

## FEASIBILITY OF CONTINUOUS ASH MEASUREMENT OF COAL

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An automatic and continuous method of measuring the ash content of coal is needed by the coal industry. Automatic control of coal quality would reduce preparation costs, improve the product, and thus indirectly increase markets for coal. Existing methods of ash analysis involve a laborious process of sampling, sample preparation and chemical analysis, and the results may not be available for several hours--perhaps long after the coal has been shipped. For bulk products like coal, sampling costs can easily become a significant proportion of production costs. The industry needs a continuous method of analysis that will permit immediate control of quality.

One possible method involves a process in which some type of nuclear radiation penetrates a considerable depth into the coal, undergoes a nuclear reaction with the chemical elements, and produces characteristic radiation that emerges from the coal and is measured. For this method, only neutrons and gamma rays will provide the required depth of penetration. When large samples of coal are bombarded by energetic neutrons, three nuclear processes can occur: (1) elastic scattering; (2) inelastic scattering; and (3) capture and subsequent decay. If the energetic neutrons are in the Mev-energy range, the usual sequence of events is for each neutron to make a number of elastic and inelastic collisions, losing energy with each collision, every inelastic collision leaving the bombarded nucleus in an excited state. When the excited nucleus returns to ground state, prompt gamma rays of characteristic energy, called inelastic gammas, are emitted. Eventually, after a large number of elastic and inelastic collisions, the neutron reaches thermal energies (about 0.025 ev) and neutron capture takes place. If the capture process excites the nuclide, prompt gammas are emitted by mechanisms similar to that of the inelastic scattering. These characteristic gamma rays are known as radiative capture gammas. Later, usually after relatively long periods depending on the half-lives of the radioisotopes, the nuclide may decay and emit a beta particle, a gamma ray, or both. These latter gamma rays are known as activation gammas. Both types of gamma rays have energies characteristic of the nucleus of the element from which they were produced, hence the number of such gamma rays can be counted and related to the concentration of elements in the coal.

Measurement and evaluation of gamma rays of different energies involves measurement by a scintillation detector and analysis by a pulse-height analyzer that produces an energy spectrum. Gamma ray spectra from coal show broad base-lines due to various interference effects and peaks that are characteristic and proportional to the individual elements in the material. This method offers the possibility of analyzing large volumes of material--say hundreds of pounds of coal--and since these processes are relatively instantaneous they would be applicable to continuous analysis of tonnage quantities of moving coal.

The mineral matter content of coal, commonly referred to as ash, includes many compounds. Alumina, silica, iron oxide, lime and magnesia generally comprise over 95 percent of coal ash.<sup>1/</sup> If each of the elements in these compounds, Si, Al, Fe, Ca, and Mg, could be measured by the nuclear method described above, a summation of these measurements should give the ash content. To explore this possibility, basic measurements were made in a physical arrangement that could be adapted to tonnage flow of coal.

<sup>1/</sup> Ode, W. H. Coal Analysis and Mineral Matter. Ch. 5 in Chem. of Coal Utilization (Supp. Vol.), ed. by H. H. Lowry. John Wiley and Sons, Inc., New York-London, 1963, pp. 202-231.

Experimental Equipment and Procedure. Apparatus used in this work is shown in figure 1. It consisted of a neutron source, a heavy metal attenuator, and a scintillation crystal detector surrounded by a cylinder of coal. The neutron sources were mixtures of plutonium and beryllium or of americium and beryllium which produce a broad spectrum of neutron energies ranging from 1 to 10 Mev by the reaction of alpha particles with the beryllium target. Radioisotope sources were chosen as neutron sources because they are relatively inexpensive and because they provide long-term unattended operation at constant flux. Commonly, 50-million neutrons per second are produced by a few curies of americium in a 1-inch-diameter steel capsule. The neutron source may be shielded by an inch of lead to reduce the gamma flux without adversely affecting the neutron output. The scintillation crystal detector is shielded from the beam of neutrons by placing it in the shadow of a cone-shaped tungsten attenuator, 8 to 12 inches in length. The detector, consisting of a 3- by 3-inch sodium iodide crystal and photomultiplier tube, is shielded from thermal neutrons by a boron shield surrounding the crystal.

About 50 pounds of coal is placed around the detector in the direct path of the neutrons. Fast neutrons from the source penetrate the coal producing inelastic and capture gamma rays, some of which interact with the detector crystal. Electrical pulses produced by the detector are proportional to the energy of the incident gamma photon. The latter are then measured by the multichannel analyzer and sorted by amplitude to represent the gamma ray spectrum.

Results. Gamma ray spectra were obtained that showed small peaks from the elements in the coal, plus a large amount of interference from unwanted radiation measured by the detector. Figure 2 shows the various interference components together with an unresolved spectrum from high-ash coal. The unresolved spectrum, or upper curve, includes background interference and is that part of the total spectrum between 0.6 and 2.5 Mev. It was obtained after coal, neutron source, and detector were kept in place for 90 minutes to approach equilibrium with respect to neutron activation. Curve A shows several small peaks and two prominent peaks corresponding to the silicon, aluminum, and hydrogen in the coal. Most of the spectrum is due to three major interference components which were estimated by several procedures and are indicated by curves A, B, and C. Curve A shows interference from neutrons transmitted through the attenuator directly into the crystal, curve B represents activation of the crystal detector by transmitted and scattered neutrons, while curve C is only the crystal activation from transmitted neutrons.

Although several improvements were made subsequently, including alteration of the composition and shape of the attenuator, better detector shielding, and different configuration of the coal samples--interrelated interference effects--it was found difficult to remove all of the interference and still retain a physical configuration adaptable to tonnage flows of coal. Improvements in one part of the spectrum often decreased the sensitivity of measurement in another part. Ultimately, however, interference effects were identified and reduced until a spectra was obtained having characteristic peaks that were reasonably sharp and consistent.

Typical results are shown in figure 3, a 50-minute measurement of 40 pounds of bituminous coal containing 9.4 percent combined silicon, aluminum, and iron. Prominent gamma peaks in the spectrum are from carbon, hydrogen, and the major ash constituents--silicon, aluminum, and iron. The 4.43 Mev carbon peak and its two pair peaks being at relatively high energy required special instrument adjustment to obtain good resolution, as shown by the insert to figure 3. The 2.22 Mev hydrogen peak contains some contribution from sulfur at 2.24 Mev, which is difficult to separate; the 1.78 Mev peak of silicon contains some contribution of the 1.77 Mev aluminum peak; and the 1.01 Mev peak of aluminum includes a few counts of the 1.04 Mev silicon peak. Iron appears prominently at 0.84 Mev and adds a small peak at 1.24 Mev, where aluminum

and silicon also contribute small peaks. A small and inconsistent peak appears at about 0.77 Mev, probably from calcium. There should be a fairly sharp peak from magnesium at 1.37 Mev but this particular coal sample did not contain enough magnesium to show it. This spectrum actually is only a part of the total spectrum which extends to 10 Mev. Some indication of an oxygen triple peak was found at 5.12, 5.63, and 6.14 Mev, and amplification gave several other peaks representing unidentified elements.

Sensitivity of Measurement. To estimate the sensitivity of measurement, tests were made on 15-pound samples of Pittsburgh-seam coal containing 3.3-percent and 21-percent ash. The 21-percent ash sample contained 5.8-percent Si, 2.3-percent Al, and 1.3-percent Fe--oxides of these elements comprising 90.4 percent of the ash in this sample. The 3.3-percent-ash sample contained 0.6-percent silica, 0.5-percent Al, and 0.39-percent Fe--oxides of these elements accounting for 90.2 percent of the ash.

Spectra of these two coals are shown in figure 4 at two amplifications. The difference between the 1.78 Mev silicon-aluminum peaks in the two spectra is readily apparent. There are 160,800 total counts under this peak for the low-ash coal and 177,300 total corresponding counts for the 21-percent ash coal, a difference of 16,500 counts. Coals containing 8.1- and 1.19-percent silicon plus aluminum respectively, gave a difference of  $16,500 \pm 581$  counts, a sensitivity of 0.02 percent. Thus a change of 0.02 percent in silicon and aluminum theoretically was detected, although this was not entirely true because silicon and aluminum were assumed to contribute equally on a weight basis to the 1.78-Mev peak. With normal coal samples this estimate probably would not be off by more than a factor of two, so it is reasonable to expect that a change of 0.1 percent in silicon or aluminum content of the coal could be detected.

Subsequently, the actual sensitivity of this method was determined in static tests with coal. Samples of anthracite containing 6.2-, 7.0-, and 7.3-percent ash, respectively, were used in these tests. The small differences in the percentage of ash in these samples approximate the combined errors of sampling and analysis. Spectra obtained from the three samples, using 50-minute counting periods for each, are shown in figure 5. The shift in the 2.22-Mev hydrogen peak indicates a channel drift from instrument instability, so the peaks are blunter than under ideal conditions. A small but definite difference is apparent between the 7.3- and 7.0-percent-ash samples. Iron, aluminum, and silica peaks for the 6.2-percent-ash coal, however, are virtually identical to the coal containing 7-percent ash. (The slight differences are indicated by the dotted line.) The reason for this was shown subsequently to be due to a slight difference in degree of activation of the crystal detector and thus the amount of interference. Although the results cannot yet be considered as completely reliable, they indicate that differences can be obtained between coals differing less than 1 percent in ash content. Minor inconsistencies in the data are just as likely to have been caused by errors in sampling and chemical analysis as by errors in the nuclear method.

Conclusion. In static measurements, the concentration of several of the elements in coal was determined with varying degrees of accuracy by means of gamma rays produced by neutrons from radioisotope sources. Spectra peaks were readily identifiable for carbon, hydrogen, silicon, aluminum, iron; smaller peaks were found for other elements in coal.

The results were promising enough to warrant extension of the work to a pilot-scale system for measurement of the ash content of coal flowing at rates of 1 to 20 tons per hour. Indications are that the concentrations of ash in moving coal can be measured continuously within 1 percent, and perhaps within 1/2 percent.

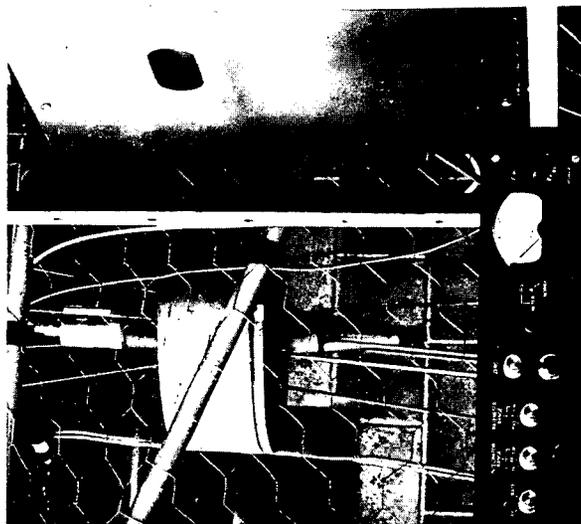
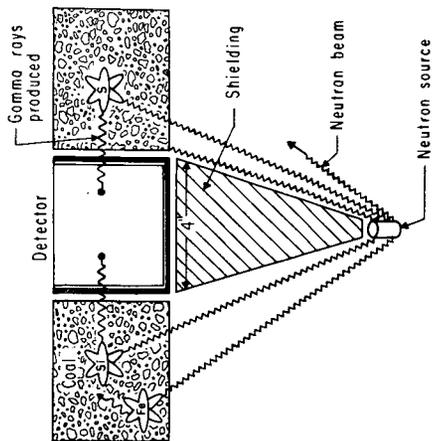
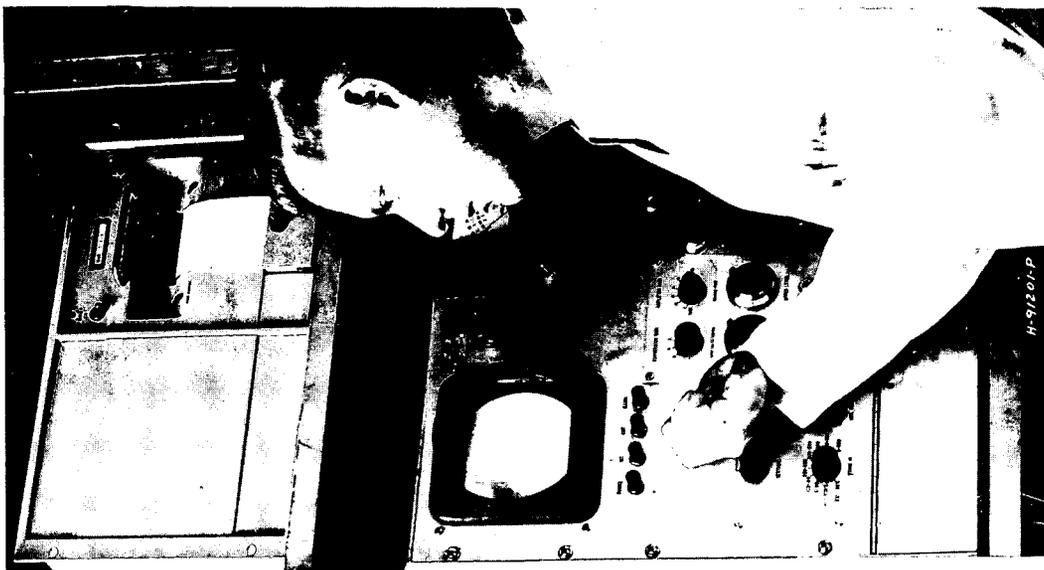


FIGURE 1—Apparatus for Measuring the Gamma Rays Produced by Neutrons Reacting With Ash Elements in Coal

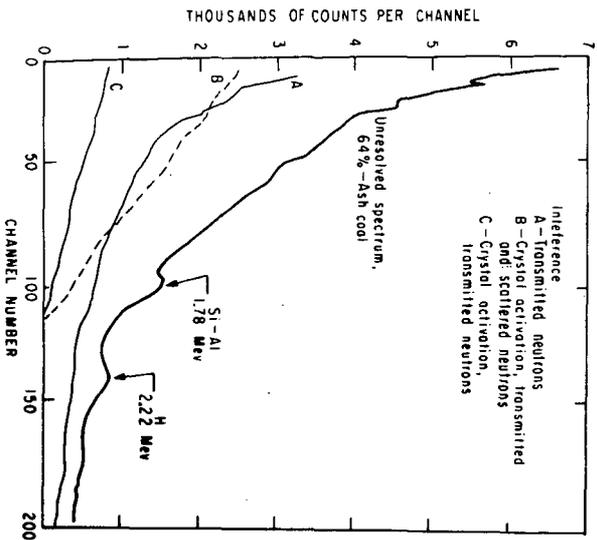


FIGURE 2—Gamma Ray Spectra and Interference Components of High—Ash Coal

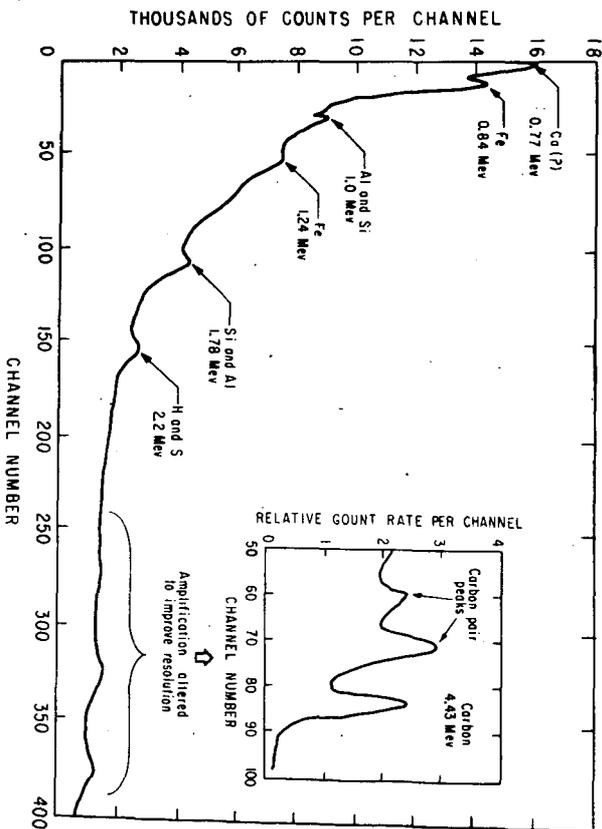


FIGURE 3—Gamma Ray Spectra from Coal Containing 23% Ash

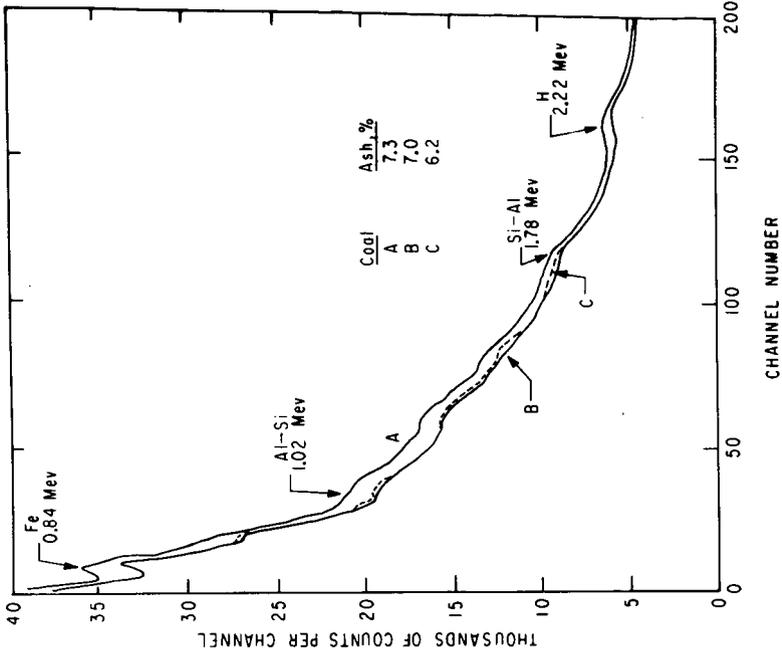


FIGURE 5 — Gamma Ray Spectro from Three Anthracite Samples

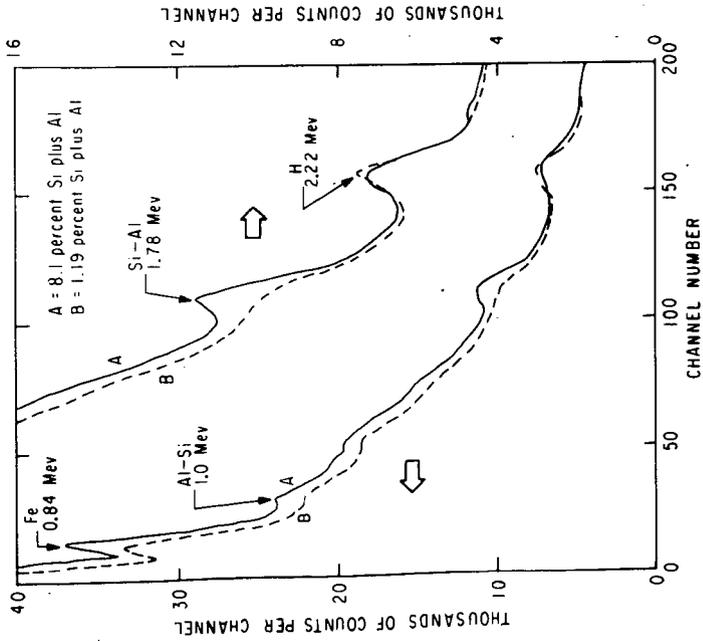


FIGURE 4 — Gamma Ray Spectro of High- and Low-Ash Coals