently possible; namely, a measurement of a γ-ray production spectrum at one angle with the very high energy resolution of the Ge(Li) pair spectrometer, combined with a subsequent measurement of the production spectrum as a function of angle using the lower-resolution Raboy-Trail system.

The requirements concerning neutron sources, target samples, and detection systems are satisfied by the characteristics of existing installations in which γ-ray production cross sections or related quantities are measured. These include the Van de Graaff installation at Los Alamos used by Hopkins and Drake, that at Texas Nuclear Corp. described in the work of Morgan and co-workers, and the similar arrangement used in Sweden by Starfelt and Bergqvist. These installations consist of Van de Graaff generators which furnish pulsed beams of microamps of protons and deuterons which are used to bombard suitable neutron-generating targets. The sample under study is placed downstream and generally an NaI detector is placed 1 meter from the sample. A crystal of large volume or the Raboy-Trail type is used to detect γ rays. Neutron background and γ rays are separated by employing nanosecond time-of-flight techniques to differentiate corresponding peaks in the time spectra. The principle difficulty at present is the lack of similar

facilities for γ-ray production measurements which use higher energy proton machines to generate neutrons in the 7-15 MeV range. It is advisable that substantial support be directed in the near future to utilization of tandem Van De Graaffs for gamma-ray measurements in this domain of energies.

Having determined in broad outline the relevant experimental techniques one can now access the limitations and precision consistent with the "state of the art". A perusal of Wash-1078 indicates that the precisions generally requested are not unreasonable. Of course one must observe, particularly at higher neutron energies, that such elusive uncertainties as neutron background levels may alter our conclusion. However, consider the most stringent requirement of a 10 microbarn/steradian MeV upper limit on a spectral measurement which is requested at a single laboratory angle of 55^{o}. If we assume an isotropic angular distribution and an energy distribution flat in energy from 0-10 MeV, the total production cross section is 1 mb. If one uses an experimental arrangement similar to that of the Los Alamos group (with a target 250 keV thick) a meaningful measurement would require one day of running. Therefore, from the point of view of hardware, experimental sensitivities are adequate to cope with the present cross-section demands. On the other hand, limitations imposed by background levels are very much more difficult to assess and one must consider the individual experimental situations before drawing any conclusions. At best, such considerations are uncertain. Little work has been carried out at the higher neutron energies where shielding problems for scattered neutrons become formidable, but the consensus of most experimenters is that one cannot use the maximum

neutron yield at his disposal. In fact, knowledgeable experimenters indicate that a realistic upper limit on a cross-section measurement would more likely be 100 microbarns/steradian MeV. In the absence of additional information, we suggest this figure as a reasonable estimate of experimental sensitivity for measurements at the present time. Clearly, a great deal of effort must be devoted to developing new experimental facilities and techniques with sufficient sensitivities at all energies to fulfill the demands of the cross-section requests.

In contrast to the question of sensitivity, the precision requested in the measurements is relatively low--10%--and there should be little difficulty in meeting this requirement. The only respect in which requests are generally incompatible with present experimental capabilities is the range of angles over which angular distributions are generally required. Frequently, points at 5 and 10 degrees are indicated and clearly such measurements are not feasible, nor are they likely to be needed (see p. 5). An obvious solution is an extrapolation of the measured angular distribution to 0°. Such an alternative should be seriously considered before requests for small angle measurements are included in the compilation.

In summary, the technology for γ-ray cross-section measurements appears adequate for fulfilling the gamma-ray differential cross sections for all regions of interest with the possible exception of measurements at small angles relative to the incident beam. However, for higher neutron energies exploratory programs will no doubt be necessary for perfecting neutron sources and supressing neutron backgrounds probably through application of time-of-flight techniques.

The following remarks concern specific requests in Wash 1078.

Requests 40, 41, 42, 50, 65, 71, and 102 all require measurement at lab angles of less than 10^{o}, which clearly is not feasible. These needs should be re-evaluated giving consideration to extrapolation of measured differential cross sections at larger angles to cover the small angle region.

Request 114 contains no indication of what resolution in neutron energy is desired.

Request 100, 125, and 245 are not clear as to resolution desired. Does requestor wish 250 $\pm$ 250 keV at the lowest energy? Neutron resolution of .5 MeV is specified from the eV to the 16 MeV region.

Request 171 is probably not feasible at the present time since no appropriate Xe^{135} target is readily available. The lifetime of this nucleus is only 9.2 hr.

TABLE IV

Target	Neutron Energy Range	ΔE_n	ΔE_γ	Precision
Be	150 eV - 15 MeV	10% (150 eV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	10% (50 keV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	30 - 40 % absolute
N	[illegible] - 16 MeV	.25 MeV	.25 MeV	10%
O	10 - 15 MeV	.25 MeV	.25 MeV	10%
Na	1 - 14 MeV	none	none	10%
Al	1 - 16 MeV	.25 MeV	.25 MeV	10%
Si	3 - 16 MeV	.25 MeV	.25 MeV	10%
Fe	Ev - 10 MeV	.25 MeV	.25 MeV	10%
Ni	0 - 175 keV	10%	.5 MeV	10%
Zr	Ev - 10 MeV	.5 MeV	.5 MeV	?
W	th. - 15 MeV	10% (150 eV - 5 MeV) .5 to 1.0 MeV (5 - 15 MeV)	10% (50 keV - 5 MeV) .5 to 1.0 MeV (5 - 15 MeV)	20%
^{235}U	150 eV - 15 MeV	10% (150 - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	10% (50 keV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	30 - 40%
^{238}U	150 eV - 15 MeV	10% (150 eV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	10% (50 keV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	30 - 40%
^{239}Pu	150 eV - 15 MeV	10% (150 eV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	10% (50 keV - 5 MeV) .5 - 1.0 MeV (5 - 15 MeV)	30 - 40%

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