27-84 JSD
1-25-84

I-12934

DR-2068-0

UCID-19964

FEASIBILITY STUDIES OF A CARBON/OXYGEN LOGGING
TOOL FOR EVALUATING THE CO₂ CONTENT OF THE
MEDIUM IN NUCLEAR DEVICE CONTAINMENT

H. I. West, Jr. and J. E. Glasgow

December 1983

MASLEN

Lawrence
Livermore
Laboratory

This is an informal report intended primarily for internal or limited external distribution. The opinions and conclusions stated are those of the author and may or may not be those of the Laboratory.

Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under Contract W-7405-Eng-48.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

UCID--19964

DE64 005227

FEASIBILITY STUDIES OF A CARBON/OXYGEN LOGGING
TOOL FOR EVALUATING THE CO₂ CONTENT OF THE
MEDIUM IN NUCLEAR DEVICE CONTAINMENT

H. I. West, Jr. and J. E. Glasgow

ABSTRACT

The feasibility of using oil-well bore-hole logging techniques for assaying the carbonate content of a potential shot site has been investigated. The procedure makes use of the detection of the 4439-keV γ ray of carbon and the 6130-keV γ ray of oxygen produced by the inelastic scattering of 14-MeV neutrons in the bore-hole medium. For the needs of the containment program, a sensitivity of detection of CO₂ to <0.02 weight fraction must be achieved. Laboratory measurements indicate that only with considerable development effort could a tool be made that would come close to achieving this goal.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

EnB

INTRODUCTION

To evaluate a potential shot site for containment one needs to know the carbonate content of the surrounding medium. In the past the containment experts at the Laboratory have relied upon chemical assays of bore-hole cuttings and side-wall samples which were reported as CO₂ content. Such an assay is a slow expensive process. It was felt that procedures based upon the detection of the 4439-keV γ rays emitted following the inelastic scattering of 14-MeV neutrons off ¹²C would serve as a sensitive measure of the carbon content of the medium. At the same time, one would measure the inelastic γ rays from ¹⁶O at 6130 keV and report the results as a carbon-oxygen ratio (C/O). Since the medium is almost universally 50% oxygen, such a normalization would eliminate the need for absolute measurements.

The containment experts require sensitivity of detection of CO₂ to 0.02 weight fraction and would like 0.01. Assuming a medium concentration of oxygen of 50%, these numbers can be viewed as weight ratios C/O of 0.012 and 0.006, respectively. In a medium of density ~2.0 gm/cm³, this means a sensitivity for carbon detection of 12 or 6 mg/cm³, respectively.

The approach described has been used in the oil-well logging business for a number of years by Dresser-Atlas,^{1,2} Schlumberger,^{3,4,5} Texaco,⁶ etc. The details of the logging tools are proprietary so that very little meaningful information is to be found in the literature. Possibly the most meaningful papers are those of Hertzog^{3,4} and of Westaway et al.⁵ describing work at Schlumberger. We use these papers as major references in this report.

Hearst⁷ recognized the possibility that the well-logging techniques could be applicable to the containment problem. He has talked with companies on several occasions. The people at Schlumberger said that in laboratory measurements they had achieved a sensitivity for the CO₂ weight fraction of 0.02, but in the field the best was 0.04. The logging systems, calibrated for oil-well work, are not adequately calibrated for our Nevada Test Site (NTS) application. The oil-well holes to be logged are typically 5" to 10" in diameter, often lined with iron pipe, and always filled with water to better define the region under neutron irradiation (also, they usually log below the water table). At the NTS holes are typically 5 to 10 ft in diameter and filling with water is not a realistic possibility (the water table is usually about 600 m). The oil-well logging companies that we felt might have the capability of doing the job at the NTS were not particularly interested in a job they believed was of limited economic opportunity. As a result, Hearst instigated the start of a logging project at LLNL. He did some feasibility studies⁷ and then proceeded to build a tool that possibly could be used for the experimental logging of holes at the NTS. However, details needed to be worked out regarding the data analysis.

At this point the first author (HIW) was asked to look into the problem of data analysis. The second author (JEG) had assumed the engineering responsibility of the tool (from R. Knipes) and later R. Bond and C. Mills, electronics technicians, were added to the project. Initially, we felt that the goals were to (1) get the present system working as well as

possible, (2) use it to define the new system to be built for field work, and (3) obtain data for working out the data analysis. After attempting to analyze the first set of data it became apparent that we needed to show that it was possible to achieve our goals. Thus, our efforts were redirected into obtaining proof of concept. We have studied the problem in some detail without having achieved the desired goals. It would probably be deceiving to say that we could never reach these goals, however, the developmental effort required, at this point, does not seem cost effective. It is the purpose of this report to document our efforts and results and show how seemingly similar systems in general used by other workers really have quite different goals.

THE TOOL

The tool usually consists of a pulsed-neutron generator separated from the detector by appropriate neutron shielding as shown in Fig. '. In oil-well logging the whole assembly is usually less than 5" in diameter. The neutron generator used in our early work gave us problems and was replaced by a generator built by the Sandia National Laboratories, Albuquerque, NM. The neutron shield between generator and detector in our case was of iron, lead, and finally an inadequate amount of borated paraffin as shown in Fig. 1. Probably a better design would be worthwhile. Typically, for the γ -ray detector, oil-well people use a 2.5" diam. by 4" long NaI(Tl) crystal, the diameter being small because of spatial limitations.

Fig. 1. Experiment block diagram.

With the tool in operation one typically pulses the neutron generator at 10 kHz. In the system described by Hertzog,^{3,4} the 14-MeV neutrons are confined to the first 18 μ s, the second 18 μ s period is used to obtain background information, and the 36-90 μ s period to obtain, primarily, capture data. From examining our data we find that there is not much decay in the thermal neutron flux (~10%) in the period between bursts, hence all data obtained after the neutron burst can be used to correct the inelastic period for contributions from capture, neutron-induced activity, and normal background. If the capture data are to be used for capture γ rays, then the contributions from induced activity and normal background must be determined by turning the generator off for, say, 1 second possibly every 10 seconds. We had no interest in using the capture data in assaying the medium and did not pursue this method although, in retrospect, we should have done so. To follow historical precedent, we collected data in three periods; we have inelastic (I), background (B), and capture (C), even though no real differentiation is to be made between the last two. In principle, one should consider using the capture period for elements producing γ rays of unique energy thus allowing these elements to be assayed, and on the basis of such information, determine the amount of elastic spectrum for the element to be subtracted. We have not proceeded to this level of sophistication.

All of the data discussed here made use of the Sandia neutron generator. Initially we had some problems with electronic noise from the generator. However, we finally were able to reduce the noise to an acceptable level, part of which was achieved by gating off the PHA for the first 2.0 μ s after the start of burst time. Figure 2 shows the relative fast-neutron output of the neutron generator determined using a plastic

Fig. 2. Output of Sandia neutron generator.

scintillator for a monitor. Here we counted the knock-on protons in the scintillator. The pulse-height distribution was roughly exponential, trailing off towards high energies. To get good data, required good stability of the pulse-height discrimination for the period of data taking. It would seem from Fig. 2 that we have a few fast neutrons for the total period, although it may be that after about 40 μ s we were counting capture γ rays. In any respect, the contribution from fast neutrons after 30 or 40 μ s is low. For the counting periods, we established 2-32 μ s for the inelastic (I) period, 34-64 μ s for the background period (B), and 66-96 μ s for the capture (C) period. The B and C periods contained much the same data and were added together in the final data analysis.

Detector

To simplify data analysis, the detector should absorb a significant number of γ rays passing through it and should have a distinctive energy response as shown, eg., by the scintillation detector. Typically, we would like the photo- or full-energy peak in the spectrum to be a significant part of the total and be narrow. With the high energies involved here, we always have pair effect with the subsequent formation of two annihilation quanta of 511 keV per event; peaks are formed at $E_\gamma - 1022$ keV (double escape) and $E_\gamma - 511$ keV (single escape). A large NaI(Tl) crystal increases the capture of annihilation radiation, thus, enhancing the full-energy peak. We considered using the high-Z high-density scintillator bismuth germanate ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$, known as BGO). The light output⁸ from BGO is only 8% of NaI(Tl) and the energy resolution is reduced accordingly. However, Harshaw Chemical Co. has talked about an energy resolution of 8.9% for 662 keV, which if attainable in large

crystals could prove interesting. In any respect, we have not tried this detector.

Obviously, the use of a high-resolution detector such as lithium-drifted germanium (Ge(Li)) would be advantageous. For example, Sowerby⁹ has used Ge(Li) for assay in the mineral industry; Senftle, Moxham, and Tanner¹⁰ have used Ge(Li) for logging minerals in a down-hole environment; and, Wormald and Clayton¹¹ have used Ge(Li) to analyze coal for trace elements. The list is by no means complete. Figure 3 shows a spectrum obtained in our Laboratory by H. Spracklin using a Pu-Be source and a 10% Ge(Li) detector (means 10% as efficient for ^{60}Co γ rays as is a 3" x 3" NaI(Tl) detector). The Pu-Be source excites the ^{12}C 4439-keV level through the $^{10}\text{Be}(\alpha, \text{nn})^{12}\text{C}^*$ reaction using ^{238}Pu α 's. In Fig. 3 we note the high resolution of the lower-energy peaks whereas the full-energy and first and second escape peaks are about 120 keV in width (full width at 1/2 maximum). Most of this width is due to the Doppler shift of the γ rays coming from the recoiling $^{12}\text{C}^*$ nuclei. Figure 4 shows data we acquired in our logging system using our carbon calibration box. Here the peaks are about 80-keV wide. When we obtained spectra for limestone, almost no carbon signal was discernable above the continuum of scattered γ rays. Obviously, Ge(Li) is not suitable for our application. This may seem to be contradictory to the results of Wormald and Clayton,¹¹ but we must remember that they were looking for trace elements in the presence of coal. In their spectra the carbon peaks show nicely and even show narrow peaks produced by other elements on top of the wide carbon peaks.

Since all of the γ rays of interest are many MeV in energy, the pair cross section in NaI(Tl) is comparable to or larger than the photoelectric cross section. This prompted us to look into the possibility of using the

Fig. 3. Germanium spectrum of $^{12}\text{C}^*$ 4439-keV γ ray from a Pu-Be source.

Note the width of the carbon peaks relative to the much narrower peaks at low energy.

Fig. 4. Germanium spectrum of $^{12}\text{C}^*$ 4439-keV γ ray excited with 14 MeV neutrons on carbon in our standard tool configuration.

scintillation pair spectrometer because it produces a unique pulse height ($E_\gamma - 1022$ keV). Here for each event one detects, in two or more side crystals, the two 511-keV annihilation quanta escaping a central convertor crystal. We tested such a system, but even though we used four side crystals to enhance the detection of the annihilation quanta, we found the efficiency too low for our needs.

From our considerations we are led inextricably back to the single NaI(Tl) detector used in the oil-well logging business. At first we used a 6" x 6" polycrystalline scintillator (made up of a number of smaller crystals in a liquid of the same refractive index) because of the concern of thermal-stress fracture. However, this crystal gave only ~10-11% resolution for the 662-keV γ ray of ^{137}Cs . We needed better resolution, and hence, obtained a 5" x 5" crystal of 8% resolution. This crystal, fitted with extra magnetic shielding and a thermal blanket of polystyrene, was used to obtain the spectra presented here. Figure 5 shows a typical Pu-Be spectrum. We note the marked enhancement of the full-energy peak relative to that obtained with Ge(Li) and the almost nonexistent double-escape peak. Figure 6 shows a spectrum of the ^{16}O γ ray at 6130 keV obtained with a Cm-C source. The γ rays are produced from the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction using ^{244}Cm α 's.

Detector Electronic System

The electronics are as shown in Fig. 1. We concentrate on aspects of the detector system. Good circuit practice in all aspects of the counting system are essential, and in some cases only state-of-the-art circuitry will do. The preamplifier is essentially an emitter follower chosen for its wide dynamic range. Since pulse clipping does not occur until the signal reaches the final stages of the linear amplifier, we must allow for

Fig. 5. NaI(Tl) spectrum of $^{12}\text{C}^*$ γ rays from a Pu-Be source. The crystal was 5" x 5".

Fig. 6. NaI(Tl) spectrum of ^{160}Gd γ rays obtained using a Cm-C source.

This spectrum shows primarily the 6130-keV γ ray and first escape peak and is quite different from the spectrum obtained with 14-MeV neutrons on water.

pileup of one or more pulses on the tails of preceding pulses. The pulses out of the linear amplifier are RC clipped and integrated so that they are essentially over by 2 μ s but unfortunately there is always a significant residual in the pulse tails (equivalent to a few channels) to about 5 μ s. Possibly the use of Gaussian pulse shaping may have helped to reduce the tail; the work reported by McQuaid et al.¹² is of particular significance in this respect.

The routing of the digitized signals to the appropriate parts of the analyzer memory was accomplished by the Gated Analog Router (Nuclear Data 569 GAR). The GAR also determined the lifetime of each of the counting channels and routed this information to the analyzer preparatory to readout to floppy disk. The analyzer readout was controlled by a LSI-11/2 microcomputer. Since the work occurred at the NTS, the floppy disks were then read at an appropriate terminal and the information stored in long-term memory at LLNL preparatory to further analysis. The data had to be converted to conventional format for further processing.

Problems with counting rate occurred. As the counting rate increases to about 10% livetime, the spectral peaks begin to shift towards higher energies (a photo multiplier problem). Experimental evaluation of this effect is shown in Fig. 7. We settled upon 512 channels per spectrum using an energy scale of 20 keV/channel or 10,240 keV full scale. As we proceeded from one sample box to the next, count rate shifts at eg. channel 300 could be 10 channels. We learned to take care of such shifts in data analysis. Our other problem was with pulse pileup. At the outset we recognized the pileup problem. Unfortunately, the pileup rejector we located here at the Laboratory was designed for Ge(Li) use and did not work well for the narrower NaI(Tl) pulses. (Apparently vastly improved

Fig. 7. NaI(Tl) spectrometer drift tests. The tests were made with a Pu-Be source of 4439-keV γ rays by varying the source to detector distance.

systems are now available commercially, eg. the EG&G-ORTEC 673 Amplifier/-Gated Integrator containing pile-up rejection.) Rather than pursue the pileup rejector further (at this time at least) we resorted to using low counting rates, i.e. a PHA dead time of no more than 5%.

For comparing various data runs it is desirable that we have a good measure of the relative neutron fluxes. The use of a plastic scintillator as a neutron monitor was discussed earlier. We could have made good use of a slow-neutron counter but never got one into operation. Obviously, a $^{10}\text{BF}_3$ proportional counter would have done the job. Also, we considered ^6Li -glass scintillators. In this respect, a ^6Li -depleted scintillator, containing primarily ^7Li , would work for fast neutrons but does not produce a good plateau for counting as does $^{10}\text{BF}_3$ and ^6Li . Fortunately, the neutron generator was stable and results were reproducible over periods of a few days. Ion current and high voltage (HV) were monitored but it was the HV that actually set the neutron flux. For the purposes of this report, the generator was operated at an output flux of 0.5 to 1.0×10^7 neutrons/s. Typical counting times were 20 min, corrected for PHA dead time.

Obviously, the electronics system that we have been using in a laboratory environment is not entirely adequate for down-hole work. As done by other workers we would need the linear amplifier, PHA, pulse routing, and HV in the down-hole tool. Buffered data would be sent up-hole to an on-line computer designed to store and process (at least partially) the data and further to control down-hole operations.

Detection Efficiency

We need an equation for the quantitative determination of γ -rays production and detection. We make use of the geometry shown in Fig. 8. The neutron flux (ignoring multiple scattering) at x , y , z is

$$\frac{dN_n}{dt} = \frac{e^{-\mu_n(r_1-r_{01})}}{4\pi r_1^2}$$

in which $\frac{dN_n}{dt}$ is the neutron source strength and μ_n is the linear absorption coefficient for fast neutrons. For our purposes it seems easier to stay with Cartesian coordinates because of programming ease, despite the obvious symmetry in cylindrical coordinates. We immediately write down the γ -ray flux at the detector

$$F_\gamma = \iiint \left\{ \frac{dN_n}{dt} \frac{e^{-\mu_n(r_1-r_{01})}}{4\pi r_1^2} \right\} \left\{ \frac{e^{-\mu_\gamma(r_2-r_{02})}}{4\pi r_1^2} \right\} \left\{ n_a \sigma_a dx dy dz \right\}.$$

We note that in the third term n_a = density of nuclei to be excited (eg. carbon), σ_a = cross section (for carbon a threshold of ~4.5 MeV with cross section averaging ~250 mb), and $dx dy dz$ = volume element. Hence, the third term is the effective area of interaction of the nuclei, which divided by 1.00 cm becomes a probability. The second term is the probability that a γ ray produced in $dx dy dz$ gets to the detector. Substituting for the r terms, our equation becomes

$$F_Y = \frac{1}{8\pi^2} n_a \sigma_a \frac{dN}{dt} \int_{z_o}^{\infty} \int_{-\infty}^{\infty} \int_0^{\infty} \frac{e^{-\mu_n [\sqrt{x^2 + (y-y_o)^2 + z^2} - \sqrt{x^2 z_o^2 / z^2 + (y-y_o)^2 z_o^2 / z^2 + z_o^2}]} }{x^2 + (y-y_o)^2 + z^2} \cdot$$

$$\frac{e^{-\mu_Y [\sqrt{x^2 + (y+y_o)^2 + (z+z_{oo})^2} - \sqrt{x^2 z_o^2 / z^2 + (y+y_o)^2 z_o^2 / z^2 + (z_o + z_{oo})^2}]} }{x^2 + (y+y_o)^2 + z^2} \cdot dx dy dz$$

Here we have allowed for symmetry between the plus and minus x axis. A small computer code (40 lines of FORTRAN) was written to evaluate the integral. We then have

$$F_Y = \frac{1}{8\pi^2} n_a \sigma_a \frac{dN}{dt} F_{NY}(\mu_n, \mu_Y, \text{geometry}) \quad \gamma \text{ rays/cm}^2\text{-s,}$$

in which F_{NY} represents the integral.

Table 1. Values of F_{NY} for 4500-keV γ rays and various geometries.

Dimensions (cm)			$F_{NY} \text{ (cm}^{-1}\text{)}$			
$2y_o$	z_o	$z_o + z_{oo}$	Carbon	CaCO ₃	H ₂ O	SiO ₂
58.4	6.35 ^a	10.8	0.0171	0.0104	0.00951	0.00105
43.2	6.35	10.8	0.0298	0.0199	0.0174	0.0197
43.2	16.5	21.1	0.0197	0.01386	0.0123	0.0140
43.2	21.6	26.1	0.0155	0.0110	0.0975	0.0116

^aNote that the center of the neutron source is located 6.35 cm in from the outer edge of the tool.

The results of this simple calculation have worked out surprisingly well in predicting absolute counting rates and variations in geometry. The results support the supposition that most inelastic collisions, leading to high-energy excitation, occur only once during a neutron lifetime and that the neutron inelastic-scattering cross section is adequately represented as independent of energy. We have also assumed that μ_n is independent of energy. To do otherwise means that we must first determine the neutron spectrum as a function of position in the medium. We note that the γ rays, when scattered, lose energy and cannot contribute to the full-energy peak; thus, the simple γ -ray absorption equation is quite accurate for the full-energy peak, but not for the continuum where multiple scattering is important. To get results better than these, we must resort to neutron-photon transport codes such as the MORSE and TART codes available at LLNL. These are complex codes to run.

DATA

Calibration test data were acquired by suspending the tool horizontally over sample boxes 6 ft x 6 ft filled to a depth of 4 ft. Table 2 shows the contents of the sample boxes; the carbon content of each sample was determined from chemical assays.

Table 2. Sample box composition.

Material	Chemical composition	Density (gm/cm ³)	Density of carbon (gm/cm ³)	CO ₂ weight fraction
Water	H ₂ O	1.00	0.0	
Coke	C	0.95	0.95	
Limestone	CaCO ₃	1.73	0.20 ^c , 0.21 ^b	0.44 ^a
30% limestone & 70% sand	CaCO ₃ /SiO ₂	1.88	0.067 ^c , 0.059 ^b	0.1145 ^a
15% limestone & 85% sand	CaCO ₃ /SiO ₂	1.76	0.033 ^c , 0.028 ^b	0.058 ^a
Sand	SiO ₂	1.78	0.0012 ^b	0.0025 ^a

^aSampling by W. McKinnis of LLNL-NTS.

^bDensity inferred from CO₂ measurements and total densities.

^cSampled by H. I. West and assayed by L. Gregory of Analytical Chemistry Section, LLNL.

The samples not only allowed testing for various levels of concentration of carbon but they allowed determining response functions by subtracting one spectrum from the other. Water is unique. Hydrogen produces only capture γ rays (2225 keV) and their contribution is readily subtracted from the I-spectrum to provide the oxygen spectrum of inelastically-scattered γ rays. The oxygen spectrum can then be subtracted from the sand I-spectrum for the silicon response. In principle, we could have used a box of CaO or Ca(OH)₂ to determine the calcium spectrum, but both are somewhat caustic requiring proper handling. In this study, it was sufficient to use the residual left over from the analysis of the limestone data for the calcium spectrum.

Elemental spectra generated during the data analysis discussed later in this report are shown in a linear-log plot in Fig. 9.

Westaway et al.⁵ show typical spectra obtained at Schlumberger for oxygen, silicon, iron, calcium, sulphur, and hydrogen during both the inelastic and capture periods. The spectra give a good overview and would be reproduced here except for copywrite problems. An important point can be made here. For the inelastic period, the iron and calcium spectra are very similar, making the analysis for separating their contribution almost impossible. In their capture spectra, however, we find marked differences. Thus, to make full use of the logging capabilities one needs both sets of data.

General Analytical Procedure in Data Analysis

Let the spectrum of a given element be written as a column vector

$$\{a_{11}, a_{21}, a_{31}, \dots, a_{n1}\}.$$

If x_1 is the amount of this spectrum in the overall spectrum, then $a_{11}x_1, a_{21}x_1, a_{31}x_1, \dots, a_{n1}x_1$ are the contents of channels 1 through n; here the a_{ij} terms are normalized* such that $\sum a_{ij} = 1$. Writing out the overall elements of the spectrum explicitly we have

$$\begin{pmatrix} y_1 \\ y_2 \\ y_3 \\ - \\ - \\ y_n \end{pmatrix} = \begin{pmatrix} a_{11} & a_{12} & \dots & a_{1n} \\ a_{21} & a_{22} & \dots & a_{2n} \\ a_{31} & a_{32} & \dots & a_{3n} \\ - & - & - & - \\ - & - & - & - \\ a_{n1} & a_{n2} & \dots & a_{nn} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ - \\ - \\ x_n \end{pmatrix}$$

*As used in this report, the sum of the a_{ij} are not normalized. The spectra used could have been divided by a normalizing factor to achieve this end, but it was not necessary.

Fig. 9. Standard spectra used in this report.

The contents of each energy channel may be written as

$$y_i = \sum_{j=1}^m a_{ij} x_j$$

in which we have m unknown elements to be determined. In more general notation we have

$$\bar{Y} = \bar{A} \bar{X}$$

in which $\bar{A}$ is $m \times n$ in dimension.

The treatment of the response matrix $\bar{A}$ and the data $\bar{Y}$ to obtain $\bar{X}$ is a highly studied problem in applied mathematics, which yields a solution through application of the methods of least-squares analysis. Hertzog briefly discusses the methods used at Schlumberger. The group at the National Reactor Testing Station^{13,14} have developed a code for the general analysis of spectra from NaI(Tl) spectrometers. Schonfield and associates^{15,16} at Oak Ridge National Laboratory have applied the methods to some specialized problems. At LLNL we have a computer utility routine, MLR, which handles the problem in general. However, there is a vast bookkeeping job in applying MLR to our job. R. Gunnink and coworkers at LLNL (private communication, 1983) have developed a code to do this called NAIGAM. This code is not documented. We have not gotten to the point in our work where we could make proper use of this code.

Before using any of the more general methods, we need to understand the details of the spectra. For this we have resorted to the peel-off method. One starts with some readily identifiable, hopefully unique, feature in the spectrum due to a given element. A spectrum of that element is subtracted until no evidence of it remains. Writing out term by term, we note that after the first peel-off our matrix equation becomes

$$\begin{pmatrix} y_1 \\ y_2 \\ y_3 \\ \vdots \\ y_n \end{pmatrix} = \begin{pmatrix} a_{12} \dots a_{n1} \\ a_{22} \dots a_{n2} \\ a_{32} \dots a_{n3} \\ \vdots \\ a_{n2} \dots a_{nm} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ \vdots \\ x_m \end{pmatrix}$$

One then proceeds to peel off the next spectrum, etc. The formal matrix representation that follows each step in the peel-off should be obvious.

Interactive Computer Code

In order to understand the details of the spectra, with the view towards eventual use of MLR response-matrix techniques, we needed to manipulate the various spectra readily -- that is, add and subtract spectra and display their results on the TMDS. It is only through an interactive procedure that one can obtain the knowledge necessary for telling the computer how to do the job. Coding evolved as the level of our needs increased. The capabilities of the last code PSP are described.

Interacting with the teletype, the operations are as follows:

- Select TMDS and RJET, display and output terminals.
- Read in up to 4 files of 512 or 1024 channels each.
- Type label.
- Option level 1.
 - Select plots linear-linear or linear-log with any scale.
 - Read out files as needed and read in files.
 - Restore modified data arrays.
- Option level 2 under linear-linear plots.
 - Select active data array.
 - Smooth data using piecewise continuous MLR fit to data.
 - Correct spectra for energy scale and zero offset.

- Integrate selected parts of a spectrum.
- Fit Gaussian-minus-straight-line to spectral peaks for centroid and area determination.
- Add and subtract spectra.
- Output TMS plots.

DETAILS OF A DATA ANALYSIS

We discuss the last data set analyzed. Our goal in the analysis was to obtain the carbon content of each sample, or viewed more completely, to obtain the carbon-oxygen ratio. We had gone through a variety of geometry changes and attempts to improve the spectra. The separation of generator and detector (measured to the center of the detector) had been changed from 23" (58.4 cm) to 17" (43.2 cm), increasing the counting rate by x2. However, the continuum in the spectra was markedly increased. It was found that by raising the tool to 4" to 10" above the samples that the continuum was minimized. For the spectra discussed here, the tool was 4" to 6" above the sample. Each spectrum required a 20-min acquisition time, corrected for the dead time of the PHA. The same neutron generator settings were used for each run to insure equal neutron fluxes.

Energy Scale

In setting up the analyzer energy scale and zero offset, we made use of radioactive sources ^{137}Cs (662 keV), ^{60}Co (1173, 1333 keV), Pu-Be (4439 keV) and Cm-C (6130 keV). (See Ref. 17 for the γ -ray energies used in this report.) However, there was always enough energy shift from one sample to the next that we had to rely upon peaks internal to the spectra. These peaks are 1779 keV ($^{28}\text{Si}(n,n')^{28}\text{Si}^*$ and $^{28}\text{Si}(n,p)^{28}\text{Al}$ β^- 2.3 min $^{28}\text{Si}^*$); 2225 keV ($^1\text{H}(n)^2\text{H}^*$); 4439 keV ($^{12}\text{C}(n,n')^{12}\text{C}^*$); and 6130 keV ($^{16}\text{O}(n,n')^{16}\text{O}^*$) in which the γ ray comes from the de-excitation

of the starred product. To find the centroid of the peaks we made use of the MLR fitting routine described in the PSP computer code. The code was then used to correct all spectra to zero offset and an energy scale of 20 keV/channel. Although the sensitivity of peak determination was usually less than 0.1 channel the overall accuracy was more like ± 0.1 channels.

Tool Response

Assume for the moment that we are obtaining data from the carbon box. The γ -ray spectrum conceivably is made up of the following parts:

- (1) γ rays produced by neutrons interacting directly with the carbon and proceeding towards the detector, with or without scatter in the medium.
- (2) γ rays produced by neutrons that had first scattered off the tool and then to the carbon.
- (3) γ rays produced in the tool by neutrons whose total history was in the tool.
- (4) γ rays produced in the tool from neutrons that had first interacted with the carbon and scattered back into the tool.

Clearly (1) and (3) are the major contributors to the γ -ray spectrum. If (2) and (4) are important, we must use a response-function procedure as discussed under the general analytical procedure for data analysis. If capture data are important, (2) and (4) probably are significant. However, for the inelastic data our experience indicates that they are not an important contribution.

For the inelastic spectra, we can get a reasonably good tool spectrum by moving the tool well away from interacting material; 10 ft seemed to be enough. A spectrum made of data from two different runs is shown in Fig. 10. Here we note that despite the general continuum we have a small peak at channel 222, the position of the carbon peak.

Fig. 10. Response of the tool and detection system to neutrons from the generator. The data were acquired with all interacting material at least 10 ft away. Data for runs on two different days are summed here. A value of 1.0 tool response, as referred to in this report, is 0.5 of this spectrum.

Carbon Spectrum

The upper curve of Fig. 11 shows $I - 0.5 (B+C)$ for the carbon sample minus the tool response. It would appear that some of the contribution to the spectrum near channel 111 is due to uncompensated hydrogen-capture from the B+C sample. This can only be partially true since no combination of (B+C) would remove more than a small amount from that shown. In the spectrum we note the full-energy peak centered at channel 222, the first escape at channel 197 and the weak second escape at about channel 171. The peak at $\sim$ channel 345 is not understood.

The bottom curve is the result of removing $0.213 \times \text{Pu-Be}$ spectrum. The factor has only relative significance. The same Pu-Be spectrum was subtracted from the other carbon-containing samples to obtain a relative measure of their contents. One will note in the neighborhood of the peaks that there are some structures that are nonstatistical. Counting rates were low so that it is hard to believe that they resulted from pile-up effects. In any respect, they represent the differences that can result even though great care was exercised in establishing the energy scales.

The overall continuum left over after the subtraction is not understood. Even the original Pu-Be spectrum showed a continuum, though smaller. Below the spectral peaks we should expect a continuum due to the scatter of the 4439-keV γ rays, but that at the higher energies must be due to aspects of the nuclear reactions not understood by the authors.

Oxygen Spectrum

Figure 12, lower curve, shows background (B) plus capture (C) data obtained from the water sample. The upper curve shows the inelastic data. We note in the B+C spectrum a bit of structure at channel 306 probably corresponding to the ^{16}O γ ray of 6130 keV. Most likely a few

Fig. 11. Carbon spectrum. The upper curve shows the inelastic data (I) minus 0.5 of the background-plus-capture data minus 1.0 tool response. The lower curve is the result of subtracting the Pu-Be spectrum of Fig. 5.

Fig. 12. Water spectrum. The upper curve is the uncorrected inelastic spectrum (I). The lower curve is the sum of the background (B) and capture data (C); it probably contains a small contribution due to inelastic scattering.

14-MeV neutrons continued to come from the Sandia neutron generator for the entire 100- μ s period. The most prominent γ ray in both spectra is from hydrogen capture at channel 111. Figure 13 shows the I spectrum with 0.55 (B+C) spectra subtracted.

The bottom curve of Fig. 13 shows the data corrected for 1.00 tool response of Fig. 10. As discussed in the last section, there was some concern as to how much tool response should be subtracted. However, the general consistency of the data strongly supports the 1.0 value used.

Engesser and Thompson¹⁸ have published spectra for a 4" x 4" NaI(Tl) crystal which would tend to support the 1.0 factor. More importantly though, they have published results for a number of elements pertinent to down-hole logging. They have attempted to associate the peaks in the spectra with the appropriate nuclear reaction and level de-excitation and have provided production cross sections. Whether their assignments are correct or not is not important for the purposes here, at least as long as the element (not isotope) is properly identified. For the $^{16}\text{O}(n,n')^{16}\text{O}^*$ reaction we have γ rays at 7117, 6919, 6130 (prominent), 2768 keV and lower. $^{16}\text{O}(n,\alpha)^{12}\text{C}^*$ gives a weak γ ray at 4439 keV and $^{16}\text{O}(n,\alpha)^{13}\text{C}^*$ γ rays at 3850, 3680, and 3090 keV (the energies are updated from Ref. 17). Production cross sections are 0.5 to 12 mb/str at 90° with most at ~ 4 mb/str.

Sand Spectrum

Figure 14 shows the capture corrected inelastic data, which in turn is corrected for tool response and oxygen spectrum to give the inelastic spectrum for silicon. Engesser and Thompson¹⁸ attempt to assign a number of transitions in the decay of $^{28}\text{Si}^*$ to their spectra. Our peaks do not show as clearly as theirs. To the left of the plot we have the 1779-keV γ

Fig. 13. Oxygen spectrum obtained from the water data. The upper curve is from $I - 0.55 (B+C)$ for the water data. The lower curve is the result of subtracting 1.0 tool response.

Fig. 14. Sand Spectrum. Curve A is the $1 - .5 (B+C)$ spectrum of SiO_2 . Curve B is the result of subtracting 1.0 tool response. Curve C is the result of subtracting 0.80 of the oxygen spectrum of Fig. 13. (The 1779-keV γ -ray peaks at $\sim 16 \times 10^3$ on Curve C.) Curve C is the resultant silicon spectrum.

ray from both the $^{28}\text{Si}(n,n')^{28}\text{Si}^*$ and $^{28}\text{Si}(n,p)^{28}\text{Al}$ 2.23 min $^{28}\text{Si}^*$ reactions, which shows strongly but the rest are weak. A large number of transitions to at least 8000 keV occur. Results that can be reasonably well identified at the higher energies are the peaks at 2838, 4498, and 5100 keV. The peak at 4498 keV is close to that of carbon and must be allowed for in our data analysis.

100% Limestone Data

Figure 15 shows composite spectra for the limestone sample. Curve A is the inelastic spectrum from which in turn is peeled off the tool spectrum, the oxygen spectrum, and carbon spectrum.

In principle, the factor used for the oxygen spectrum should be 1.0 when allowances are made for differences in material density and neutron scattering cross sections between water and CaCO_3 . This assumes that the neutron generator was operated at the same level (within the limit of its settings). Calculation shows that a 2.0 inch difference in tool-sample distance would produce the difference.

Curve D, the result of subtracting the carbon spectrum of Fig. 11, is primarily due to calcium. The peaks at 3736 and 3920 keV are identified with the $^{40}\text{Ca}(n,n')^{40}\text{Ca}^*$ reaction. The remainder of the spectrum is still to be understood, but is believed to be due almost entirely to calcium.

Figure 16 shows the same limestone data using smoothing. The spectra were fit piecewise continuous to a 3-term power series, 5 channels at a time. More extensive smoothing (more channels in the fit) could be done but the procedure does not seem to produce much good and statistical information is lost.

Fig. 15. 100% limestone data. Curve A is $I - 0.5 (B+C)$. Curve B is $A - 1.0$ tool response. Curve C is $B - 0.8$ of the oxygen spectrum (Fig. 13). Curve D is $C - 0.145$ of the carbon spectrum of Fig. 11. The resultant is attributed to calcium and is used in the analysis of the 30% and 15% limestone data.

Fig. 16. 100% limestone data. The data of Fig. 15 are smoothed. The
I - .5 (B+C) spectrum, tool response, oxygen spectrum, and carbon
spectrum were each smoothed before peel-off was carried out.

30% Limestone Data

Figure 17 shows our analysis of the 30% limestone-70% sand mixture. Starting with Curve A, oxygen (0.80), silicon (0.65 ± 0.03), calcium (0.25), and carbon (0.06 ± 0.02) were subtracted. Oxygen, of course, has a distinctive spectrum which makes its subtraction not too subjective. For silicon subtraction, we made use of the intense peak at 1779 keV. There is not much structure in the calcium spectrum at the higher energies. Below 2 MeV there are peaks that should prove useful but did not net very much. Hence, the calcium subtraction was subject to some uncertainties. By subtracting the calcium spectrum to reduce the spectrum above approximately channel 222 of the carbon peak to about zero, we found a residual below channel 222 characteristic of the carbon peak. The degree of peel off of these two spectra was then adjusted for best fit.

There did not seem to be any significant systematic problems so it seemed appropriate to analyze the calcium and carbon as if they were the only remainder. In effect we can write

$$\begin{pmatrix} y_1 \\ y_2 \end{pmatrix} = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \end{pmatrix}$$

We let y_1 be the sum of the counts in channels 140-240, and y_2 the sum of channels 241-341. The column vector $\{a_{11}, a_{21}\}$ is from the calcium spectrum of Fig. 15, and $\{a_{12}, a_{22}\}$ from the carbon spectrum of Fig. 11. Then x_1 and x_2 are the respective amounts of the calcium and carbon spectrum. We obtained 0.296 for calcium and 0.0486 for carbon. The statistical errors in the numbers are but a few percent but a comparison of these numbers to the earlier numbers gives us a better indication of the error.

Fig. 17. 30% limestone-70% sand data. Curve A is $I - 0.5 (B+C) - 1.0$ tool response. Curve B is $A - .80$ of the oxygen spectrum. Curve C is $B - 0.65 \pm .03$ of the silicon spectrum of Fig. 14. Curve D is $C - 0.25$ of the calcium spectrum of Fig. 15. Curve E, averaging approximately to zero, is $D - 0.06 \pm .02$ of the carbon spectrum of Fig. 11.

15% Limestone Data

Figure 18 shows the results of analyzing the 15% limestone-85% sand data. Starting with the fully corrected inelastic data of Curve A, we have in turn, (B) the subtraction of 0.80 of the oxygen spectrum, (C) 0.85 ± 0.01 of the silicon spectrum, (D) 0.1 ± 0.05 of the calcium spectrum, (E) $0.03^{+0.15}_{-0.03}$ of the carbon spectrum.

Curves D and E result from the subtraction of small amounts of the calcium and carbon spectra. Here the curves run together but there is little point in showing them separated. Actually the results are consistent with zero carbon contribution. However, we can analyze the calcium-carbon remainder as we did for the 30% limestone sample by summing channels 140-240 and 241-341. For calcium we obtain 0.0817 and for carbon 0.0434. The statistical errors are but a few percent but the actual error is much higher.

Carbon Detectability

Figure 19 shows the composite results of the carbon analysis. The ordinate is given as a fraction of the carbon spectrum (Fig. 11) with density corrections made, correcting the limestone results to the case of pure carbon (x1.40). The density corrections are based on the theory presented earlier (see the 17" (43.2 cm) results of Table 1).

A procedure based on the peel off of the Pu-Be spectrum was used in early analyses. For this approach, after the oxygen contribution was subtracted, the carbon peak at channel 222 was reduced to zero by the subtraction of the Pu-Be spectrum. This gave results completely consistent with the present results down to concentrations the order of the 30% limestone mixture. Below that there were problems because of the silicon contribution. However, for quick results the subtraction of a carbon spectrum following the subtraction of the oxygen spectrum can be useful.

Fig. 18. 15% limestone-85% sand data. Curve A is $I - 0.5 (B+C) - 1.0$ tool response. Curve B is $A - 0.80$ of the oxygen spectrum. Curve C is $B - 0.85 \pm 0.01$ of the silicon spectrum. Curve D is $C - 0.1 \pm 0.05$ of the calcium spectrum and Curve E is $D - 0.03^{+0.15}_{-0.03}$ of the carbon spectrum.

Fig. 19. Output response as a function of carbon concentration in the samples. The limestone data points have been increased by $\times 1.4$ relative to the pure carbon point to correct for differences in scattering and self shielding. The abscissa is based on both assays listed in Table 2. The points marked by crosses were determined using 4-point response matrices for the calcium-carbon analysis as described in the text.

One should remember for a log-log plot as in Fig. 19, that for the output to be proportional to input, the data must fall on a line of slope 1.00. This linear proportionality is a must; any deviation is an indication of systematic errors. The results meet this criterion except for the 15% point; however, we cannot say that knowing the answer did not influence our results. We believe that the error bars are reasonable. The errors assigned are visual errors of fit. Above and below these values the spectral peel off clearly was not correct. Our error bars are equivalent to $\pm 0.025 \text{ gm/cm}^3$ or a variation in C/O of ± 0.028 , or a variation in CO_2 of ± 0.056 .

Critique

Clearly, determination of the carbonate content to ± 0.056 weight fraction is far short of the 0.01 to 0.02 that seem to be needed according to the containment people. What do we do to improve the results?

Our first question is, have we removed the systematic errors. We found that the spectral continuum is reduced by moving the tool 4" to 10" away from the sample but we have not shown that in this distance variation that the contribution to elemental response functions does not change. What happens in moist environment? The neutron attenuation changes obviously, and we have an enhanced hydrogen-capture peak, which we can handle easily. But what happens to the response functions?

Obviously, improving the statistics in the present analysis would have helped considerably. Those runs that were used to obtain the oxygen, silicon and calcium spectra could have been greatly improved by increasing the counting periods from 20 min to 2.0 hours or more for each. Certainly, the judicious use of MLR techniques would have helped. For those spectra in which prominent γ rays show, eg. hydrogen capture, silicon capture, and

inelastic data, and even the oxygen inelastic data, using visual methods for establishing peel off goes fairly well. For almost-featureless spectra, eg. calcium, the visual method can be faulty. Of course, the lack of features in the calcium spectrum is one of resolution. In principle, one could include a Ge(Li) detector in the system for assaying such elements. Indeed, all but carbon could be done this way. However, this makes for a highly complicated system. Realistically, we could improve the present results a factor of 2.0, getting close to what Schlumberger reports, but hardly more.

Hearst has commented that some logging people do a very simplistic analysis by simply summing the region of the major carbon contribution (eg. 3.17 to 4.65 MeV) and dividing by the counts in the region of the oxygen peak (4.86-6.62 MeV). The channel widths are those used by Hopkinson et al.² of Dresser Atlas. It is not clear that the experimenters even went so far as to subtract the tool response from their data. For the purposes here, we do so, since it improves the appearance of the results and in no way affects statistics. Here we have used the counts in channels 150-240 for the carbon determination and 260-320 for the oxygen determinations. In addition, we used the 4-term response matrix procedure used for the calcium-carbon analysis of the 15% and 30% limestone samples. The results plotted vs. CO₂ weight fraction are shown in Fig. 20. Clearly we get a very nice linear response suggesting high sensitivity. What the data tell us is that barring no other changes, the addition of a small amount of carbon can be detected. Of course, the Dresser Atlas people derive other indices, characteristic of the lithology, and on the bases of all of the data decide whether a small change is due to carbon or some other effect. Clearly, there is no way by

Fig. 20. Simplistic data analysis. The 1 - .5 (B+C) ~ 1.0 tool results were first obtained. Then the carbon and oxygen were determined using a four-term response matrix and the carbon results (x_2) divided by the oxygen results (x_1). The assays of W. McKinnis (see Table 2) are used for the abscissa.

using such an analysis alone to do anything but obtain a rough measure of the carbonate content. However, this does suggest the possibility that by using chemical assays at selected points in a bore hole, the regions in between might be analyzed.

We can get a handle on the detection limit by going to the extreme of subtracting the 15% limestone spectrum from the 30% limestone spectrum. Figure 21 shows the subtraction of 0.97 of the raw I-15% spectrum (B, C, and tool not subtracted), subtracted from the I-30% limestone spectrum. The 0.97 factor has no significance other than that it is close to 1.00 indicating that the neutron generator output was stable and reproducible, and that the residual above ~ channel 222 is about zero. Indeed, for channel 240-370 the count is 848 ± 1177 and for channel 100-230 is 14311 ± 1608 for a net difference of 13463 ± 1992 . This is in a region, channel 100-370, in which the initial count is 1,481,064. The origin of the statistics should be explained. For channels 240-370 the statistics were determined by the square root of the sum of the squares of the individual channel deviations from the average. The same procedure was used for the data of channels 100-230. We could also treat this later data as the difference of two numbers, giving about the same result. One may argue that the difference between the two spectra is partly due to differences in calcium and silicon content but, mostly, it is due to the carbon difference of 0.034 gm/cm^3 . We can then put a number on the minimum error of $\pm 0.005 \text{ gm/cm}^3$, which translates to a CO_2 weight fraction of ± 0.01 . Obviously, the necessary spectral peak offs will introduce a factor of 2 to 4 in the error, thus providing an estimate of the statistical limit.

Fig. 21. The 30% limestone data minus 0.97 of the 15% limestone data.

Examination of the residual shows a remnant below about channel 230 due primarily to the difference in the carbon content of the two samples.

ACKNOWLEDGMENTS

We acknowledge J. Hearst's part in instigating the program and his continued interest. R. Knipes laid much of the early ground work in data acquisition. R. Bond and C. Mills made contributions in electronics and data acquisition. J. Cates helped with mechanical problems. J. Truelsen coordinated special jobs. K. Raschke helped in control of radiation hazards and in special radiation counting problems. W. McKinnis provided a sampling and assay of our sample boxes. L. Maynard helped set up special counting systems. H. Spracklin helped with the detector system and was the sounding board for many of our instrumentation ideas. The

REFERENCES

1. R. B. Culver, E. C. Hopkinson, A. H. Youmans, Carbon Oxygen (C/O) Logging Instrumentation, Soc. of Pet. Eng. J. (Oct. 1974), p. 463.
2. E. C. Hopkins, W. H. Fertl, and D. W. Oliver, The Continuous Carbon/Oxygen Log-Basic Concepts, Soc. of Pet. Eng. J., SPE-9613 (Mar. 1981).
3. R. C. Hertzog, Neutron-Excited Gamma-Ray Spectrometry for Well Logging, IEEE Trans. Nucl. Sci. NS-26, 1558 (1979).
4. R. C. Hertzog, Laboratory and Field Evaluation of an Inelastic-Neutron-Scattering and Capture-Gamma Ray Spectroscopy Tool, Soc. of Pet. Eng. J., Technical Paper SPE 7430 (Oct. 1980), p. 327.
5. P. Westaway, R. Hertzog, and R. E. Plasek, The Gamma Spectrometer Tool Inelastic and Capture Gamma-Ray Spectroscopy for Reservoir Analysis, Soc. of Pet. Eng. J., Technical Paper SPE 9461 (Sept. 1980).

6. W. E. Schultz, H. D. Smith, Jr., Laboratory and Field Evaluation of a Carbon/Oxygen (C/O) Well Logging System, J. Pet. Tech. (Oct. 1974), p. 1103.
7. J. K. Hearst, Laboratory Studies of Improved Carbon/Oxygen Log, Preprint UCRL-81650, 1978, The Log Analyst 19, 16 (1978).
8. A. E. Evans, Jr., Gamma-Ray Response of a 38-mm Bismuth Germanate Scintillator, IEEE Trans. Nucl. Sci. NS-27, 172 (1980).
9. B. D. Sowerby, Elemental Analysis by Neutron Inelastic Scatter Gamma Ray with a Radioisotope Neutron Source, Nucl. Instr. Meth. 166, 571 (1979).
10. F. E. Senftle, R. M. Moxam, and A. B. Tanner, A Comparison of Radiative Capture with Decay Gamma-Ray Method in Bore Hole Logging for Economic Minerals, Nucl. Inst. Meth. 104, 485 (1972).
11. M. R. Wormald and C. G. Clayton, In-Situ Analysis of Coal by Measurements of Neutron-Induced Prompt γ -Rays, Int. J. Appl. Radiat. Isot. 34, 71 (1983).
12. J. H. McQuaid, D. R. Brown, T. Gozani, and H. Bozorgmanesh, High Rate Spectroscopy for On-Line Nuclear Coal Analyzer (NUCOALYZER), IEEE Trans. Nucl. Sci. NS-28, 304 (1980).
13. R. G. Helmer, R. L. Heath, D. D. Metcalf, and G. A. Cazier, A Linear Least-Squares Fitting Program for the Analysis of Gamma-Ray Spectra including a Gain-Shift Routine, AEC Research and Development Report, IDO-17015, 1964.
14. R. G. Helmer, R. L. Heath, L. A. Schmitteroth, G. A. Jayne, and L. M. Wagner, Analysis of Gamma-Ray Spectra from NaI(Tl) and Ge(Li) Spectrometers: Computer Programs, Nucl. Inst. Meth. 47, 305 (1967).

15. E. Schonfeld, Improved Accuracy in Determination of Radionuclide Concentrations in Solutions Containing Fast-Decaying Isotopes by Least-Squares Resolution of the Gamma-Ray Spectra, Proceedings International Conference: Modern Trends in Activation Analysis (1965).
16. E. Schonfeld, A. H. Kibbey, and W. Davis, Jr., Determination of Nuclide Concentrations in Solutions Containing Low Levels of Radioactivity by Least-Squares Resolution of the Gamma-Ray Spectra, Nucl. Inst. Meth. 45, 1 (1966).
17. C. M. Lederer, V. S. Shirley (editors), Table of Isotopes, Seventh Edition, John Wiley and Sons, Inc., New York, 1978.
18. F. C. Engesser and W. E. Thompson, Gamma Rays Resulting from Interactions of 14.7 MeV Neutrons with Various Elements, J. Nucl. Energy 21, 487 (1967).