55
3/6/79

44. 2302

LA-7617-PR
Progress Report

MASTER

NDA Technology for Uranium
Resource Evaluation
January 1—June 30, 1978

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

CLASSIFIED

UNCLASSIFIED

The four most recent reports in this series, unclassified, are LA-6840-PR, LA-6996-PR, LA-7139-PR, and LA-7251-PR.

This work was supported by the US Department of Energy, Division of Uranium Resources and Enrichment.

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.

NDA TECHNOLOGY FOR URANIUM RESOURCE EVALUATION

Compiled by

M. L. Evans

ABSTRACT

This progress report describes work performed during the time period from January 1, 1978 to June 30, 1978, on the contract for NDA Technology for Uranium Resource Evaluation in Group Q-1. The work has focused on gamma-ray calculations and computer code modifications in support of borehole logging measurements and surface gamma-ray surveys. Experimental effort has concentrated on improvements in uranium fission neutron detection efficiency.

The Phase II calculations deal with gamma-ray surface and airborne surveys. The initial calculations are to determine the KUT spectra as a function of position and angle above and below a plane interface. Diagnostic calculations have been performed to independently verify the appropriateness of the model used by ONETRAN in calculating the angular fluxes of Phase II and to determine the optimum values for the model parameters. However, all attempts to eliminate unphysical oscillations in the high-energy angular fluxes by judiciously choosing the model parameters have not been successful. Alternatively, a criterion based on single Compton scattering has been devised by which angular flux values calculated by ONETRAN are accepted if they are physically reasonable but rejected if they are unphysical. The Monte Carlo code MCNG has been used to calculate angle-integrated currents for comparison with those obtained from ONETRAN. The ONETRAN direct currents agree very well with those of MCNG for both high (for example, 2.655-MeV) and low (for example, 0.46-MeV) photon energies. The sky-shine currents of ONETRAN

agree well with those of MCNG at low energy but are inaccurate at high energy due to the inadequacy of the ONETRAN P_n approximation in describing the photon scattering at high energies.

Development continues on the Monte Carlo computer code GAMRES, which computes NaI detector response functions. The code has been optimized with respect to execution speed by streamlining the root-solving algorithm used to determine electron and photon energies and scattering angles, and by appropriately choosing the convergence criteria for each of the simulated photon interactions. A scheme for generating and testing the response function map has been devised. The computer program PHSURF, which performs two-dimensional spline interpolation on the energy vs pulse-height map generated by GAMRES, has been modified to provide both quantitative and qualitative checks of the interpolated energy vs pulse-height surface.

An alternative approach to solving the disequilibrium problem in KUT logging is being investigated. The scheme involves the use of a probe containing a gamma-ray source, gamma-ray shielding, and a NaI or perhaps Ge(Li) detector. Photons from the probe source irradiate the borehole formation, inducing X-ray fluorescence from nearby uranium deposits, which is measured by the probe detector. Initial results from a Monte Carlo simulation of the technique look promising.

Development of the prototype photoneutron logging probe has focused primarily on improvements in the efficiency of detection of the uranium fission neutron signature. The fundamental difficulty in this area is achieving optimum neutron detection efficiency while operating in the high gamma-ray flux produced by the photoneutron source. Two areas of detector technology are being explored in this development effort: 1) recoil proportional counters and 2) liquid scintillator detectors.

In the first category, methane (CH_4) recoil proportional counters are being evaluated as a potential replacement for ^4He counters. Initial tests conducted with an unmoderated fission neutron source in a 1 R/h gamma-ray background indicate that significant improvement in detection efficiency can be achieved with

high-pressure CH₄ detectors. Additional measurements in a borehole mockup to determine the effects of neutron spectral softening on these conclusions are in progress.

The development of a liquid scintillator detector for the prototype photoneutron logging probe offers the potential for a large increase in fission neutron detection efficiency. Unfortunately, commercially available detectors are not designed to meet the stringent space requirements imposed by the borehole environment. Thus, a liquid scintillator detector appropriate to the prototype photoneutron logging probe has been designed and constructed at LASL. The initial tests indicate that the detector is exhibiting the proper sensitivity to incident radiation. Further measurements of the effectiveness of the shielding and gamma-ray discrimination are in progress.

I. GAMMA-RAY SPECTRAL CALCULATIONS

A. Phase II Calculations

(M. L. Evans and M. Jain)

The Phase II calculations deal with the gamma-ray fluxes of surface and airborne gamma-ray surveys for uranium ore. The calculation of angular flux values for the geometry of Phase II (see Fig. 1) is very difficult owing to the rock-air interface and the absence of sources in the air medium.¹⁻⁴ In particular, unphysical oscillations in the angular fluxes for P_n orders greater than one and gamma-ray energies greater than about 1 MeV have persisted despite many attempts to eliminate them.

Studies were performed to determine if the inclusion of a very weak sky-shine source such as cosmic rays would be sufficient to eliminate the oscillations. Inclusion of a sky-shine source in the air results in angular fluxes that are more isotropic, and thus, in fluxes that are more likely to be accurately calculated using a low-order cross-section expansion (that is, low P_n order). Initial results looked promising, but further study indicated that the oscillations could be significantly reduced in

Fig. 1.
Geometry used in the Phase II
calculations.

amplitude only by the inclusion of a relatively large sky-shine source; a source that also resulted in significant distortions in the sky-shine fluxes that did not suffer from the oscillations.

Various numerical quadrature techniques were also studied in an attempt to eliminate the oscillations. In performing numerical integration of a rapidly varying function, unphysical oscillations can result in some regions of the integration interval depending on what quadrature scheme (that

is, Gaussian, Chebychev, Lobatto, etc.) is being used and on how the quadrature points are chosen. Quadrature oscillations can sometimes be overcome by refining the quadrature mesh in the region exhibiting the oscillations. If the mesh refinement technique fails, then typically, the function is integrated only over the region for which the integration scheme yields accurate results. The region containing the oscillations is ignored as being nonintegrable if the contribution from that region is estimated to be small.

Figure 2 summarizes the results of applying three different integration schemes in ONETRAN to obtain the angular fluxes for the Phase II geometry. All data were taken at the rock-air interface ($z = 0$) in the energy bin 2.51 - 2.61 MeV. A single gamma ray having an energy of 2.56 MeV was used as the source in the 26-group calculations. The P_n order of the photon cross-section expansions was kept constant at five. The quadrature schemes utilized different quadrature points as shown in Fig. 2. The 96-point double Gaussian (DP) quadrature employed 48 points over the interval $-1 \leq \mu \leq 0$ and resulted in a very fine mesh

structure. The 12-point quad Gaussian technique involved a partitioning of the interval $-1 \leq \mu \leq +1$ into four equal quadrature regions (instead of two regions as for double Gaussian integration) over which separate integrations were performed. The 12-point double Chebychev quadrature used 6-point Chebychev integration over each of the intervals $-1 \leq \mu \leq 0$ and $0 \leq \mu \leq +1$.

Figure 2 illustrates very clearly that the oscillations are not amenable to elimination by the available choices of numerical quadrature schemes. Angular fluxes from the three schemes fall along the same curve and all schemes result in the same unphysical oscillations. The magnitude and shape of the oscillations appear to depend only on the P_n order of the cross-section expansions as shown in Fig. 3. As the P_n order increases, the ONETRAN angular fluxes become a more accurate estimate of the true sky-shine fluxes, but the unphysical oscillations become more rapidly varying, not unlike the Gibbs phenomenon⁵ in performing a Fourier series approximation to a step function. In both instances, unphysical oscillations occur when an insufficient number of terms is used to approximate a rapidly varying or discontinuous function.

The sky-shine flux in the energy bin 2.65-2.66 MeV shown in Fig. 3 falls off very rapidly with increasingly negative values of μ because of the geometry of Phase II. The energy bin can receive flux contributions only from source gamma rays (having energies in the range 2.65-2.66 MeV) that originate in the rock medium near the interface, travel nearly parallel to the interface unattenuated, and then suffer an interaction that scatters the gamma ray back toward the rock medium (that is, in the sky-shine direction: $-1 \leq \mu \leq 0$). If the angle of scattering is too large, the scattered gamma ray will have an energy that is below the energy bin 2.65-2.66 MeV and will contribute no flux to the bin. Figure 4 shows the limiting value of μ for gamma rays having energy E_γ starting in the rock medium and suffering only single Compton scattering. The curves of Fig. 4 are parameterized by the energy bin width ΔE_γ used to bin the sky-shine flux. The curves indicate that as the energy of the source gamma ray

Fig. 2.

Phase II sky-shine angular fluxes obtained using three different ONETRAN angular integration schemes. A monoenergetic 2.56-MeV gamma-ray source and a cross-section expansion order P_5 were used for all three cases. The quad Gaussian and double Chebychev integrations each employed twelve quadrature points (S_{12}).

Fig. 3.

Phase II sky-shine angular fluxes at $Z=0$ obtained using double Gaussian integration, S_8 quadrature, a monoenergetic 2.655-MeV gamma-ray source, and cross-section Legendre expansions of order 0 to 4.

Fig. 4.

The limiting value of μ for gamma rays having energy E_γ starting near the interface, suffering only single Compton scattering in the air medium, and yielding a sky-shine gamma ray having an energy in the range E_γ to $E_\gamma - \Delta E_\gamma$.

Fig. 5.

Phase II sky-shine angular fluxes at $Z=0$ obtained from a P_3 , S_8 , double Gaussian integration calculation using a monoenergetic 2.655-MeV gamma-ray source.

decreases, Compton scattering allows the gamma ray to scatter through larger angles while retaining an energy that is still in the source gamma-ray energy bin. More importantly, comparison of the two curves for a constant source energy (for example, $E_\gamma = 2.655$ MeV) shows that as the energy window ΔE_γ (used to bin the sky-shine flux) is widened, the limiting value of μ becomes

more negative (that is, gamma rays scattering through larger angles and having lower energies are contributing to the flux in the bin). These effects are also demonstrated in Fig. 5, which shows the sky-shine flux (in three energy bins) resulting from source gamma rays in the rock medium having energies in the range 2.65-2.66 MeV. The flux in the highest bin (2.65-2.66 MeV) is limited to small negative values of μ because the gamma rays cannot scatter through large angles and still have an energy that falls in the bin. However, source gamma rays starting in the range 2.65-2.66 MeV can scatter through larger angles and fall in the 1.65-1.66 MeV bin. Thus, the sky-shine flux extends to more negative values of μ . Finally, if the source gamma rays are allowed to backscatter, the gamma rays can be emitted from the rock medium directly upward (that is, $\mu = +1$) and can still contribute to the sky-shine flux at $\mu = -1$ for the energy bin 0.15-0.16 MeV. Thus, source gamma rays can be emitted from the rock medium in any direction and can scatter through any angle and still contribute to the sky-shine flux. As a result, the sky-shine angular flux becomes nearly isotropic and there is no limiting angle in μ . Figure 4 indicates that for source gamma rays having lower energies, the limiting values of μ are more negative and the fluxes are more isotropic than those of Fig. 5.

The implication of these results for the P_n order of the cross section expansions is clear. Only extreme forward-scattering of the source gamma rays can contribute to the sky-shine flux at high energies. Thus, accurate fluxes at high energies depend on an accurate representation of the extremely forward-peaked Compton cross section at those energies. Low-order expansions of the Compton cross section are poor representations of the interaction at high energies and result in anomalies in the resulting angular fluxes. At lower gamma-ray energies, the Compton cross section becomes more isotropic and can be represented with fewer terms in the cross-section expansion, resulting in more accurate fluxes at lower energies (this topic will be discussed in more detail later).

In theory, the oscillations in the sky-shine angular fluxes could be eliminated by including a very large number of terms in the cross-section expansions. In practice, the gamma-ray cross sections used by ONETRAN include a maximum of eleven terms (tenth order). Results indicate that even eleven terms are insufficient to accurately represent the scattering at high energies (more will be said about this later).

The practical limits on the P_n order and the inability of various numerical integration schemes to eliminate the oscillations in the sky-shine fluxes suggest that the oscillations cannot be overcome by state-of-the-art numerical techniques and must be ignored instead. This philosophy will prevail in all subsequent work in Phase II. The angular fluxes will be calculated using the highest P_n order available (that is, $P_n = 10$) and the single Compton scattering criterion discussed above will be used to determine at each energy the region of μ for which the fluxes are accurately calculated. That is, the highest source energy for a given spectrum (that is, potassium, uranium, or thorium) will be used with the expression

$$\mu_{\text{lim}} = -\sin \left[\cos^{-1} \left(1 - \frac{m_0 c^2}{E_\gamma} \left\{ \frac{E_\gamma}{E'_\gamma} - 1 \right\} \right) \right] \quad (1)$$

derived from single Compton scattering and the Phase II geometry to determine the limiting value of μ for each energy bin of the spectrum. In the above expression,

- μ_{lim} = limiting value of μ ,
- $m_0 c^2$ = rest mass energy of the electron,
- E_γ = upper edge of the bin containing the most energetic line of the spectrum, and
- E'_γ = lower edge of each of the energy bins of the spectrum.

Thus, for each energy bin of the spectrum, a limiting value of μ will be determined. Angular flux values having μ in the range $\mu_{lim} < \mu \leq 0$ will be accepted as accurate fluxes, but fluxes having μ in the range $-1 \leq \mu \leq \mu_{lim}$ will be set to zero (to eliminate the unphysical oscillations).

Angular fluxes modified by the single Compton scattering criterion are shown in Figs. 6-9. The angular fluxes are those for $z = 0$ cm (the rock-air interface) and are due to source photons started in the rock formation having energies in the range 2.61-2.62 MeV (the same energy as the most energetic line of the thorium spectrum). The fluxes are the result of a P_5 , S_{12} , DP, 265-group ONETRAN calculation.

Fig. 6.
Phase II sky-shine angular fluxes at $z=0$ as a function of gamma-ray energy and angle (that is, $\mu = \cos\theta$). The μ axis projects into the plot and is obscured by the surface.

Fig. 7.
Logarithmic (base 10) plot of the sky-shine angular fluxes of Fig. 6 viewed from the same perspective.

Figure 6 is a linear three-dimensional plot of the angular flux as a function of photon energy and angle (that is, μ , where $\mu = \cos\theta$ and θ is defined in Fig. 1). The fluxes correspond to μ values in the range $-1 \leq \mu \leq 0$, that is, sky-shine fluxes. The energy axis is in the foreground and encompasses the range 0-2.66 MeV with the energy increasing from left to right. The $-\mu$ axis projects into the plot from the perspective of Fig. 6 and extends from 0 to +1. The flux tail at low energies obscures the $-\mu$ axis

Fig. 8.
Logarithmic (base 10) plot of the sky-shine angular fluxes of Fig. 6. The perspective corresponds to a 45° counterclockwise rotation of the plot of Fig. 7. The μ axis (actually $-\mu$ axis) is clearly visible in this perspective.

Fig. 9.
Logarithmic (base 10) plot of the sky-shine angular fluxes of Fig. 6. The perspective results from a 90° clockwise rotation of the plot of Fig. 7. The energy axis is obscured by the surface.

from view in Figs. 6 and 7. The flux axes in Figs. 6-9 are in units of photons/cm²/sr/unit source, that is, standard angular flux units. Figure 6 is a linear plot with the fluxes ranging in value from 0 to about 1 photon/cm²/sr/unit source. The plot indicates that for all values of μ the energy dependence of the flux is about the same. Starting at 0 MeV and moving to higher energies, one can identify certain features. First, there is the usual low-energy tail due to the interplay between the Compton and photoelectric processes. At about 0.2 MeV the Compton back-scatter peak can be observed and at about 0.5 MeV, the pair annihilation peak is a very prominent feature. The flux then decreases slowly with increasing energy to form a very low level plateau. There appears to be very little dependence of the angular flux on μ , except perhaps near 2.6 MeV, where the plateau decreases with more negative values of μ (that is, more positive values of $-\mu$). This is due, of course, to the fact that the fluxes above the annihilation peak in energy are very small and are difficult to discern on the linear plot.

Logarithmic (base 10) plots of the angular fluxes of Fig. 6 are shown in Figs. 7-9, where an artificial base has been given to the plots. The flux axes have the range -4 to 0 and any flux values that are smaller than 4×10^{-4} photons/cm²/sr/unit source (that is, about -3.4 on the log scale) have been set to that value. This technique permits the plotting of those regions of Fig. 6 wherein the angular flux values are zero.

The perspective of Fig. 7 is identical to that of Fig. 6. The same energy-dependent features are observable in both plots. However, the dependence on μ of the angular fluxes is much more evident in Fig. 7. The perspective of Fig. 8 corresponds to a 45° counterclockwise rotation of the plot of Fig. 7. In Fig. 8, both the energy and $-\mu$ axes are clearly visible. The perspective of Fig. 9 resulted from a 90° clockwise rotation of the plot of Fig. 7. In Fig. 9, the energy axis is obscured by the flux values and the $-\mu$ axis is in the foreground. The origin of the plot is also obscured but lies below the label "FLUX", deep into the plot. The energy values increase from 0 MeV at the origin (deep

into the plot) to 2.66 MeV in the foreground. This perspective best illustrates both the rapid decrease in the angular flux with more negative μ values (more positive $-\mu$ values), and the effect of the application of the single Compton criterion to the angular fluxes. The angular fluxes decrease rapidly with increasingly negative values of μ for all energies greater than about 0.5 MeV. The decrease is especially rapid for energies greater than about 2 MeV. The P_5 representation of the scattering cross sections is not sufficient to accurately describe extreme forward scattering at these energies, resulting in angular fluxes that do not fall off as rapidly as they should for increasingly negative values of μ . Consequently, the single Compton criterion truncates the angular fluxes for a given energy for values of μ more negative than the limiting value of μ at that energy. This truncation is clearly evident for the fluxes of Fig. 9. At the highest energy the truncation is quite severe, but as the energy decreases, the P_5 cross-section approximation is more accurate in describing the less rapid flux falloff and the truncation becomes less severe. Below about 0.5 MeV, the limiting value of μ is -1, and no truncation of the angular fluxes occurs. The plot of Fig. 9 has an artificial base at a value of 4×10^{-4} photons/cm²/sr/unit source. At that level, the angular fluxes are not truncated below about 2 MeV. The apparent truncation near 1.6 and 0.7 MeV is due to the resolution of the plot in the μ direction. Thus, the single Compton criterion appears to be a valid technique for eliminating the unphysical oscillations in the angular fluxes while retaining the more physical values.

In performing discrete ordinates transport calculations, a model is chosen that hopefully will be an accurate representation of the physical system being simulated while remaining tractable to computer solution. Typically, model parameters (such as boundary conditions and the extent of physical regions or media) and method parameters (such as P_n order, S_n order, and the mesh spacing) are varied in a systematic manner in search of those parameter values that yield convergent flux values. Usually, the fluxes are sensitive to a relatively small number of parameters,

so that the search for convergence is relatively brief and does not require undue effort. However, the angular fluxes of Phase II depend strongly on a large number of model and method parameters; namely, the P_n and S_n order, the mesh point spacing, the extent of the rock and air media, and the boundary conditions. Also, the effects of the individual parameters on the angular fluxes are not independent but are coupled. Consequently, the scheme utilized for determining the proper parameter values was not to seek convergence of the angular fluxes as a function of the parameters, but instead, to use Monte Carlo results to guide the selection of the parameter values. This scheme assures that not only have the angular fluxes converged properly, but also that they have converged to accurate values.

The Monte Carlo photon transport code MCNG⁶ was used to calculate photon currents crossing the rock-air interface of the Phase II geometry.³ The Monte Carlo model of the Phase II geometry was virtually identical to that used in the ONETRAN calculations. Photons having energies in the range 2.65-2.66 MeV were started from origins uniformly distributed in the rock medium and with directions chosen isotropically. Photons crossing the rock-air interface ($z=0$) with energies in the range 2.65-2.66 MeV were tallied as a function of μ . Thus, the MCNG code yielded photon currents integrated over specified angular subintervals about discrete values of μ . The angular fluxes for the group 2.65-2.66 MeV of the ONETRAN calculation were integrated over the same subintervals so as to be directly comparable to the MCNG results.

The direct currents (that is, those having $0 \leq \mu \leq +1$) are shown in Fig. 10 for the ONETRAN and MCNG calculations. The agreement between the two codes is excellent, indicating that the ONETRAN model adequately represents physical reality for the positive half of the angular domain.

The sky-shine fluxes (that is, those having $-1 \leq \mu \leq 0$) for this energy are very small compared with the direct fluxes (see Fig. 5). Calculation of the sky-shine currents using a single-step MCNG calculation would require many days of continuous

Fig. 10.

Comparison of Phase II direct currents at $z=0$ obtained from a Monte Carlo and a ONETRAN calculation. The discrete ordinates calculation was performed using a P_5 cross-section expansion, S_{12} quadrature, double Gaussian integration, and a monoenergetic 2.655-MeV gamma-ray source.

Fig. 11.

The direct currents of Fig. 10 are replotted to illustrate the linear dependence of the current over the range $0.00 \leq \mu \leq 0.14$.

computer execution to obtain a statistical precision comparable to that of the direct current. However, a two-step MCNG calculation has been used to obtain sky-shine currents having fair precision.

The geometry of Phase II suggests that only photons originating near the rock-air interface and emitted from the rock medium with an angle nearly parallel with the interface (that is, for positive μ near $\mu = 0$) can contribute to the current at the highest

energy (that is, 2.65-2.66 MeV). Figure 4 indicates that photons having energies in the range 2.65-2.66 MeV and emitted exactly parallel to the interface can contribute scattered current to values of μ no more negative than $\mu = -0.04$. Photons emitted at a small positive angle from the interface contribute current at an even more positive value of μ , while photons emitted at an angle greater than $\mu = 0.04$ do not contribute at all. Thus, only direct current emitted from the interface in the interval $0 \leq \mu \leq 0.04$ can contribute to the sky-shine current.

Sky-shine currents having acceptable precisions for reasonable computer execution times (that is, less than 30 min) were obtained by performing two separate MCNG calculations. The first computation yielded only the direct currents (that is, $0 \leq \mu \leq +1$) of Fig. 10. Those currents are replotted in Fig. 11 for the range $0.00 \leq \mu \leq 0.14$. The functional dependence (linear) of the direct current over the range $0.00 \leq \mu \leq 0.04$ was then used as input to the second MCNG calculation. That is, the sky-shine currents were calculated by starting photons at the interface with energies distributed uniformly in the range 2.65-2.66 MeV and starting angles distributed linearly in the range $0.00 \leq \mu \leq 0.04$ according to the functional dependence of the direct current shown in Fig. 11. Biasing the source with respect to starting direction resulted in the vastly decreased computer execution time.

The sky-shine currents obtained using the two-step technique are compared in Fig. 12 with ONETRAN currents obtained from three different calculations. Comparison of the MCNG sky-shine currents with those of the 265-group, P_5 , S_{12} , DP calculation reveal the gross overestimation of the current at this energy by the ONETRAN results. As stated earlier, the P_5 representation of the cross sections is very inadequate in describing the extremely forward-peaked scattering of photons at this energy. The 26-group, P_{10} , S_{24} , DP results are a better approximation but are very far from being accurate. The 26-group, P_5 , S_{24} , DP calculation is included to demonstrate that the 26-group, P_{10} , S_{24} , DP results are consistent with the 265-group, P_5 , S_{12} , DP results, that is, that the group structure and S_n order have

little effect on the currents and that any difference between the two calculations can be attributed solely to the difference in P_n order.

The discrepancy between the MCNG and ONETRAN sky-shine currents shown in Fig. 12 are probably due to MCNG modeling errors and inconsistencies as well as to the inadequacy of the low-order P_n approximations used by ONETRAN. The biasing of the starting photon angles and the assumption that the energy of the photons should be uniformly distributed between 2.65 and 2.66 MeV are probably not completely valid. However, it should be emphasized that these errors are probably not large and that most of the discrepancy observed is due to the inadequacy of the ONETRAN P_n approximation.

The accuracy of the ONETRAN results at lower energies was studied³ by using MCNG to calculate the current crossing the interface due to photons originating in the rock medium. The MCNG and ONETRAN models used were identical to those used in the previous comparison except that photons having

Fig. 12.

Comparison of Phase II sky-shine currents at $z=0$ obtained from a Monte Carlo calculation and three different ONETRAN calculations. The 265-group ONETRAN calculation used a monoenergetic 2.655-MeV gamma-ray source and the 26-group ONETRAN calculations utilized a monoenergetic 2.56-MeV gamma-ray source.

energies in the range 0.41-0.51 MeV (instead of 2.65-2.66 MeV) were started in the rock medium. The ONETTRAN results for this case indicated that the sky-shine currents were not dramatically smaller than the direct currents so that a single-step MCNG calculation (that is, obtaining both the direct and sky-shine currents in a single run) could be used.

Direct currents for the energy range 0.41-0.51 MeV from the MCNG calculation are compared in Fig. 13 with those from three ONETTRAN calculations. The MCNG results agree very well with all the ONETTRAN calculations, although the P_5 results probably are the most accurate. It is not surprising that the P_1 results are also accurate, since the direct current consists primarily of uncollided flux.

Sky-shine currents for the energy range 0.41-0.51 MeV from the MCNG calculation are compared in Fig. 14 with those from three ONETTRAN calculations differing only in P_n order. The P_5 results are the most accurate and the agreement with the MCNG results is good, although the P_5 results might be systematically larger than the MCNG results. A possible explanation of the slight discrepancy can be found in the cross sections used by the MCNG and ONETTRAN calculations. MCNG uses point cross sections while the ONETTRAN results shown in Fig. 14 were obtained using cross sections averaged over the energy bin 0.41-0.51 MeV. The averaging used a uniform weighting scheme instead of being weighted by the actual flux shape in the bin. At this energy, the Phase II fluxes increase with decreasing energy, resulting in average cross-section values that would be larger than those obtained using uniform weighting. Thus, the ONETTRAN cross sections do not remove (primarily via the photoelectric cross section) as much flux from the bin as they should or as the MCNG cross sections do, and the ONETTRAN currents are larger than the MCNG currents. The narrower energy bin widths of the 265-group calculations (10-keV width vs the 100-keV width of the 26-group calculations) will minimize the effect for the final Phase II results.

Fig. 13.

Comparison of Phase II direct currents at $z=0$ resulting from a Monte Carlo calculation and three different ONETRAN calculations. The ONETRAN calculations used double Gaussian integration, S_{12} quadrature, a monoenergetic 0.46-MeV gamma-ray source, and cross-section Legendre expansions of order P_1 , P_3 , and P_5 . The results for the P expansion are identical to those for the P_5 expansion and have not been plotted.

Fig. 14.

Comparison of Phase II sky-shine currents at $z=0$ obtained from a Monte Carlo calculation and three different ONETRAN calculations. The ONETRAN calculations were performed using double Gaussian integration, S_{12} quadrature, a monoenergetic 0.46-MeV gamma-ray source, and cross-section Legendre expansions of order P_1 , P_3 , and P_5 .

The MCNG studies indicate that the accuracy of the direct currents (and angular fluxes) is very good for all photon energies. This result is not too surprising, since, as mentioned before, the direct fluxes consist chiefly of uncollided flux from the rock medium. Consequently, the P_n approximation of the scattering cross sections has little effect on the angular fluxes.

Fig. 15.

Phase II sky-shine angular fluxes at the interface ($z=0$) for the energy group 0.51-0.61 MeV. The results were obtained from three different ONETRAN calculations using a 26-group energy structure, S_{24} quadrature, double Gaussian integration, a monoenergetic 2.56-MeV gamma-ray source, and cross-section expansions of order P_1 , P_5 , and P_{10} .

Fig. 16.

Phase II sky-shine angular fluxes at the interface ($z=0$) for the energy group 1.51-1.61 MeV. The results were obtained from six different ONETRAN calculations using a 26-group energy structure, S_{24} quadrature, double Gaussian integration, a monoenergetic 2.56-MeV gamma-ray source, and cross-section expansions of order P_1 , P_3 , P_5 , P_7 , P_9 , and P_{10} .

The accuracy of the sky-shine currents (and angular fluxes) is good at low energies, but worsens as the energy increases and the low-order P_n approximation becomes insufficient to accurately describe the scattering. This trend in accuracy as a function of energy is verified somewhat by the Phase II ONETRAN P_n studies shown in Figs. 15-17. All data were obtained using 26-group, S_{24} , DP calculations with the source photons having energies uniformly distributed in the range 2.51-2.61 MeV. Figure 15 shows the sky-shine angular fluxes at the interface ($z=0$) for the energy group 0.51-0.61 MeV. The fluxes appear to have converged for the P_5 calculation. This is consistent with the MCNG/ONETRAN comparison at low energies.

The sky-shine angular fluxes for the energy group 1.51-1.61 MeV are shown in Fig. 16 for different P_n orders. The fluxes appear to have converged for the P_9 calculation, although one cannot be certain from the data of Fig. 16. Finally, Fig. 17 shows the sky-shine angular fluxes for the energy group 2.51-2.61 MeV, the source group. Clearly, the fluxes have not converged, even for the P_{10} calculation. This again verifies the trend in accuracy seen earlier in the MCNG/ONETRAN comparison at high energy.

Fig. 17.
Phase II sky-shine angular fluxes at the interface ($z=0$) for the energy group 2.51-2.61 MeV. The results were obtained from the six ONETRAN calculations that yielded the results shown in Fig. 16.

Fig. 18.

Spectral plots of the Phase II direct (upper curve) and sky-shine (lower curve) currents at the interface ($z=0$). A P_5 , S_{12} , DP, 265-group ONETRAN calculation having a monoenergetic 2.615-MeV gamma-ray source (that is, the most energetic line of the thorium spectrum) was used to obtain the results.

Fig. 19.

Spectral plots of the Phase II direct (upper curve) and sky-shine (lower curve) currents at $z=0$. The results were obtained from a P_5 , S_{12} , DP, 265-group ONETRAN calculation using the 20 most intense lines of the thorium spectrum as the gamma-ray source.

The inaccuracies in the sky-shine currents (and angular fluxes) at high energies are not quite so disconcerting if one considers the relative magnitude of the sky-shine and direct currents. The upper curve of Fig. 18 is the total direct current at the interface; that is, the direct angular fluxes have been integrated over the angular region $0 \leq \mu \leq +1$. The lower curve is the total sky-shine (that is, $-1 \leq \mu \leq 0$) current at the interface. The results were obtained using a P_5 , S_{12} , DP, 265-group ONETRAN calculation with the source photons having energies in the group 2.61-2.62 MeV. The sky-shine spectrum results simply from an integration over μ of the angular fluxes of Fig. 9. In fact, the energy shape is essentially identical to those of Figs. 6 and

7. The total sky-shine current is less than 10% of the direct current for energies greater than about 0.8 MeV and less than 1% for energies greater than about 2.3 MeV. Thus, at high energies where the sky-shine fluxes are inaccurate, their contribution to the total (both direct and sky-shine) flux is negligible.

The contribution of the sky-shine fluxes to the total flux is typically smaller for the case of a thorium spectrum than for the simple spectrum of Fig. 18. This is illustrated in Fig. 19 where, again, the upper curve is the total direct current and the lower curve is the total sky-shine current at the interface. The results were obtained using a P_5 , S_{12} , DP, 265-group ONETRAN calculation with the source photons having the energy distribution of the 20 most intense lines of the thorium spectrum.⁷ The total sky-shine current is about 0.001% of the direct 2.614-MeV line, about 0.1% of the 1.588-MeV line, and about 0.5% of the 583-keV line. Thus, it comes as no surprise that the sky-shine current is even more negligible when compared with the peaks of the direct current instead of the Compton continuum.

Preliminary studies of the Phase II angular fluxes have indicated that relatively accurate results can be obtained using the discrete ordinates method of ONETRAN to calculate the fluxes and using the single Compton scattering criterion to eliminate any unphysical values. Results obtained to date indicate that accurate angular fluxes can be computed at high energies only by using a very high order P_n expansion of the photon cross sections. The current version of ONETRAN can only accommodate 265-group cross-section expansions of order up to five (for the Phase II geometry). A new version of the code will soon be available that can accommodate up to a P_{10} expansion of the cross sections. This higher order expansion will permit calculation of accurate angular fluxes for energies as high as about 1.5 MeV. A cross-section expansion of order 50 (or maybe even higher) would be required to accurately calculate the highest energy angular fluxes. Final parameter values for the ONETRAN model of the Phase II geometry will be chosen after preliminary results have been generated utilizing the new (higher P_n order) version of ONETRAN.

B. Detector Response Function

(M. L. Evans)

The computer code GAMRES⁸ was developed at the Los Alamos Scientific Laboratory (LASL) for the calculation of gamma-ray response functions for various NaI(Tl) or plastic scintillation detectors commonly used for uranium exploration. In mapping the detector response function, the code will be run for a large number of incident photon energies (for example, about 90 energies are required to map the response over the energy domain 10 keV - 3 MeV). The large number of incident energies is necessary to ensure accurate interpolation of the basic map (this topic will be addressed more completely in subsequent progress reports). Thus, GAMRES must be capable of producing precise pulse-height spectra in short running times.

GAMRES is an analogue Monte Carlo code that determines the detector response function by tracing the history of photons impinging on the scintillator. Interactions of the photons with the scintillator (and surrounding container or shielding) are simulated by sampling approximate expressions describing the Compton, photoelectric, and pair production cross sections. Electron and photon energies and scattering angles are determined by solving transcendental expressions for the appropriate roots. Studies were performed to determine whether the Newton-Raphson method⁹ would be more efficient than the bisection method¹⁰ in estimating the electron energy in Compton scattering, the electron scattering angle in the photoelectric effect, and the fraction of kinetic energy taken by the positron in pair production. The bisection method has been used to calculate the spectra obtained to date.

Difficulty arose in ensuring convergence of the Newton-Raphson method for all photon energies of interest (that is, 10 keV - 3 MeV). The method was modified in an attempt to overcome the problem but to no avail. Even for the cases for which the method converged, it was only slightly faster than the bisection method (to the precision required by the code). Thus, the bisection method (which always converges) was chosen as the preferred

root-solving algorithm, and the coding for the method was optimized for execution speed and consistency among the various subroutines of GAMRES.

The accuracy to which the electron and photon energies and scattering angles are computed determines the speed of execution of the code. A study of the tradeoff between accuracy and execution speed must be made if a reasonable choice of convergence criteria is to be made. Figure 20 summarizes such a study for the GAMRES code. Simulation of each type of photon interaction (that is, Compton scattering, photoelectric effect, and pair production) requires the solution of a transcendental equation. Finding the root using the bisection method requires multiple calls to function subprograms to evaluate the transcendental expression.

Plotted in Fig. 20 is a curve for each type of simulated interaction relating the number of function calls to the convergence parameter for that interaction. All data were obtained for 10 000 incident photons having an energy of 2 MeV. Preferred values of the convergence parameters are indicated by arrows in Fig. 20. The value $\epsilon = 0.004$ for Compton scattering provides for better than 1-keV resolution in the energy of the Compton electron. This was necessary because of the small pulse-height bin widths encountered in the interpolation of the energy (E) vs pulse-height (PH) surface (this topic will be discussed in more detail in subsequent reports). The convergence parameter

Fig. 20.
Relationship between the number of function subprogram calls and the convergence parameter ϵ for the three photon interactions simulated by GAMRES. The bisection method of root approximation gives rise to the general shape of the curves.

value $\epsilon = 0.002$ for the photoelectric effect resulted in better than 0.001 precision in determining the cosine of the scattering angle of the photoelectron. Thus, the imprecision in the scattering angle was limited to less than 2.6° for the worst case (that is, forward scattering). Finally, the value $\epsilon = 0.002$ for pair production yields a precision better than 0.001 in the fraction of kinetic energy taken by the positron. This results in a resolution of about 2 keV in the positron energy for the worst case to be treated using this code, that is, for incident photons having an energy of 3 MeV.

The convergence values chosen above together with the code optimization discussed earlier have led to a 50% improvement in execution speed, that is, from about 1000 incident photon histories per second to about 1500. These values are averaged over incident photon energies in the range 10 keV - 3 MeV. Thus, a response function map containing 90 discrete pulse-height spectra having 10 000 events each can be obtained in approximately 10 min of CDC 7600 computing time.

This faster GAMRES code was used as the starting point in developing a version of the program to be used in the response function mapping process. A flowchart outlining the scheme for generating and testing the response function map is shown in Fig. 21, where rectangular boxes denote computer programs, oval boxes denote data files, and the trapezoidal-shaped box denotes output. The program GAMRESP is a production version of the fast GAMRES code that includes a scheme (to be described in a subsequent progress report)

Fig. 21.
Flowchart outlining the scheme for generating and checking a detector response function map.

for selecting the incident gamma-ray energies to be used in the map and for the automatic restarting of the code to provide a spectrum for each of those energies. GAMRESP also includes coding for selecting the binning to be used in the pulse-height spectra that is consistent with the incident gamma-ray energies chosen. The result of the GAMRESP execution is a file called BASMAP which consists of a header containing descriptive information and parameter values, followed by the pulse-height spectra that comprise the basic energy vs pulse-height surface to be interpolated.

The program GAMCHK is a modified version of GAMRESP that is used to produce a low-precision energy vs pulse-height map to be used in performing a least-squares comparison with the interpolated surface. The incident gamma-ray energies are chosen to be midway between those of the basic map, that is, where the interpolation distances are largest and, hence, the interpolation errors are likely to be most severe. Thus, the least-squares comparison should provide a measure of the upper bound on the interpolation errors.

Execution of the GAMCHK code results in a file called CHKMAP that has the same structure as the file BASMAP, but which contains the least-squares checking map, not the high-precision basic map used in the interpolation. BASMAP and CHKMAP serve as input to the program PHSURE, which reads the basic map from BASMAP and performs two-dimensional spline interpolation on the energy vs pulse-height surface. The code permits linear, quadratic, and/or cubic interpolation in both the energy and pulse-height directions. Recent modifications enable the code to accept a basic map containing up to 90 channels for each independent variable (that is, a 90-point x 90-point basic map) and to provide interpolated values of the energy vs pulse-height surface on a grid containing up to 200 channels for each independent variable (that is, a 200-point x 200-point grid).

The larger grid is input to a subroutine called PIC3D which is a three-dimensional plotting routine. PIC3D provides high-resolution plots for visual inspection of the interpolated energy vs pulse-height surface. The plots can be used for identifying

regions of the surface wherein the interpolation yields unphysical results such as oscillations and under-shooting (that is, negative interpolated values). The current version of PIC3D is a high-precision version of the plotting routine used to produce the energy vs pulse-height plots shown in the preceeding progress report.⁸

In addition to having the capability of making plots utilizing a very dense grid, PIC3D also permits the numbering and labeling of the plot axes, the use of a window by which an exploded view of a part of the surface can be obtained, and the plotting of different parts of the surface in different colors for use as a color coding tool. PIC3D is also faster in execution speed and less memory intensive than its predecessor.

Examples of PIC3D usage in the visual examination of the interpolated energy vs pulse-height surface are shown in Figs. 22-27. The basic map used in the interpolation consisted of 89 pulse-height spectra each of which was due to 10 000 photons of a different energy (in the range 15 keV-

Fig. 22.
The response function map calculated for a 7.62-cm-diam x 7.62-cm-long cylindrical NaI scintillator for incident photons having energies in the range 15 keV-2.94 MeV. Both the energy (E) and pulse height (PH) axes are visible.

Fig. 23.
The response function map of Fig. 22 rotated clockwise through an angle of about 30° with respect to the viewer. The energy axis is obscured by the surface.

Fig. 24.
Logarithmic (base 10) plot of
the response function map of
Fig. 22 viewed from the same
perspective.

Fig. 25.
Logarithmic (base 10) plot of
the response function map of
Fig. 23 viewed from the same
perspective.

3 MeV) impinging on the flat face of a 7.62-cm-diam x 7.62-cm-long cylindrical NaI scintillator from a point source located on the cylinder axis 10 cm in front of the flat face of the detector. The scintillator was surrounded by a 0.254- μ m (10-mil) iron container in a manner identical to that used to obtain the pulse-height spectra presented in earlier progress reports.⁸

The response function (that is, energy vs pulse-height surface) plotted in Figs. 22-27 is the same, but different perspectives and display options are used in the different figures. A linear plot of the response function is shown in Figs. 22 and 23. In Fig. 22 the energy axis (E) is located on the left side of the plot, increasing in magnitude from deep within the plot to the foreground (0.015 MeV to 2.94 MeV). The pulse-height axis (PH) is located in the foreground and increases in magnitude from left to right (0.0075 MeV to 2.97 MeV). The response axis (in units of counts/channel) is located on the left side of the plot near the origin symbol (O) and increases in magnitude from 0 to 5500.

Fig. 26.

Three-dimensional plot of the response function map of Fig. 23 using the window option of the PIC3D to obtain an exploded view of the high-energy, high-pulse-height region of the E vs PH surface. The perspective is slightly different from that of Fig. 23, that is; the viewer is much closer to the base (response = 0) of the plot.

Fig. 27.

Exploded view of the high-energy, high-pulse-height region of the E vs PH surface shown in the logarithmic plot of Fig. 25. The perspective is identical to that of Fig. 25 but the scale of the response axis is different.

The most prominent feature of the plot is the full-energy ridge that lies diagonally across the E vs PH plane. The ridge results from the very nearly linear relationship at these energies between incident photon energy and full-energy peak pulse-height values. That is, a photon detected by the scintillator gives rise to a full-energy peak that has a mean pulse-height value approximately equal to the incident photon energy. The width of the full-energy peak increases with increasing energy, even though the resolution improves at higher energies. The increasing photopeak efficiency at lower energies is manifested by ever larger full-energy peak amplitudes. However, for incident photon energies less than about 70 keV, the iron container surrounding the NaI scintillator greatly attenuates the incoming photon flux, resulting in a strongly reduced response at very low energies.

Other features of the response function such as the valley between the full-energy ridge and the Compton continuum are difficult to discern. The perspective of Fig. 23 affords a slightly enhanced view of the surface, especially at the higher energies. In Fig. 23 the surface of Fig. 22 has been rotated clockwise (through an angle of about 30°) with respect to the viewer. The origin of the plot is obscured by the E vs PH surface and the energy and response axes are not numbered. The valley between the full-energy ridge and the Compton continuum is just barely recognizable as are the first and second escape ridges running nearly parallel to the full-energy ridge at high energies. The linear scale for the response function greatly impairs the observation of statistical imprecision in the surface, especially for small pulse-height values in the Compton continuum.

The plots of Figs. 24 and 25 are virtually identical to those of Figs. 22 and 23, respectively, except that the common logarithm of the response function has been plotted instead of a linear plotting of the response. The valley between the full-energy peak and the Compton continuum is clearly discernible, especially in Fig. 25, and the first and second escape ridges are easily observable. The Compton shoulder for the highest energy can be seen and the statistical imprecision is clearly evident. At large values of energy and small values of pulse height the Compton continuum is relatively small in magnitude (about 10 counts/channel) and exhibits the resulting imprecision.

The window option of PIC3D is utilized in Figs. 26 and 27 to obtain exploded views of the high-energy, high-pulse-height region of the E vs PH surface. The perspective of the linear plot of Fig. 26 is different from that of Fig. 23 (linear plot of the entire surface), as is the scale of the response axis. Both changes enhance the view of the high-energy, high-pulse-height region to an extent that makes the plot useful for examining the fine structure of the response function. The perspective of the log plot of Fig. 27 is identical to that of Fig. 25 (log plot of the entire surface), but the scale of the response axis is different. Again, the exploded view affords a more detailed examination of the E vs PH surface.

The three-dimensional plotting capabilities available with the interpolation code PHSURF provide for a complete but somewhat subjective check of the interpolated E vs PH surface. Visual examination of the surface is supplemented by the least-squares comparison mentioned earlier. The comparison between the check map (file CHKMAP) and the interpolated E vs PH surface is performed for each of the individual pulse-height spectra contained in the check map. That is, a least-squares comparison is made for each incident photon energy of the check map. In this way, one can identify those energy regions of the interpolated surface for which the spline interpolation does not yield an accurate representation of the basic map. The code PHSURF also provides a composite least-squares comparison that indicates the validity of the interpolated surface as a whole. Thus, PHSURF provides for both subjective and quantitative checking of the interpolated E vs PH surface.

The output of PHSURF consists of parameter values and data that are printed, plotted, and written onto a magnetic disk or tape. Response function values from the basic map, check map, and interpolated surface are printed for ease of detailed inspection and analysis. The results of the various least-squares comparisons are also printed out. Three-dimensional plots of the interpolated surface can be produced on either 35-mm film (color or black and white) or on microfiche. If the subjective and quantitative checking indicates that the interpolated surface is, in fact, an accurate representation of the basic map, a 265- x 265-channel response function map is produced and written into the file GAMMAP. The energy and pulse-height values used for the 265- x 265-point interpolation are, of course, consistent with the 10-keV binning structure of the ONETRAN calculations of Phases I, II, and III. Thus, the GAMMAP file can then be used as input to a separate computer program that enfolds the detector response function with the photon fluxes of the discrete ordinates calculations.

II. DISEQUILIBRIUM PROBE CALCULATIONS

(M. Jain and M. L. Evans)

Modification of the TWOTRAN¹¹ code to accept externally mixed cross sections has been completed. This alleviated computer core storage problems as discussed in the last progress report.¹² However, with the large two-dimensional geometry, excessively long computer execution times became a major problem. For example, a single P_2 , S_8 calculation with ^{137}Cs and KUT sources took about 10 min of CDC-7600 CPU time. Therefore, the process of finding optimum values for the parameters P_n and S_n for the given probe dimensions and of completing the set of calculations for different uranium concentrations became extremely time consuming.

Furthermore, recent experimental data¹³ indicate that the signature obtained from formations in disequilibrium may be due to processes other than a change in average Z of the formation. The ratio of counts in the filtered detector without a source to the unfiltered detector with a source increased for uranium-deficient formations rather than decreasing as would be the case if changing formation $\bar{Z}$ were causing the signal (see Ref. 12, Fig. 4). A possible explanation for this effect could be x-ray fluorescence (XRF) of the uranium in the ore. Since the TWOTRAN code has no provision for XRF, a Monte Carlo code, MCNP¹⁴, was used for the subsequent calculations.

Initially, a geometric model similar to that which had been used in the TWOTRAN calculations was used to compute the gamma-ray current across the surfaces of the NaI detectors. The calculations were performed using the 662-keV gamma-ray line of ^{137}Cs as the source. Due to the unfavorable geometry the ratio of the number of gamma rays reaching the NaI detector to the number of source gamma rays was about 10^{-4} . To increase the solid angle, a new model was implemented in which the 15.24-cm-thick cylindrical tungsten shield was replaced by a cone of height 5.08 cm and by a 2-cm-thick cylinder (see Fig. 28). In addition, the ^{137}Cs source was biased with regard to the selection of gamma-ray starting directions. That is, gamma rays

were started from the source with only positive Z-components of the direction cosine, resulting in gamma rays emitted only "toward" the detector.

The new model geometry enhanced the gamma-ray flux incident on the detector, but excessively long computer execution times were still needed to obtain adequate statistical precision. As a result, the point-detector flux option of MCNP was used in an attempt to improve the precision of the calculations. In this approach the contribution to the flux at a given point (say, at the center of the detector) is calculated from the points in the system at which interactions take place. From a point of interaction, the contribution to the uncollided flux at the detector position is computed using the cross sections of the intervening media. Thus, the point-detector flux option can yield improved precision in the flux estimates because of an effectively larger sample size (that is, every interaction taking place in the system contributes to the flux).

The calculations were performed with and without XRF for 1% and 2% uranium concentrations (by weight) in sandstone. Figures 29 and 30 show the fluxes without XRF for 2% and 1% uranium concentration, respectively. The flux values for 2% U are smaller than those for the 1% U case and reflect the increased photoelectric absorption of the ore containing the larger uranium concentration. Figures 31 and 32 show the fluxes (including the XRF contribution) for 2% and 1% U concentration, respectively. Again, the flux values for the 1% U case are larger than those for the case with 2% U concentration. However, for both cases there is a peak in the flux in the region of uranium XRF radiation (that is, 94-115 keV).

The enhancement due to XRF is directly seen in comparing Figs. 31 and 29 for 2% U concentration (and Figs. 32 and 30 for 1% U). The flux increases in the U x-ray region for both concentrations.

These calculations demonstrate the importance of the XRF contribution to the signal. For concentrations of about 1% this

Fig. 28.

Model geometry used in the initial Monte Carlo feasibility studies of the x-ray fluorescence (XRF) downhole probe. All dimensions are in centimeters.

Fig. 29.

Gamma-ray spectrum at the point detector for a 0.5- μCi ^{137}Cs source and for a sandstone formation ($\rho = 2.6262 \text{ g/cm}^3$) containing 2% uranium by weight. XRF radiation was not taken into account.

Fig. 30.

Gamma-ray spectrum at the point detector for a 0.5- μCi ^{137}Cs source and for a sandstone formation containing 1% uranium by weight. XRF radiation was not taken into account.

signature looks very promising as a tool for in situ uranium assay. Further studies are in progress to elucidate the method, especially at lower uranium concentrations.

III. PROTOTYPE PHOTONEUTRON LOGGING PROBE DEVELOPMENT

A. Recoil Proportional Counters

(M. P. Baker and H. O. Menlove)

Methane (CH_4) recoil proportional counters are being evaluated for use as fast-neutron detectors in the prototype, photoneutron-based logging tool.¹⁵ Previous laboratory uranium ore measurements were made using an 18-atm ^4He recoil proportional counter.¹⁶ The evaluation of methane counters as a

Fig. 31.

Gamma-ray spectrum at the point detector for a 0.5- μCi ^{137}Cs source and for a sandstone formation containing 2% uranium by weight. XRF radiation was included in the transport.

Fig. 32.

Gamma-ray spectrum at the point detector for a 0.5- μCi ^{137}Cs source and for a sandstone formation containing 1% uranium by weight. XRF radiation was included in the transport.

Fig. 33.
Geometry for evaluation of 5-cm-diam x 30-cm-long recoil proportional counter with ^{252}Cf spontaneous fission neutron source and mock-up of borehole photoneutron source.

Fig. 34.
Pulse-height spectrum obtained from an 18-atm ^4He detector with incident neutron and gamma ray fluxes described in Fig. 33.

potential replacement for the ^4He counter is motivated by two considerations.

- 1) For a moderated fission neutron energy spectrum, the energy dependence of the H scattering cross section favors CH_4 .
- 2) CH_4 has four H atoms per molecule while ^4He has only one He atom per molecule. Thus, if all other factors were equal, a CH_4 counter would have four times the counting efficiency of a ^4He counter.

The initial stage of the evaluation was performed in the configuration shown schematically in Fig. 33. The 5-cm-diam x 30-cm-long detector to be evaluated was irradiated with ^{252}Cf spontaneous fission neutrons from one side and a mockup of the borehole photoneutron source from the other side. This mockup consisted of a $\sim 100\text{-mCi } ^{88}\text{Y}$ gamma-ray source inserted into a hole along the axis of a 7.5-cm-diam x 10-cm-long Be cylinder. This combination produced a gamma dose of 1-2 R/h at the detector.

The anode of the recoil proportional counter was operated at positive high voltage with the cathode at ground potential. The signals from the counter were processed by a charge-sensitive pre-amplifier and an Ortec 450 amplifier before passing to a multi-channel analyzer for pulse-height analysis.

The pulse-height spectrum obtained with the 18-atm ^4He counter is shown in Fig. 34. The spectrum is characterized by two distinct regions. At low pulse heights, a rapidly falling exponential distribution from gamma-ray interactions is observed. At larger pulse heights the events due to neutron interactions are clearly discernable. The dotted line shows the spectrum obtained with only the gamma-ray source present. Thus, events above the discrimination level shown in the figure are due only to neutron interactions.

For a fixed source-detector geometry, the neutron counting rate above the discrimination level might be expected to depend on several variables, for example, high voltage, amplifier time constant, gas pressure, etc. Figure 35 illustrates the effect of two of these variables for the 18-atm ^4He detector. The neutron counting rate is plotted as a function of high voltage with the amplifier time constant as parameter. For a given time constant, the neutron counting rate has a roughly parabolic dependence on high voltage. As the time constant is shortened, the maximum achievable neutron counting rate increases. This is due to the longer pulse rise time for a gamma-ray interaction compared to a neutron interaction. A short clipping time will then effectively diminish the pulse height of gamma-ray events relative to neutron events. The disadvantages of the shorter amplifier time constants

are that a larger amplifier gain is required for a given output signal size and somewhat higher voltage is required on the detector.

In order to evaluate the performance of CH_4 detectors using the criteria discussed above, a series of 5-cm-diam x 30-cm-long CH_4 detectors were obtained from Reuter-Stokes. The detectors were fabricated with 0.8-mm-thick stainless steel walls and had fill pressures that varied from 3 to 18 atm as shown in Table I. A series of measurements similar to those shown in Fig. 35 were carried out for the CH_4 detectors and again indicated that an amplifier time constant of 0.1 μs gave the highest counting rate. Of major interest, however, was the fill pressure and high voltage behavior of these detectors. The results of changes in these parameters on neutron counting rate are shown in Fig. 36.

The 3-atm and 5-atm CH_4 detectors behave as expected with the maximum neutron counting rate per unit pressure roughly four

Fig. 35.
Effective neutron counting rate as a function of applied high voltage and amplifier time constant (τ) for 5-cm-diam x 30-cm long recoil proportional counter.

Fig. 36.
Effective neutron counting rate as a function of applied high voltage and detector gas and fill pressure for 5-cm-diam x 30-cm-long recoil proportional counters.

times that obtained for the 18-atm ^4He detector. However, for CH_4 detector fill pressures higher than 5 atm, the response per unit pressure begins to fall rapidly. For example, the 7-atm detector gives only ~6% more response than the 5-atm detector while a 40% increase would have been expected.

TABLE I
Fill Pressures, Approximate Operating Potentials, and Relative Neutron Counting Rates for 5-cm-diam x 50-cm-long CH_4 Recoil Proportional Counters

Pressure (atm)	Approximate Operating Voltage (V)	Relative Counting Rate
3	2500	5.6
5	3200	9.9
7	4000	12.5
10	5000	10.4
13	6400	6.8
18-atm ^4He	1900	3.6

The saturation and eventual reduction in the fast-neutron response of the CH_4 detectors with increasing fill pressure can only be partially explained by the increase in gamma sensitivity. Measurements conducted with monoenergetic neutrons produced by group Q-1's Van de Graaff accelerator indicated that, even with a relatively small flux of gamma radiation incident, the fast-neutron response per unit pressure decreased as the detector pressure was increased. This effect was accompanied by a broadening in resolution with increasing pressure. The reason for this behavior is not understood, but may involve increased recombination of ions and electrons in the detectors as the pressure is increased.

Increased gamma sensitivity with increasing detector pressure is at least generally understood. The response of a gas recoil proportional counter to gamma radiation is due to interactions both in the gas and in the container walls. In the latter case, only interactions in which an electron escapes from the wall into the gas will result in a pulse in the detector. As the pressure of the gas in the detector is raised, the probability of direct interaction in the gas increases essentially linearly. In addition, the energy deposited in the gas by recoil electrons from either source increases with pressure. The combined result of these effects is that the gamma-ray background, as shown in Fig. 2, extends to larger pulse heights as the gas pressure is

raised. Thus, the discrimination level must be raised to maintain the separation between neutrons and gamma rays. Therefore, due to the increasing gamma sensitivity, the effective neutron sensitivity increases with increasing fill pressure at something less than the expected linear rate.

The observed effects of the gamma sensitivity on fast-neutron measurements depends critically on the gamma-to-neutron ratio, the gamma-ray and neutron energy spectra, and the type of gas used. For CH_4 the results with $\sim 1\text{R/h}$ of high-energy gamma radiation and ~ 300 fission spectrum neutrons/ $\text{cm}^2\text{-s}$ are shown in Fig. 36. For this case the 7-atm CH_4 counter is about 20-25% more sensitive than the 18-atm ^4He detector.

The situation is likely to be quite different for the uranium borehole logging geometry in that both the neutron and gamma-ray spectra will be considerably "softened" compared to the present case. In order to determine the effects of this spectral softening, measurements similar to those described above will be performed in a borehole mockup.

B. Liquid Scintillation Detector

(M. P. Baker)

Liquid scintillators are also being evaluated for use as fast-neutron detectors in the prototype photoneutron logging probe.¹⁷ These detectors are very attractive for this purpose in that the hydrogen density and thus the fast-neutron interaction probability is very high. Unfortunately, the electron density and thus the gamma-ray interaction probability is also quite high. (The interaction probabilities for 1-MeV neutrons and gamma rays in a 1-cm-thick slab of liquid scintillator are $\sim 20\%$ and $\sim 6\%$, respectively.) In addition, the light output from recoil electrons is considerably higher than that from recoil protons of the same energy. Thus, discrimination between neutrons and gamma rays on the basis of pulse height alone is not possible in organic scintillators. Fortunately, neutrons and gamma-rays can be discriminated by their pulse shape differences in certain liquid organic scintillators.

In general, suitable pulse-shape discrimination of neutrons and gamma rays requires that the ratio of gamma-ray and neutron

fluxes not be excessive. Thus, fast-neutron detection with the liquid scintillator requires more gamma-ray shielding than would be needed in conjunction with a gas recoil proportional counter. Since the total (shielding + detector) diameter must be conserved in the borehole logging tool geometry, an increase in gamma-ray shielding implies a decrease in detector volume. Thus, in using a liquid scintillator in the borehole logging tool, a large gain in intrinsic efficiency is offset by a loss in detector volume.

In order to investigate the areas outlined above, a 1.2-cm-diam x 7.6-cm-long liquid scintillator has been designed and constructed. The design features are based on the use of the scintillator in the prototype photoneutron logging tool as shown in Fig. 37. The scintillator is constructed from a high-density ($\rho = 18 \text{ g/cm}^3$) tungsten alloy cylinder prepared by a powder metallurgy process in order to achieve maximum gamma-ray shielding. The scintillator solution is located in a cylindrical volume on the axis of the tungsten cylinder with dimensions as stated above. The container in which the solution resides is highly polished stainless steel.

The detector is filled via the channels through the tungsten. One channel is connected to a Teflon tube, which is wrapped around the outside of the tungsten cylinder, as shown in Fig. 38. This tube is also filled with liquid scintillator solution and serves as a reservoir, which maintains the detector volume at a constant value during changes of ambient temperature. The detector body is threaded at one end to accommodate connection to a photomultiplier tube assembly. The assembly consists of an

Fig. 37.
Liquid scintillation counter for detection of fast neutrons in the prototype photoneutron logging tool.

Fig. 38.
Photograph of liquid scintillation detector and photomultiplier tube assembly

Fig. 39.
Photograph of liquid scintillation detector assembly.

RCA 8575 photomultiplier tube in a high-density tungsten gamma-ray shield and an ORTEC 265 resistive divider chain. The photomultiplier tube views the liquid scintillator solution through the Pyrex window attached to the detector body with Torr-Seal. Figure 39 shows the complete detector package with the photomultiplier tube assembly threaded into the detector body.

The detector has been filled with NE218 liquid scintillation solution purchased from Nuclear Enterprises, Inc. The filling procedure is complicated by the fact that the xylene-based solution is highly corrosive when in contact with a wide variety of common materials and that the pulse-shape discrimination properties are compromised by dissolved oxygen. Thus, the detector was filled inside a plastic bag (fitted with gloves), which had nitrogen flowing through it. After bubbling nitrogen through the solution, the detector was filled by forcing the solution through the Teflon tube and into the scintillator volume.

Initial tests indicate that the detector is yielding pulses of the proper magnitude. Measurements of the pulse-shape discrimination properties and the effectiveness of the tungsten shield are in progress.

REFERENCES

1. M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-6840-PR (June 1977), pp. 12-13.
2. M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-6996-PR (October 1977), pp. 2-6.
3. M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7139-PR (February 1978), pp. 3-5.
4. M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7251-PR (April 1978), pp. 2-3.
5. Erwin Kreyszig, Advanced Engineering Mathematics, John Wiley & Sons, New York (1968), p. 469.
6. E. D. Cashwell, J. R. Neergaard, W. M. Taylor, and G. D. Turner, "MCN: A Neutron Monte Carlo Code," Los Alamos Scientific Laboratory report LA-4751 (January 1972); E. D. Cashwell, J. R. Neergaard, C. J. Everett, R. G. Schrandt, W. M. Taylor, and G. D. Turner, "Monte Carlo Photon Codes: MCG and MCP," Los Alamos Scientific Laboratory report LA-5157-MS (March 1973); MCNG is a hybrid version of MCN and MCG.
7. M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-6840-PR (June 1977), p. 4.
8. M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-6996-PR (October 1977), pp. 7-12; M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7139-PR (February 1978), pp. 11-18; and M. L. Evans, D. A. Close, and M. Jain, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7251-PR (April 1978), pp. 3-5.
9. Carl-Erik Froberg, Introduction to Numerical Analysis (Addison-Wesley, Reading, Massachusetts, 1969), pp. 21-28.
10. R. W. Hamming, Numerical Methods for Scientists and Engineers (McGraw-Hill, New York, 1973), pp. 62-63.
11. K. D. Lathrop and F. W. Brinkley, "TWOTRAN II: An Interfaced, Exportable Version of the TWOTRAN Code for Two-Dimensional Transport," Los Alamos Scientific Laboratory report LA-4848-MS (July 1973).

12. M. Jain, M. L. Evans, and D. A. Close, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7251-PR (April 1978), pp. 5-6.
13. W. Miller, Geosciences Inc., personal communication, June 1978.
14. LASL Group TD-6, "MCNP - A General Monte Carlo Code for Neutron and Photon Transport," Los Alamos Scientific Laboratory report LA-7396-M (July 1978).
15. M. P. Baker, M. L. Evans, and H. O. Menlove, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7251-PR (April 1978), pp. 8-9.
16. M. P. Baker and H. O. Menlove, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-6840-PR (June 1977), pp. 16-22.
17. M. P. Baker, "NDA Technology for Uranium Resource Evaluation," Los Alamos Scientific Laboratory report LA-7139-PR (February 1978), pp. 21-23.