DOE/PC/80537-77

Final Report

Grant No. DE-FG-22-85-PC80537

Use of Generator Produced Neutrons in

Coal Analysis

Submitted by:

Dr. Arthur N. Thorpe

Howard University

Washington, DC

and

Dr. Frank Senville and Jon L. Mikesell

U.S. Geological Survey

Reston, Va

December 1988

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.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

55 12:12 PM 13 SEP 76

ASSISTANCE & MODIFICATION

RECEIVED
DDOE/PETC

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Table of Contents

INTRODUCTION	1
THE BASIC ELECTRONIC CIRCUITS	3
THE NEUTRON GENERATOR	5
1- The Reservoir and Associated Circuit	5
2- The DC-DC Converter	7
3- The Pulse-Generating Assembly	8
THE (D.D.) OPERATED TTA	9
R.F. ENHANCED ION SOURCE	12
NEUTRON FLUX MEASUREMENT	13
FAST HIGH-ENERGY NEUTRON DETECTOR	14
GAMMA-RAY SPECTRA IN COAL	15
1- Preliminary Measurements	15
2- Comparison of Low and High Energy Spectra	18
SUMMARY AND CONCLUSIONS	21
REFERENCES	23

INTRODUCTION

Usually, chemical analyses on small samples of coal do not yield statistically meaningful data. In a borehole, one encounters a statistically large sample, but it is difficult to make an analysis in the confines of the borehole. This problem has been circumvented to some extent by using neutron activation^{1,2}. As a neutron source, small but relatively strong americium-beryllium or californium-252 neutron sources have been used with relatively good results. However these sources are frowned upon by the coal industry because of their continuous neutron emission requiring large shields, strict licensing requirements, and special handling equipment needed to ensure personal safety. An alternative and more attractive source of neutrons is a neutron generator which can be turned on and off.

The most common neutron generators accelerate deuterium ions into a tritium target and via the $T(D,n)He$ reaction, produce 14 MeV neutrons. High energy neutrons penetrate the coal to such an extent, that by the time the energy of the neutrons is moderated absorption takes place at some distance from the borehole. The activated sample is thus far removed from the detector and results in a large reduction in sensitivity. In addition, there are copious high-energy capture gamma-rays produced which significantly increase the background. If a neutron generator can be made to produce lower-energy neutrons, the sensitivity of a neutron-activation technique using machine generated neutrons could be greatly enhanced. It is well known that, if the accelerator target is loaded with deuterium instead of tritium, 2.5 MeV neutrons can be produced via the $D(D,n)T$ reaction. Although 2.5 MeV neutron generators have been

tested in the laboratory, little work has been done to test the application to a borehole neutron generator. The specific problems of using a neutron generator as applied to bulk coal are not generally recognized, need to be worked out, and tested in greater detail. Also, a study of the nuclear reactions, relative sensitivity for specific elements etc., as applied to coal needs to be evaluated for both 2.5 and 14 MeV neutrons before a neutron generator can be applied usefully. Most of the tests under field conditions have been made with isotopic neutron sources. A few initial tests have been made with an accelerator in the laboratory using 14 MeV neutrons³. As far as we know no field tests with a 2.5 MeV generator have been made to determine the elemental analysis of coal, either in the laboratory or in situ. Also, no good comparative analyses of coal using both 14 MeV and 2.5 MeV neutrons is available.

It was the initial plan to build a conventional borehole accelerator for producing 14 MeV neutrons except to use a target which is loaded with deuterium instead of tritium. In this way, 2.5 MeV neutrons could be produced via the $D(D,n)He$ reaction rather than 14 MeV neutrons by the $T(D,n)He$ reaction. In addition to simply replacing the target, the accelerating voltage, and other electronic parameters must also be changed. However, at the start of this research it was not known what changes in the electronics would be required to produce 2.5 MeV neutrons with a flux of 10^7 or better.

THE BASIC ELECTRONIC CIRCUIT.

The basic neutron generator is patterned after that developed by the Sandia Laboratories for producing 14 MeV neutrons. The accelerator tube (called a Zetatron) is a sealed glass unit consisting of three parts: (1) an ion source with a tritium/deuterium reservoir, (2) a tritium/deuterium loaded target and (3) a pulse transformer. The assembly is potted in epoxy and is referred to as the tube-transformer assembly (TTA). In the TTA, the neutrons are accelerated through a potential of about 120 kilovolts and collide with a metal target loaded with tritium. Higher acceleration voltages are generally needed with a deuterium target because of the lower reaction cross-section for the (D,D) reaction compared to the (D,T) reaction.

An initial pulse is produced in a circuit called a pulse generating assembly (PGA), and fed into one of two subsequent pulse forming networks which produces a 2.4 kV pulse. The 2.4 kV pulse is applied to the ring anode in the TTA. The other network is used to apply a 900 volt pulse to the high voltage pulse transformer in the TTA. The output of the transformer is a negative 120 kV pulse which is applied to the accelerating electrode and target in the TTA.

In addition, the PGA has three major sections which supply the necessary bias, control voltage, and initial pulses to the TTA circuits. These sections are (1) a gas reservoir control circuit, (2) a D.C/D.C. converter circuit and (3) a pulse generating circuit. The overall configuration is shown in Fig-1.

The neutron tube reservoir is a small cylinder containing metal hydride powder that releases a mixture of deuterium and tritium gas when electrically heated. An external

Figure 1. Neutron Generator Configuration

circuit controls the gas pressure within the tube by varying the current through the reservoir heater. The circuit consists of a linear-feedback system that monitors the ion source voltage waveform each time the TTA is triggered, and varies the drive current to the tube reservoir accordingly. If the tube pressure is low, the discharge (strike) portion of the ion source voltage waveform occurs later in time and the strike integral will be greater than the normal value, causing the circuit to provide more reservoir drive current. The strike portion of the waveform being defined as the portion of the waveform that drops quickly from the 2.4 kV amplitude to the burn voltage. Conversely, if the tube pressure is too high, the strike integral becomes smaller causing the circuit to adjust the reservoir pressure to a predetermined relatively constant value. This operation requires about 5 seconds depending upon the particular tube reservoir response time and the magnitude of pressure change required. The predetermined pressure at which the circuit controls the tube is determined experimentally and henceforth is controlled by the feedback control circuit.

Limits of high and low tube gas pressure must be observed. At too high a pressure, forward and reverse breakdowns within the tube become more prevalent. These breakdowns are suspected to reduce the life of a tube. In addition, forward breakdowns within the tube can cause damaging transients within the probe electronics. At too low a pressure the ion source voltage may fail to strike and no tube current flows. Thus, the tube provides no loading to the secondary of the TTA transformer which may cause the secondary voltage to exceed design values. Continued operation may result in breakdown of the high voltages within the transformer winding or from the winding to

the external ground.

THE NEUTRON GENERATOR

During operation of the TTA, the deuterium-tritium gas within the tube must be held at a low and constant pressure (about 3 Pa or 4.35×10^{-4} psi). A titanium reservoir in which deuterium and/or tritium is absorbed is the source of the gas. To keep the tritium/deuterium gas pressure constant the reservoir is gently heated and cooled by a control circuit. In this way, a constant current of deuterium ions is produced and accelerated down the TTA to interact with the tritium in the target electrode. Another circuit, the quiescent drive, provides reservoir drive current when the tube is not being triggered at repetition rates greater than 20 Hz.

1-The Reservoir and Associated Circuit:

Figure 2 is a block diagram of the reservoir control circuit depicting the major functional parts of the circuit. In order to simplify the description of this circuit, assume that the probe is triggered at 20 Hz or higher, and that the quiescent circuit had been previously adjusted to properly warm up the reservoir. The trigger pulses are integrated and the resultant dc level is fed into the negative input of the quiescent amplifier, driving the output of this amplifier low and removing the quiescent level from two steering diodes in the control circuit. Thus, the quiescent signal is disabled. At the input of the feedback control circuit a source voltage waveform will be present after each trigger. The waveform is reduced from its peak value of 2.4 kV to the level shown in the same figure

Figure 2. Reservoir Control Circuit Block Diagram.

(Fig.2). It is then clipped by a network including a Zener diode to remove the burn voltage from the waveform. Next, each resulting pulse is integrated with a time constant of 2.4 ms. The amplitude of these integrated output pulses corresponds to the integral of the source voltage waveform taken above the burn voltage level. A peak detector then converts this pulse into a dc voltage proportional to the integrator pulse amplitude. With this arrangement the reservoir control circuit is independent of the input trigger frequency.

The peak detector has an adjustment that sets the feedback input-level to the amplifier section of the control circuit. The peak detector is fed into a high-input-impedance buffer. This stage is needed to keep the peak detector from being loaded by the amplifier stage of the control circuit. The amplifier circuit input impedance is equal to the series combination of the feedback resistors. When megohm resistors are employed to get the input impedance high, the stray capacities across the resistor causes the response of the control circuit to become unstable. The buffer solves this problem and compensation in the form of a single capacitor can be applied to the amplifier stage. The major factor contributing to the response of the control circuit, and thus its stability, is the response time of the reservoir itself. Response time is defined as the time the reservoir takes to change pressure in the tube a predetermined amount (1 Pa) after receiving a step function increase in heating current from the control circuit. Zetatron tubes of known responses covering the range from relatively slow to fast were employed to design the amplifier gain and compensation network of the control circuit.

The dc voltage out of the amplifier drives a pulse-width modulator. The output of

the integrating circuit consists of two, 180° out-of-phase pulses. The duty cycle of these pulses is inversely proportional to the dc input from the control circuit amplifier over a range of +2 volt to +6 volts. These pulses in turn drive a switching power supply that converts +28 V into a low voltage, high current (2A-4A) reservoir drive.

When the triggering phase is over, the signal at the quiescent amplifier controls the pulse width modulator through its steering diode. The circuits are then ready for the next pulse.

2-DC-DC Converter:

The DC-DC converter is a switching, regulated, power supply that converts the 500 V_{dc} to 28 V_{dc} . The 28 V_{dc} is used to power the internal electronic circuits of the system. Its operation is similar to the reservoir control circuit in that it uses a pulse width modulator in the feedback loop to regulate the +28 V output. A block diagram of the converter circuitry is shown in Fig. 3.

The 500 V is fed into a start-up circuit following the trigger pick-off pulse transformer. A capacitor in the start-up circuit is charged up to 500 V_{dc} when the 500 volt supply is turned on. At the same time, a small voltage is applied to a Zener diode network which feeds the 24 V_{dc} diode regulator. The 24 V is then fed to the power supply inputs of all the circuits within the DC-DC converter/reservoir control module. The time constant of the start-up circuit is designed to hold up for as long as it takes the DC-DC converter to start and produce 28 V. The 28 V is fed into the same 24 volt Zener network to produce a constant 24 V for the circuitry within the module. When the 24 V is first

Figure 3. DC/DC Converter

Figure 4. Pulse Generating Assembly Block Diagram.

applied to the DC-DC converter, the pulse width modulator causes the switching circuit to chop the 500 V. A transformer then converts the chopped 500 V to a lower ac voltage. The voltage is rectified, filtered and fed back into an operational amplifier. If the voltage is higher than 28 V, the resultant output of the amplifier will cause the pulse width modulator to decrease the duty cycle of the switching circuit. If the voltage is lower than 28 V, the amplifier output causes the pulse width modulator to increase the switching circuit duty cycle. This change in duty cycle decreases or increases the DC-DC converter's output until the desired 28 V amplitude is reached. A potentiometer at the control amplifier's input provides an adjustment of this amplitude.

3-Pulse Generating Assembly (PGA)

The function of the pulse generating assembly is to provide medium voltage pulses to the target via the TTA transformer and also to the ring electrode in the neutron tube. It provides the neutron detector trigger pulse and also has logic and timing circuits that prevent noise pulses from retriggering the probe after a trigger pulse has been received.

In Fig. 4., an LC resonant doubler circuit charges the two pulse forming networks to about 900V. The voltage is somewhat less than twice the 500V supply voltage because of the voltage drop in the transmission cable, and losses in the doubler circuits. The trigger pulse is taken from the 500 V line and is applied to a gate and timing circuit. The purpose of this circuit is to allow only the trigger pulse to switch the Switching Control Rectifier (SCR). All noise pulses occurring during the time immediately following the probe

trigger will be blocked out for about 7 milliseconds. Another timing circuit is employed to generate a pulse that is wide enough to allow the SCR to completely discharge the two pulse forming networks.

After the SCR is switched, one pulse forming network applies a 900 volt pulse to the primary winding of the high voltage transformer. At the same time, the other pulse forming network applies a 900 volt pulse to the TTA transformer which in turn applies a 2.4 kV source voltage pulse to the anode of the ion source. The series resistor is used to set the source current to about two amperes. Two of the source network divider resistors are connected to the source voltage output. The voltage across one resistor is employed as a source-voltage monitor; the other provides the feedback signal to the reservoir control circuit.

THE (D.D.) OPERATED TTA

The previous discussion of the electronic circuitry refers to a tritium loaded TTA used for generating 14 MeV neutrons. This equipment was completed and tested early in the investigation period. While this was in progress and order was let to the General Electric Company for two deuterium loaded TTA's. Construction of the units could not begin until authorization was obtained from the Department of Energy (Oak Ridge). For reasons which are not clear the authorization was held up for over a year. In order not to hold up the work it was decided to simulate the TTA so that the electronics could be tested. Special resistive loads were obtained to simulate the filament, ion source and the 120 kV transformer. This worked initially, but trouble developed with the gas pressure

control circuit and this approach was finally abandoned.

About this time, we located a deuterium loaded accelerator tube at Sandia Corporation, but it had no high voltage transformer. Using this tube and a transformer taken from one of our defunct tritium loaded TTA's, we attempted to construct a TTA. Before the transformer could be mounted on the new D-D tube, it had to be removed from the old TTA. The transformer and resistor are connected to the tube by means of threaded stud centered in the end of the accelerator tube. After they are screwed together, the whole assembly is potted in epoxy resin. To separate the two, it is essential not to disturb the potting compound around the transformer or resistor. Because the resin is opaque, it is difficult to tell just where to make a thin cut so that the accelerator tube can be unscrewed from the resistor. In addition, the tube contains 7 Curies of a radioactive isotope, tritium. Accidentally cutting into the tube chamber would release this gas into the air.

To solve these problems, a rectangular wooden jig with a lead scale was made to fit firmly over the TTA and then the assembly was X-rayed at a local hospital. From the X-ray pictures (Figure 5), engineering literature, and drawings from the Sandia National Laboratory, we were able to establish the dimensions and locations of the various components. With the help of the pictures and drawings, the potting compound was removed in the region where the two components were screwed together. The cut had to be made true without intersecting any of the components in the assembly. In this way, the tube was removed without damaging the transformer or resistor.

As the D-D tube could not be permanently embedded in epoxy resin, the

Figure 5. X-Ray of TTA showing pulse transformer, target and ion source.

transformer and tube had to be immersed in a liquid fluorocarbon to prevent high voltage arcing when the tube is operated. Although it could not be used in a borehole, a plexiglas box was built (Figure 6A) to hold the components immersed in the liquid fluorocarbon so that bench tests could be made. The fluorocarbon acted as a dielectric and coolant and completely covered the D-D tube and transformer. Never-the-less vigorous arcing was experienced and no neutron output was observed. The arcing was undoubtedly due to overvoltage caused by the tube not drawing sufficient current. After considerably adjustment of the electronics, it was found that the heater voltage feedback circuit used with the D-T tube had to be substantially changed when the circuit was used with a D-D tube. By proper adjustment, the D-D tube appeared to fire properly. It should also be noted that probably as a result of the arcing experienced initially several of the electronic components had been damaged and needed to be replaced. Upon completion of the bench tests, a sealed plastic shroud was made to hold the fluorocarbon dielectric) (Fig. 6B).

Once the correct shape of the strike pulse was obtained, the D-D tube started to fire properly, the arcing ceased, but only a relatively low neutron flux was obtained. Several activation experiments were made to ascertain that neutrons were being emitted. The important step here was that we were able to make the D-D tube work even though the transformer had been taken from another TTA assembly and no high voltage potting was used.

The above experiments were made by running the accelerator at 100 pulses per second. To increase the neutron output it is clear that the repetition rate must be

Figure 6A. Box to hold fluorocarbon dielectric and the TTA.

Figure 6B. TTA showing the sealed plastic jacket made to hold the fluorocarbon.

increased. Modifications were made to the high voltage power supply and the trigger circuit , and adjustments made to the control circuits, so that the repetition rate could be increased to about 500 pulses per second. At 500 pulses per second the resolution can be significantly enhanced.

R.F. ENHANCED ION SOURCE

Another way to enhance the neutron flux output of the generator would be to increase the output of the Penning ion source. Between ion pulses, the ion concentration drops to a low value. If a constant uniform ionization can be maintained in the ion source between pulses, then additional concentration of ions produced during the pulse will increase the total ion output. To accomplish this, a radiofrequency coil was wound around the ion source end of the TTA envelope. The radiofrequency coil, which was driven by a 25 megahertz tunable generator, was used to induce a field in the TTA to produce a constant ionization between pulses. The generator output could be set for low-, medium-, and high-intensity. To determine effect of r.f. ionization on the neutron generators, a comparison was made using both generators under the same conditions. A BF_3 neutron detector was placed approximately one meter from the respective targets and counts were recorded for 60 seconds. A low pulse repetition rate of 50/sec was used to preclude any possible overheating effects in the TTA. The results shown in Figure 7 for three r.f. intensities and indicate a significant positive enhancement in the neutron flux in both cases compared to the flux where there was no radiofrequency induced ionization. Time did not permit further development of this technique for use in a borehole configuration.

NEUTRON FLUX MEASUREMENT

In the process of increasing the repetition rate to 500 neutron pulses per second, a relatively large number of successive changes and adjustments were made to the electronics. After each change, the relative neutron flux per pulse must be measured. The average flux is conventionally measured by activating and counting metal foils. However, this is a time consuming method in cases such as ours where many measurements must be made. To circumvent this problem, we considered developing and building a fast detector for measuring 2.5 MeV neutrons. However, as the neutron output of our generator was so low, and, as we also needed a fast neutron counter for 14 MeV neutrons, we considered the following method using the 14 MeV generator. If this worked well, we would build a similar detector for 2.5 MeV neutrons using a different reaction.

To measure 14 MeV neutrons, a reaction with a fairly high threshold energy is desirable. The $^{16}\text{O}(n,p)^{16}\text{N}$ has a threshold of 10.3 MeV and results in a 7.13 second decay of ^{16}N . The high energy gamma rays emitted by ^{16}N would seem to be ideal for indirectly measuring 14 MeV neutrons.

To test the concept, some preliminary experiments were performed in a large plastic barrel of water. The target end of the neutron generator was immersed in about 40 cm of water and positioned 15 cm from the edge of the barrel. A high-purity germanium detector was mounted opposite the generator target and about 15 cm from the outside of the barrel. Initially, the neutron generator was turned on for 20 seconds at a neutron-burst repetition rate of 30 per second. At the end of the irradiation period,

counts were collected in a window straddling the ^{16}N decay gamma rays at 6128 keV and its escape peaks at 5617 and 5106 keV. Decay measurements of these lines indicated a half life of about 7 seconds showing that the window is properly set for oxygen. The neutron burst repetition rate was then increased and the experiment repeated. Figure 7 shows how the total number of gamma rays from oxygen change as the neutron burst repetition rate increases. Experiments were also made with 5 and 10 cm of water between the generator and the detector as shown. At a repetition rate in excess of 120 per second, the neutron flux per burst drops rather fast. The drop in neutron output is due to the electronic pressure-adjustment circuitry losing control of tube pressure. It is this problem we had to address, but to accomplish this test, we must have a fast response 14-MeV neutron counter. These tests show that the oxygen technique is quite adequate to measure the 14-MeV neutron output from the generator in significantly shorter times than if activation foils are used. Thus, this method can be used to measure relative neutron flux and thus guide us as we make the necessary electronic changes to increase the neutron burst repetition rate.

FAST HIGH-ENERGY NEUTRON FLUX MEASUREMENT

In order to make real-time measurements of the 14 MeV neutron flux using the oxygen technique discussed above, it was decided to make an instrument designed specifically for this task. In addition to a need for such an instrument for timing and adjusting the neutron generator, such a flux measuring device could be most useful for a number of nuclear experiments.

Figure 7. Relative neutron flux using low-, mid- and high-level radiofrequency compared under the same conditions without radiofrequency enhancement.

Figure 8. Change in the relative neutron counting rate as a function of pulse repetition rate for three intervening thicknesses of water.

Basically the instrument should be a sensitive gamma-ray detector surrounded by a sheath containing oxygen. Because of its high density and efficient gamma-ray detection, barium germanate (BGO) was selected as a scintillator. Thus, a 1.5 inch photomultiplier tube was mated to a BGO detector of the same diameter and about 1 inch thick.

Several oxygen-containing compounds were considered for the sheath material. H_2O could be used, but hydrogen will thermalize and absorb neutrons. After some consideration, SnO_2 , which contains 21.2 percent oxygen was picked as the most promising material. The Sn has a very low neutron absorption cross-section and no interfering gamma-rays. Figure 9 shows a diagram of the detector-sheath combination. The sheath contained about 2 kg of SnO_2 . The instrument works remarkably well and provides a method of real-time high-energy neutron flux measurement. A similar instrument could undoubtedly be fabricated for low energy neutrons, but there was insufficient time to build one.

GAMMA-RAY SPECTRA IN COAL

As the final simulated borehole facility utilizing a multiton sample of coal was not completed, some preliminary measurements were made on smaller samples.

1-Preliminary Measurements:

A cylindrical three foot diameter wooden barrel full of high volatile bituminous coal was mounted over the target area of the accelerator which was laid close to the floor.

Figure 9. 2kg SnO_2 sheath surrounding the scintillation detector using a BGO crystal.

A high purity germanium detector was mounted on the top of the barrel.

The generator was set to operate at 100 pulses per second for this experiment so that fast neutron reactions, thermal capture reactions, and decay reactions could all be observed. Each neutron pulse was 5 μ s in duration and yielded 3×10^5 neutrons or an average emission rate of 3×10^7 neutrons per second. The neutron emission rate during the burst is equivalent to that of 26 mg of ^{252}Cf , a large isotopic neutron source. Because of this large neutron output, the gamma-ray detector was completely overloaded during the burst. The generator output had to be reduced by a factor of two and the source to detector distance increased to about 120 cm in order to obtain good prompt gamma spectra during the burst.

Inelastic gamma-ray formed by neutron scattering were measured during and slightly after the neutron pulse called Gate 1 (see Figure 10). Neutron capture gamma-rays were measured for several hundred microseconds between 0.1 to 2 ms after the end of the neutron pulse (Gate 2). A third and much wider gate some 5 to 10 milliseconds after the neutron pulse was used to measure the decay gamma-rays due to thermal neutron capture. Figure 11 shows the spectra taken over a period of 10 μ s starting at the beginning of each neutron burst. In this time frame, only prompt gamma rays from inelastic neutron scattering can be seen. Prompt gamma rays from the $^{16}\text{O}(n,n'\gamma)^{16}\text{O}$, $^{12}\text{C}(n,n'\gamma)^{12}\text{C}$, and $^{56}\text{Fe}(n,n'\gamma)^{56}\text{Fe}$ reaction are observed and the counting rates are low. In part the low count is due to the low neutron burst repetition rate used in this experiment.

A spectrum taken from 0.1 to 2 milliseconds after each neutron burst is shown in

Figure 10. Sheath showing the relative positions of the counting windows.

Figure 12. In this time window, the thermal neutron capture gamma rays from many elements can be seen. Some of these have been indicated in the figure. From the point of view of coal analysis, thermal capture spectra will yield the most information about elemental concentrations. Again the number of counts are low and can be enhanced by increasing the neutron burst repetition rate.

Finally, a spectrum taken 5-10 milliseconds after each neutron burst is shown in Figure 13. Because of the long delay the only activity observed in this spectra is due to decay gamma-ray reactions. Most of the gamma-ray peaks are due to naturally radioactive elements. A few of the elements were indicated on the graph, but at greater neutron pulse rates and enhanced neutron flux, more elements might be observed. It is interesting to note that decay gamma rays from the oxygen (n,p) can be seen. With pulsed neutrons from a D-D generator, the neutron energy will be too low to observe this oxygen reaction. This will be a disadvantage in using D-D neutrons because the fast neutron detector cannot be used to monitor the flux. However, low energy neutron emission can be measured in other ways.

The result of this experiment indicates that if a 2.5 MeV neutron generator is used to perform the same type of neutron activation as described above, the neutron flux must be improved by at least two orders of magnitude. This will improve the thermal capture spectrum. With the lower energy and lower neutron output of the D-D tube, the source-to-detector distance can be substantially shortened.

Figure 11. Prompt gamma-ray spectrum due to inelastic scattered neutrons.

FAST NEUTRONS - PROMPT

Figure 12. Capture gamma-ray spectrum due to thermal capture.

THERMAL CAPTURE

Figure 13. Decoy gamma-ray spectrum due to thermal capture neutrons.

2-Comparison of Low and High Energy Spectra.

One of the objects of this investigation is to compare the spectra obtained in coal using 14 MeV and 2.5 MeV neutrons. Initially it was planned to use machine-generated neutrons in both cases. However, because of our inability to produce a strong enough flux of machine-generated 2.5 MeV neutrons, it was decided to use Cf252 neutrons (~2.2 meV) as a substitute. To do this a large coal sample (several tons) is required. The test facilities available at the USGS for activating large ore samples are constructed of steel and are not hermetically sealed. Iron has a number of strong photopeaks in a capture gamma-ray spectrum that can be used to measure the iron in coal, but the large amount of iron in these steel containers makes this measurement impossible. Also, if the coal loses water by evaporation, the neutrons will not be moderated as in a natural environment. Therefore, to reproduce the natural environment as closely as possible, it was decided to construct a new hermetically-sealed borehole facility fabricated of fiberglass and plastic instead of steel. A large fiberglass drum was purchased from the Raven Corporation in Washington, Ohio. A 3 1/2 inch ID plastic tube to simulate the borehole was mounted along the central axis of the drum. The upper end of the tube was supported by a plywood ring and the lower end by a laminated bakelite support. Freshly mined bituminous coal (pea size) was specially trucked to the laboratory and placed in the drum as soon after removal from the mine as possible so that the moisture content did not change significantly. When filled, the drum was hermetically sealed with silicone rubber. An analysis of a representative sample taken at the time of filling is shown in Table 1.

Table 1. Chemical Analysis of Coal Sample from the Franklin Seam (near Mt. Storm, W. Va.)

	As-received	Dry	Dry ash-free
MOISTURE	2.87		
ULTIMATE ANALYSIS			
Hydrogen	4.46	4.26	4.94
Carbon	73.22	75.38	87.41
Nitrogen	1.45	1.50	1.74
Sulfur	3.00	3.09	3.58
Oxygen	4.50	2.01	2.33
Ash	13.37	13.76	
	<hr/> 100.00	<hr/> 100.00	<hr/> 100.00

FREE SWELLING INDEX 9.0

To obtain some comparative information using fast neutrons (14 MeV) and ^{252}Cf neutrons (≈ 2.5 MeV), spectra were taken with the (D,T) neutron generator and a californium isotopic source. The experimental arrangement is shown in Figure 14. The neutron source or the generator target, as the case may be, was placed about 80 cm from the bottom of the coal canister. The germanium detector was placed outside the coal canister so that the germanium crystal was opposite the source and 22 cm from the outside of the canister. Counts were recorded with the californium source ($\approx 85\mu$) and also with the neutron generator, each for 45 minutes.

The spectrum for each of the neutron sources is shown in Figure 13. The most obvious difference between them is the presence of the oxygen (n,n') photopeaks (full energy 6130 keV) in the 14 MeV spectra, but not in the spectrum made with californium neutrons. There are some other interesting features. The presence of the natural potassium in the high energy neutron spectrum is due to the lower count rate and background levels in this spectrum. It was not seen in the low energy spectrum. Two photopeaks of germanium, at 142 and 198 keV, show up in the fast neutron spectrum only. The 198 keV peak is a sum peak of the 23 and 175 keV peaks, and occurs because both of these gamma rays are emitted simultaneously and detected within the germanium crystal. The 142 keV line is caused by thermal neutron capture by ^{74}Ge . The presence of these peaks in the spectrum indicates that when 14 MeV neutrons are used, thermal neutrons penetrate and escape from the two foot thick coal sample. This does not happen when lower energy ^{252}Cf neutrons are used. The escape of neutrons is also shown by features in another part of the spectrum and is considerably larger than that found with Cf-252 neutrons. At the same time, many of the atoms which are activated

Figure 14. Experimental arrangements of neutron generator and germanium detector with respect to the coal canister.

Figure 15. 14 MeV and ^{252}Cf neutron capture spectra for coal.

beyond a two foot radius in an infinite sample thickness situation are so far from the borehole detector that the gamma rays will have a high probability of scattering. Activation by the slow neutron source will be considerably more efficient in spite of the higher penetration and larger sample formed by high energy neutrons. In these experiments, the neutron flux was 3×10^7 n/sec for the neutron generator and 4.9×10^7 n/sec for the Cf-252 neutron source. In the fast neutron spectrum 2.55×10^6 gamma rays were recorded whereas 1.50×10^7 gamma rays were recorded for the Cf-252 source. These measurements point out the desirability of using a source of neutrons with energies less than 14 MeV, and confirms the original rational of the project to develop a D,D borehole generator.

SUMMARY AND CONCLUSIONS

A 14 MeV neutron generator is satisfactory for making capture gamma-ray analysis in a borehole in rocks for a limited number of elements. In coal, a thermal-capture gamma-ray spectrum will yield data for a relatively larger number of elements. However, by the time 14 MeV neutrons are reduced to thermal energies and absorbed by the target elements the neutrons are a considerable distance from the neutron source and the detector. Consequently, the activated coal sample is dispersed and a long way from the detector, and results in a reduced sensitivity. If the neutron generator source emitted 2.5 MeV neutrons by a (D,D) reaction, the neutrons would be thermalized close to the detector. In this case, the sensitivity of the neutron activation method would be substantially enhanced.

To produce a borehole generator which would produce 2.5 MeV neutrons, we started with 14 MeV generator using a tube-transformer assembly (TTA) as developed by the Sandia Corporation. A similar TTA was constructed using a deuterium rather than a tritium loaded target. The new TTA did not respond adequately with the exiting electronics. Many tests had to be made under adverse conditions. Finally, we were able to obtain a pulsed neutron flux but the intensity was very low and insufficient to perform practical neutron activation analyses. Several techniques such as increasing the pulse rate, using an R.F. enhanced ion source etc. improved the flux rate but not sufficiently for practical neutron activation. The reaction cross-section for the accelerating voltage available was just too low for the (D,D0) reaction. Higher voltages could have been used to enhance the cross section but were incompatible with the insulation capabilities in the

confines of the borehole.

In the course of this investigation, a fast-response high-energy neutron detector using the $^{16}\text{O}(\text{n,p})^{17}\text{N}$ reaction was developed which worked quite well and was a great help in adjusting the electronic parameters of the 14 MeV neutron generator.

As the flux rate of the 2.5 MeV neutron generator was inadequate for activation analysis of coal, Cf-252 was substituted in order to obtain comparable spectra using 14 and 2.2 MeV neutrons. Using a specially constructed coal irradiation facility with a central borehole, experiments were made using neutrons of both energies and comparable fluxes. It is clear from the results that lower energy neutrons yield spectra which are more useful for coal analysis.

REFERENCES

1. Senftle, F., Tanner, A., Philbin, P., Boynton, G., and Schram, C., 1978, "In situ analysis of coal using a ^{252}Cf Ge-Li borehole sonde." Mining Engineering, (AIME) vol. 30, pp 666-674.
2. Mikesell, J., Dotson, D., Senftle, F., Zych, R., Koger, J. and Goldman, L., 1983, In situ capture gamma-ray analysis of coal in an oversize borehole, Nucl. Instr. and Methods, 215, 561-566.
3. Mikesell, J., Senftle, F. and Tanner, A., 1985 "Borehole elemental analysis of coal using fast neutrons." Proceedings of IAEA Advisory group meeting on gamma, X-ray, and neutron techniques for the coal industry, Vienna, Austria.