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NDA Technology for Uranium Resource Evaluation

Compiled by

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NDA TECHNOLOGY FOR URANIUM RESOURCE EVALUATION

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ABSTRACT

The following progress report describes the work performed during the first quarter (October 1-December 1, 1976) on the contract for NDA Technology for Uranium Resource Evaluation. The initial phases of the work focused on gamma-ray calculations and computer code modifications in support of borehole logging measurements and surface gamma-ray surveys. The existing gamma-ray transport computer codes at Los Alamos Scientific Laboratory (LASL) are being modified to give the energy group resolution desired for the uranium calculations. Monte Carlo computer calculations have been compared with the discrete ordinates ONETRAN code results for simple cases to gain confidence in the results.

Preliminary calculations have been performed to investigate the applicability of transport and Monte Carlo computer calculations to uranium ore measurements. One of the initial problems is to determine the effect of rock composition, density, moisture content, and borehole diameter on the KUT gamma-ray spectra in the detector.

Feasibility studies are in progress to investigate the use of a photoneutron source for direct measurement of the uranium concentration surrounding a borehole. An ^{124}Sb -Be neutron source was used for the initial laboratory test experiments, and a ^4He gas proportional counter was used to count the induced prompt fission neutrons. More efficient fast-neutron detectors are being evaluated and Monte Carlo computer calculations are being used to optimize the source, shield, and detector parameters.

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A new approach is being investigated for producing photoneutrons by means of converting a beta-decay source to a bremsstrahlung spectrum. The high-energy portion of the spectrum then interacts with a beryllium sleeve to give the photoneutrons. This photoneutron source has the advantage of being a subthreshold neutron source with a convenient half-life. Also, by removing the bremsstrahlung conversion material, such as lead or thorium, the gamma-ray and neutron source can be partially turned off when not in use. Initial measurements have been performed using a ^{90}Sr beta-decay source combined with lead for the bremsstrahlung conversion.

I. GAMMA-RAY SPECTRA COMPUTER CALCULATIONS

(D. A. Close, M. L. Evans, and M. Jain)

In the previous internal progress report examples were given of gamma-ray calculations using Monte Carlo computer codes for boreholes of various diameters and for different rock compositions. It is clear that for repetitive calculations, a considerable savings in computer time can be achieved by using discrete ordinates computer codes.

To gain experience with the ONETRAN* discrete ordinates code applied to gamma-ray calculations, two calculations have been performed for comparison with identical Monte Carlo calculations. For these comparisons, a gamma ray having an energy of 352 keV was transported through dry sandstone ($\rho = 2.63 \text{ g/cm}^3$). This gamma ray is the second most intense gamma ray in the decay of ^{238}U and the most intense gamma ray having an energy less than 500 keV. It allowed the study of the relative importance of the photoelectric and Compton scattering cross sections for low-energy photons where the calculations might conceivably have been in error. Both the Monte Carlo and the few-group ONETRAN results were tabulated in eighteen 20-keV energy bins from 0.0 to 360.0 keV. Cylindrical geometry was considered, with one calculation having a vacuum outer boundary and the other having a reflecting outer boundary; no borehole was introduced into the geometry. Thus, the calculation employing the reflecting outer boundary simulates the infinite-media calculation asked for in Phase I of the calculational work statement. The calculation involving the vacuum outer boundary was performed as a check on our understanding of the code.

Figures 1 and 2 show the comparison between the Monte Carlo and the ONETRAN calculated flux (the number of gamma rays per square centimeter per second per source gamma ray) for the two cases. The Monte Carlo results are absolute; the ONETRAN results have been normalized to the Monte Carlo results in the 340- to 360-keV energy bin. These comparisons give us confidence that we

*See subsection A for a description of the steps involved in generating the ONETRAN spectra.

Fig. 1.

Comparison of gamma-ray spectra calculations using the ONETRAN discrete ordinates code with the Monte Carlo code for the 352-keV gamma ray from uranium decay.

understand the ONETRAN results for the Phase I type of calculations.

Work is continuing on the Phase III type of calculations where a borehole is introduced into the problem. Again, initial ONETRAN calculations are being done on the same 18-group problem described above. Currently, we are having difficulties understanding the ONETRAN results for the borehole problem. These problems may be caused

Fig. 2.
Comparison of ONETRAN and Monte Carlo gamma-ray spectra calculations for the 352-keV gamma ray in sandstone with a reflecting boundary.

by not using enough Legendre coefficients* in the cross-section angular distributions or by not using a high enough order for the angular quadrature.

When the above problem has been resolved, work will proceed quickly on generating the 265-group cross sections (many-group) for the elements contained in sandstone and shale.

*This problem is discussed in subsection B.

A. Structure for the Gamma-Ray Spectral Calculations

It is necessary to make modifications in the existing codes to give the gamma-ray energy resolution desired for the uranium ore spectra. The logical flow of the spectral calculations is depicted in Fig. 3. All computer codes are CDC 7600 FORTRAN-compatible. These codes (except the few-group ONETRAN code) are presently under development by LASL Groups T-1 and T-2 and have not yet been documented.

1. Code Descriptions

a. GAMINR. The GAMINR and CCCC codes originally were parts of a single code, but were modified so extensively (to provide many-group capabilities) that it was deemed more efficient to

Fig. 3.
Computer codes and the sequence used to calculate the gamma-ray spectra.

develop two independent codes. GAMINR takes point cross-section values for Compton (incoherent and coherent) scattering, the photoelectric effect, and pair production, and generates group-averaged cross sections. The angular dependence of the cross sections is expressed in terms of a Legendre polynomial expansion of a specified order.

b. CCCC. The CCCC code takes the group-averaged cross sections of GAMINR and arranges the transition elements into an isotope-ordered format, i.e., for problems involving mixtures of several isotopes (or elements), CCCC orders the cross sections by isotope. This cross-section file is the standard input for the few-group ONETRAN code (discrete ordinates code).

c. Many-Group ONETRAN Preprocessor. Because the many-group ONETRAN code requires group-ordered cross sections as input, a preprocessor code is required to change the cross section ordering from the isotope-ordered format of the CCCC output to the group-ordered format required by the many-group ONETRAN.

d. Many-Group ONETRAN. The many-group ONETRAN code is a modification of LASL's standard ONETRAN code (referred to earlier as the few-group ONETRAN). The code is a discrete ordinates finite element program for the solution of the one-dimensional multigroup (up to 400 groups) transport equation.

2. Calculational Procedure

The following steps will be followed in calculating the gamma-ray spectra of Phases I, II, and III:

a. For each elemental constituent of the uranium ore (B, C, O, Mg, Al, Si, K, Ca, Fe, U, and Th) a group-averaged, group-ordered cross-section file (preprocessor output) will be generated. Each element will be processed independently to avoid excessively long computer runs.

b. The files generated in (a) above will be combined into a single file to facilitate later processing. Once the master file has been created, no further cross-section manipulation need be done, unless additional elements are added to the ore composition.

c. The many-group ONETRAN code mixes the elemental cross sections in accordance with the

atomic densities particular to a given problem, and then solves the transport equation for the given problem geometry. Thus, for each problem (i.e., each combination of borehole and detector parameters) only a single ONETRAN computer run need be made; no additional cross-section manipulation is required (other than simple mixing).

B. Expansion of the Cross Sections in Legendre Polynomials

One of the computational problems in the discrete ordinates calculations is to decide upon the maximum order of Legendre polynomials to be used for the representation of the gamma-ray differential cross sections. We have written a program to calculate the Compton scattering differential cross section. The results are expressed in terms of coefficients of an expansion in Legendre polynomials (with $\cos \theta$ argument), where θ is the gamma-ray scattering angle in the laboratory. For a 2-MeV gamma ray the ratio of the coefficients for $P_1(\cos \theta)$ and $P_0(\cos \theta)$ is $1/12$, while for a 0.1-MeV gamma ray the ratio is $1/5000$. Therefore, it will require several investigative computer runs to empirically determine the balance between retaining higher order Legendre polynomials (thereby incurring longer ONETRAN execution times) and the required accuracy of the calculations. At this stage, it seems that the best solution would be to make the highest order of Legendre polynomials to be used a monotonically increasing function of the gamma-ray source energy. This requires creating cross-section files of sufficient Legendre polynomial order to adequately represent the maximum gamma-ray source energy. Then it would be necessary to vary the maximum order of polynomials as a function of energy in the ONETRAN code. We are currently investigating this procedure. It will probably be necessary to decide upon a suitable order of Legendre polynomials based on a reasonable compromise between numerical accuracy and speed of the calculations.

II. PHOTONEUTRON SOURCE EVALUATION FOR LOGGING

A. Feasibility Test for an ^{124}Sb -Be Based Instrument (H. O. Menlove and M. P. Baker)

Feasibility studies are in progress to investigate the use of photoneutron sources for direct

uranium ore grade determination. The principle of the method is to induce fission reactions in the ^{235}U surrounding the borehole with a sub-threshold source and to count the induced prompt fission neutrons with a fast-neutron detector. The detector is biased to count only neutrons above the interrogation source energy; thus no neutron source pulsing or modulation is necessary.

To investigate this technique, laboratory tests have been performed using an ^{124}Sb gamma-ray source ($\sim 1\text{ Ci}$) placed in the center of a 5-cm-diam by 10-cm-long beryllium metal cylinder to produce the neutrons. A 7-cm-long tungsten cylinder was positioned between the ^{124}Sb source and the fast-neutron detector to shield the detector from the direct source gamma rays. The available fast-neutron detector was an 18-atm ^4He gas tube 5 cm diam by 30 cm long. This detector type is very insensitive to gamma-ray background and can be operated in fields of 1-5 R/h with no gamma-ray shielding. The absolute neutron counting efficiency for this detector is relatively low ($\sim 0.5\%$). However, the neutron efficiency matches the shape of the prompt fission neutron spectrum as shown in Fig. 4, giving good intrinsic rejection of the lower energy (26-keV) interrogating neutrons. Figure 4 also shows the shape of the ^1H scattering cross section that is proportional to the efficiency of a liquid or plastic scintillator. These detectors have much higher counting efficiencies than the ^4He gas tubes but additional measures must be taken to suppress the gamma-ray sensitivity. Figure 5 shows a schematic drawing of the basic components for a photoneutron-based direct uranium logging system.

The present experimental test apparatus used no radial shielding on the neutron ^4He detector. To mock up borehole measurements, several 55-gal barrels of crushed sandstone ore ($\sim 0.2\%$ uranium) were obtained from the Kerr-McGee Corporation near Grants, New Mexico. Ten-centimeter-diam through-pipes were positioned on the center axis of the barrels to simulate boreholes. An additional barrel was fabricated with small tubes (1.5 cm diam) positioned at different radii out from the center hole as shown in Fig. 6. This

Fig. 4. Cross sections for fast-neutron detection with ^4He and liquid scintillator (^1H) detectors compared with the ^{235}U fission neutron spectrum.

barrel was filled with normal damp sand, and UO_2 reactor fuel rods were positioned at different radial locations to measure the penetration of the interrogating neutrons and the subsequent return of prompt fission neutrons to the detector. A set of 3 PWR-type fuel rods 120 cm long containing UO_2 with 3% ^{235}U enrichment were used for the radial penetration studies. The results of the measurements are given in Fig. 7. It can be seen that the penetration is satisfactory with the response falling off to one-half its initial value at a radius of 15 cm. Note that the neutron penetration in the present crushed sandstone is greater than would be expected for an ore body because of the lower density in the barrel and an actual borehole is likely to be filled with water. On the other hand, the relatively small size of the present barrel results in a large leakage of the source neutrons at the surface. Thus a larger ore body would result in a higher

Fig. 5.
Schematic diagram showing primary components of the photoneutron-based direct uranium logging instrument giving ranges of design parameters.

Fig. 5
End view of 55-gal barrel test apparatus used for initial evaluation of the photoneutron direct uranium logging method. The small radial holes are used to introduce uranium fuel rods at different radii.

Fig. 7.
Measurements of the prompt fission neutron response with the ^4He detector as a function of radius in the sandstone using the $^{124}\text{Sb-Be}$ photoneutron test apparatus.

neutron flux at the outer radial positions. Computer calculations are being made to better relate the present test setup with actual borehole conditions.

Previous computer calculations for damp sand ($\rho = 1.3 \text{ g/cm}^3$) and a 26-keV neutron source have shown that the thermal neutron flux peaks at about 22 cm from the center of the borehole. The induced fast fission neutrons then must travel back to the detector in the borehole, which shifts the response function to the general shape shown in Fig. 7.

To improve the counting efficiency, liquid scintillators are being obtained to replace the ^4He tube. The new detectors are expected to have more than an order of magnitude increase in counting efficiency.

R. Monte Carlo Computer Calculations for an Sb-Be Downhole Neutron Probe (M. L. Evans)

Computer calculations performed during this quarter have been directed toward determining the feasibility of using a liquid scintillator neutron probe. The crucial issue is whether adequate gamma-ray shielding can be provided for the scintillator under the constraints imposed by the borehole diameter, while maintaining sufficient scintillator volume to ensure adequate fission neutron counting rates.

Monte Carlo gamma and neutron codes have been used to estimate the intensities of the gamma and neutron fluxes for the probe shown in Fig. 5. The parameters used for the calculation were:

- probe diameter - 6.3 cm
- scintillator diameter - 1.8 cm
- scintillator length - 10 cm
- scintillator shield thickness - 2.25 cm
- height of cone shield - 10 cm
- Be source length (from apex of cone to neutron reflector) - 5 cm
- ore concentration - 2000 ppm U.

In addition, the computer model of the probe had no neutron reflector, and the gamma rays were emitted isotropically from a point source located along the beryllium metal cylinder axis 2.5 cm from the apex of the cone shield.

Estimates of the gamma and neutron fluxes incident on the liquid scintillator were obtained using three separate Monte Carlo codes.

The first was a gamma transport code that traced the 1.69- and 2.09-MeV gamma rays of ^{124}Sb from the center of the beryllium cylinder into the surrounding ore and probe. For a 1-Ci ^{124}Sb source, approximately $5 \times 10^4 \text{ } \gamma/\text{s}$ would be incident on the scintillator volume. In addition to providing an estimate of the gamma flux, the code was modified to provide an estimate of the prompt neutron flux created within the beryllium cylinder. For a 1-Ci source, approximately 1.5×10^7 neutrons/s are produced. Unfortunately, the gamma code does not have a feature whereby it can trace the neutrons produced by the $^{235}\text{U}(\gamma, n)$ reaction, and, consequently, a separate neutron code is used to start and trace the neutrons throughout the system. Thus, all information regarding the direction of the photoneutrons is lost, and the neutron code starts the neutrons isotropically from origins distributed uniformly throughout the beryllium cylinder volume. Using this technique, the photoneutrons (with energies of 26 keV and 563 keV) were traced throughout the probe-ore system, causing fissions of the ^{235}U in the ore. The code then traced the prompt fission neutrons back to the scintillator volume, resulting in an estimate of the fission neutron flux of 150 neutrons/s (in the energy range 0.5-20 MeV) for a 1-Ci ^{124}Sb source.

The third Monte Carlo code was used to estimate the neutron detection efficiency of the scintillator as a function of neutron energy. For the 1.8 cm by 10 cm liquid scintillator, the average neutron detection efficiency over the range 0.5-20 MeV is about 10% (with the energy bias at 0.4 MeV). For gamma rays with energies in the range 0-2 MeV, the scintillator should be about 5% efficient on the average. Thus, the expected gamma and neutron rates should be approximately $2.5 \times 10^3/\text{s}$ and 15/s, respectively, resulting in a γ/n ratio of about 150/1.

Experiments are being designed to verify these estimates and to evaluate different n- γ discrimination techniques. Previous experience indicates that neutron separation can be achieved for large γ/n ratios, so it should be possible to reduce the scintillator gamma shielding volume and thus increase the detection efficiency. With

increased neutron rates, the possibility of reducing the ^{124}Sb source strength to reduce health hazards in handling is also enhanced.

After the scintillator-based ^{124}Sb -Be laboratory system has been proven feasible experimentally, further calculations will be performed to optimize the probe configuration with respect to neutron rate, γ/n ratio, and ^{124}Sb source strength.

III. PHOTONEUTRON SOURCES USING BREMSSTRAHLUNG FROM THE β -DECAY OF ^{90}Sr (M. S. Krick)

Photoneutron sources are sources that produce neutrons via $(h\nu, n)$ reactions. Generally the $^9\text{Be}(h\nu, n)$ and $^2\text{H}(h\nu, n)$ reactions are used because the small negative Q -values (-1.67 and -2.23 MeV, respectively) permit the use of isotopic low-energy gamma-ray sources such as ^{124}Sb and ^{228}Th . Nondestructive assay instrumentation based on photoneutron interrogation has been investigated by the Nuclear Safeguards Research Group at Los Alamos for many years.^{2,3}

One difficulty with isotopic photoneutron sources is that the half-lives of the radioisotopes that emit gamma rays with energies above the (γ, n) thresholds are usually short; for example, the half-life of ^{124}Sb is 60.2 days. Longer-lived isotopes such as ^{88}Y ($T_{1/2} = 107$ days), ^{144}Ce ($T_{1/2} = 285$ days), ^{226}Ra (1620 yr), and ^{228}Th ($T_{1/2} = 1.91$ yr) are also useful or potentially useful sources. However, these isotopes are either not yet available in sufficient quantities for practical applications or are very expensive.

Bremsstrahlung sources are being considered for photoneutron production at LASL. An isotope that β -decays with an end-point energy above the photoneutron threshold can produce photoneutrons via $(h\nu, n)$ reactions from bremsstrahlung photons ($h\nu$). The neutron yield per beta decay is very low, since (1) only a small fraction of beta particles have energies above the photoneutron threshold and (2) only a small fraction of these beta particles will produce a photon above the photoneutron threshold. The thick-target bremsstrahlung energy distribution for monoenergetic electrons is⁴

$$\frac{dE}{d\nu} \approx C \left[Z(\nu_m - \nu) + 0.0025Z^2 \right],$$

where

$\frac{dE}{d\nu}$ is the bremsstrahlung energy per unit frequency,

Z is the atomic number of the target,

ν_m is the maximum photon frequency, and

C is independent of Z and ν .

Except when $\nu \approx \nu_m$, $\frac{dE}{d\nu}$ is proportional to Z .

The most promising isotope for a bremsstrahlung source is ^{90}Sr , since (1) the maximum beta-decay energy from the decay chain is 2.27 MeV (0.56 MeV above the $^9\text{Be}(h\nu, n)$ threshold), (2) the half-life is 28.1 yr, and (3) ^{90}Sr is available in large quantities as a by-product from nuclear fuel reprocessing operations. The useful beta particle is emitted by ^{90}Y in the decay chain

To evaluate a bremsstrahlung-based photoneutron source, a 92-Ci ^{90}Sr source was obtained in the form of 2 g of SrCO_3 , mixed with 40 g of lead and sealed in a cylindrical stainless steel capsule. Since lead is a high- Z element, its addition enhanced the bremsstrahlung production per curie of ^{90}Sr . The photon distribution was measured with a Ge(Li) detector and is shown in Fig. 8; the vertical line at 1.665 MeV shows the $^9\text{Be}(h\nu, n)$ threshold. The photoneutron yield was measured with a modified long counter and compared to the yield of an ^{124}Sb -Be source. A 7.50-cm-diam by 10-cm-high beryllium sleeve was used for both sources. The ^{90}Sr -Pb-Be source yielded 370 neutrons/s·Ci and the ^{124}Sb -Be source yielded roughly 3×10^6 neutrons/s·Ci; thus to produce a given neutron source strength about 8000 Ci of ^{90}Sr is required for each curie of ^{124}Sb .

For portable systems the shielding requirements for photon radiation are particularly important for photoneutron sources. The dose rate from 1 Ci of ^{124}Sb at a distance of 1 m in air was measured at 11 rem/h; the corresponding dose rate for 8000 Ci of ^{90}Sr -Pb is 125 rem/h. At a

Fig. 8
Measured bremsstrahlung photon energy spectrum from ^{90}Y mixed with lead using a Ge(Li) detector.

distance of 50 cm through 5 cm of lead the dose rates are ~ 0.2 rem/h for ^{124}Sb and ~ 3.5 rem/h for 8000 Ci of $^{90}\text{Sr-Pb}$. The lead is very effective in reducing the dose from the $^{90}\text{Sr-Pb}$ source because of the predominance of low-energy gamma radiation in the spectrum as shown in Fig. 3.

Since the bremsstrahlung dose rate is roughly proportional to the atomic number of the converter material (e.g., lead), the bremsstrahlung dose rate could be reduced somewhat by separating the converter from the source to partially turn off the gamma source. However, even a dilute

distribution of ^{90}Sr in a homogeneous matrix will have a bremsstrahlung yield only one or two orders of magnitude lower than that of a $^{90}\text{Sr-Pb}$ or $^{90}\text{Sr-U}$ source (per curie of ^{90}Sr). Thus, for a given neutron yield, a $^{90}\text{Sr-Pb-Be}$ source is more difficult to shield than an $^{124}\text{Sb-Be}$ source. Furthermore, the potential radiological hazard from a ^{90}Sr source is much greater than that from a ^{124}Sb source.

For portable systems it appears that ^{90}Sr bremsstrahlung photoneutron sources offer no advantage except long half-life over other photoneutron sources. If the half-lives of ^{124}Sb and ^{88}Y are unacceptably short, a $^{226}\text{Ra-Be}$ or $^{228}\text{Th-D}$ source would probably be preferable to a $^{90}\text{Sr-Sb-Be}$ source for portable systems.

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