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Prompt Gamma Analysis of Coal Samples using an Accelerator-based PGNAA Setup

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Abstract. An accelerator based PGNAA setup was developed to analyze the elemental composition of bulk sample of cement and concrete. In this study the elemental composition of a coal sample was determined using the accelerator based PGNAA setup. The size of the coal sample was calculated through prompt gamma ray yield optimization as a function of sample radius and sample length using Monte Carlo simulations. Then powder coal samples with the optimum size were prepared and irradiated with neutron beams. The prompt gamma ray spectra of pure coal and coal containing known amount of chloride salt were acquired. Strong interference has been observed between chlorine gamma rays and those from the coal sample constituents. This study has demonstrated the useful application of an accelerator-based PGNAA setup in the elemental analysis of coal samples

Introduction

Prompt Gamma ray Neutron Activation Analysis (PGNAA) is commonly used in the coal and cement industries for elemental analysis of raw material [Braun H. and F. Riffel, 1982; Gozani, T., 1984; Herzenberg C. L., 1979; Khelifi R., et. al., 1999; Kirchner, A. 1991; Wormald, M.R., et. al., 1993; Lim C. S., et. al., 2001; Naqvi A. A.. et. al.,2006; Naqvi A. A. et. Al. 2004; Pfutzner, H. and Mahdavi, M. 1994; Saleh H. H. and Livingston, R. A. 2000; Tickner J. 2000]. In the coal industry, elemental analysis of raw coal is used for several purposes including price tagging (according to different grades), control parameters for environmentally safe combustion of coal, and analysis of the coal by-products such as ash [Herzenberg C. L., 1979; Kirchner, A. 1991]. The coal industry requires a rapid, non-destructive, and non-invasive on-line analytical technique for elemental characterization in bulk coal samples. Additionally the analysis should not leave a residue of hazardous byproducts. Prompt Gamma ray Neutron Activation Analysis (PGNAA) and X Ray Florence (XRF) techniques have proven to be viable techniques in the coal (Braun H. and F. Riffel, 1982; Gozani, T., 1984; Herzenberg C. L., 1979; Kirchner, A. 1991; Wormald, M.R., et. al., 1993; Pfutzner, H. and Mahdavi, M. 1994;) and the cement (Khelifi R., et. al., 1999; Lim C. S., et. al., 2001; Naqvi A. A.. et. al.,2006; Naqvi A. A. et. Al. 2004; Saleh H. H. and Livingston, R. A. 2000; Tickner J. 2000) industries. The PGNAA technique is a rapid, non-destructive, powerful multi-elemental analysis technique and is ideally suited for industrial process control. In a PGNAA analysis, the elemental composition of the sample is determined from the intensity of prompt γ-rays produced through neutron inelastic scattering (n, n'γ) and thermal neutron capture (n_{th},γ) [Naqvi A. A.. et. al.,2006; Naqvi A. A. et. Al. 2004]. Like many analytical techniques, XRF and PGNAA, have difficulties in analyzing low atomic number elements such as C and O. In order to analyze C and O as well, the PGNAA technique is modified to the Pulsed Fast and Thermal Neutron Analysis (PFTNA) technique, in which the gamma ray spectrum is recorded from both prompt gamma rays and activation gamma rays [Wormald, M.R., et. al., 1993]. In the present study, a prompt gamma ray analysis was carried out of coal samples using an accelerator based PGNAA setup at the King Fahd

University of Petroleum and Minerals, Dhahran, Saudi Arabia [Naqvi A. A.. et. al.,2006; Naqvi A. A. et. Al. 2004]. Prior to gamma ray analysis, Monte Carlo calculations were carried out to obtain the optimum size of a coal sample. Then the samples were irradiated in the PGNAA setup. In the present paper, the results of the simulation study and the experimental tests conducted on the PGNAA setup using the coal samples are reported.

Materials and Methods

Calculations of Optimum Size of Coal Sample

The optimum size of the coal sample was calculated through Monte Carlo simulations using the MCNP4B2 code [RSICC- Computer Code Collection-MCNP4B2, 1999.]. These calculations were carried out using the procedure described elsewhere [12]. Figure 1 shows a schematic diagram for the PGNAA setup with a rectangular moderator, which was used in the simulations. The rectangular moderator is made of paraffin wax while its front moderator is made of high density polyethylene. The chemical composition of the coal sample used to determine the optimum size of a coal sample is listed in Table 1.

Table. 1.

Element	wt(%)
C	78.71
H	4.99
Cl	0.11
N	1.43
Si	1.66
Al	1.05
Fe	0.36
S	0.75
Na	0.03
K	0.11
Ti	0.07

The sample size was optimized to maximize the detection of the prompt gamma rays due to chloride impurity in the coal. In this procedure, assuming a small quantity 0.11 wt. % of chlorine impurity to be present in coal, the length and radius of the coal sample was optimized to detect the maximum yield of chlorine prompt gamma rays. Chlorine has several prompt gamma rays which may interfere with the gamma rays from the coal constituents. In this study, only two gamma rays with extreme energies 1.161 and 6.11 MeV energies were chosen for optimization.

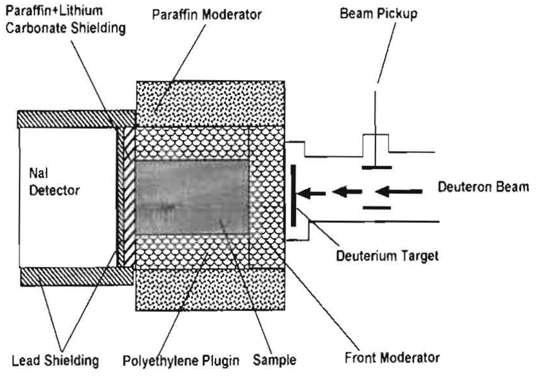

Fig. 1. Schematic of the PGNAA setup used in the study.

The bulk density and moisture content of the coal samples were assumed to be 1.09 g/cm^3 and 1 wt % respectively. The chlorine gamma rays yield was calculated as a function of front moderator thickness, sample radius and sample length. The optimum values of these three parameters i.e. front moderator thickness, sample radius and sample are those values for which the maximum yield of the prompt gamma rays was observed. It required approximately 55-60 minutes of computation time to reduce the statistical uncertainty in the prompt gamma ray yield calculations for the most prominent lines to 1-2 %. In this study, the radius and length of a cylindrical concrete sample was determined to obtain the maximum yield of the prompt gamma rays at a NaI detector location. The dimension of the detector was 25 cm x 25 cm (diameter x height). The results of these calculations suggest an optimum value of the sample radius to be about 11.5 cm and a sample length of 14 cm. The maximum value of the prompt γ-ray yield has been observed for 4 to 5 cm thick front moderator. In the present design of the PGNAA setup, the thickness of the front moderator was chosen to be 5 cm.

Prompt Gamma Ray Yield Measurement from Coal Samples

The prompt gamma ray yield from the coal samples was measured using the KFUPM PGNAA setup with a pulsed beam of 2.8 MeV neutrons produced via a D(d,n) reaction [Naqvi A. A.. et. al.,2006]. The deuteron pulse had a width of 5 ns and a frequency of 31.25 kHz. The typical beam current of the accelerator was 3-4 μA. Due to continuous implantation of deuterons by the incident beam in the target backing, target thickness was not well defined but neutron intensity at the target was estimated to be 5x10^5 n/s.

The coal samples, whose size was calculated in the previous section, was prepared by grinding the coal into powder form and filling them in a thin walled 14 cm long polyethylene container which had an 11.5 cm inner radius. The samples were irradiated in the rectangular moderator. The data was acquired for a preset number of charges measured at the electrically-isolated neutron producing target. The prompt γ-ray spectrum of the NaI detector was acquired in a gated mode; the gate signal being derived from the beam pick-up signal. Figure 2 shows the prompt gamma ray spectra from a 14 cm long pure coal sample with a 11.5 cm radius taken with a 5 cm thick front moderator. Figure 2 shows a portion of the pulse height spectrum of prompt gamma rays from the pure coal on an enlarged scale with energies higher than 3 MeV.

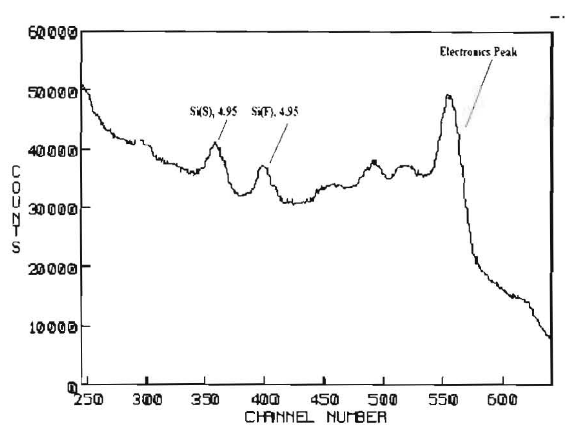

Fig. 2. Experimentally measured prompt γ-rays spectrum from pure coal sample with energies above 3 MeV.

The prompt gamma ray spectra were measured from two more coal samples containing chlorine contamination in 2wt.% and 4 wt % proportions. These spectra are shown in Fig. 3 and 4 respectively.

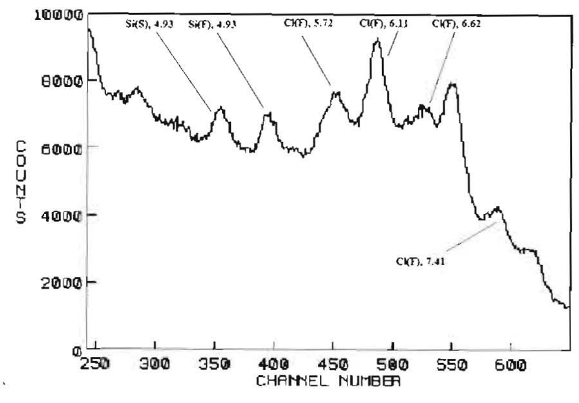

Fig. 3 Experimentally measured spectrum of prompt γ-rays with energies above 3 MeV from a coal sample containing 2 wt. % chlorine .

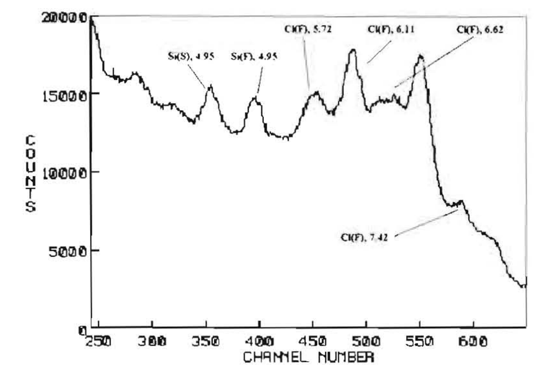

Fig. 4. Experimentally measured spectrum of prompt γ-rays with energies above 3 MeV from a coal sample containing 4 wt. % chlorine

Results and Discussion

The major constituents of coal are carbon. Neither 2.8 MeV nor thermal energy neutrons can excite carbon nuclei. The 2.22 MeV prompt γ-ray peak due to the capture of thermal neutrons in hydrogen in the moderator material is very intense and prominent. Figure 2 shows the full energy (F) and single escape (S) peaks in the pulse height spectrum of prompt gamma rays from silicon higher than 3 MeV in pure coal sample on an enlarged scale.

The large peak on the right side of the spectrum is the electronic sum peak of the detector, which is produced due to the addition of noise signals from six photomultipliers viewing the NaI crystal. The prompt γ-ray peaks from the capture of thermal neutrons in silicon in coal, are located in the spectrum at energies higher than the 2.22 MeV hydrogen peak. Figure 2 shows the full energy (F) and single escape (S) peaks in the pulse height spectrum of prompt gamma rays from silicon at 4.935 and 3.539 MeV (not marked) .Figure 3 shows prompt gamma ray spectra of coal mixed with chloride salt. The main feature of Fig. 3 is the presence of chlorine gamma ray peaks along with gamma ray peaks from the coal. The chlorine has several high energy prompt gamma rays at 1.161, 5.72, 6.11, 6.62 and 7.41 MeV. The chlorine full energy gamma rays may interfere with full energy gamma rays of the coal sample constituents. Additionally full energy gamma rays of chlorine may interfere with the single and double escape peaks of other chlorine gamma rays. For example the single escape peak of the 6.619 MeV chlorine peak may interfere with the chlorine full energy peak at 6.110 MeV. This results in an increased intensity of 6.11 MeV peak, as shown in Fig. 3. Similarly the single escape peak of 6.11 MeV peak may interfere with full energy peak at 5.72 MeV, resulting enhanced intensity of the 5.72 MeV peak. Figure 4 shows the similar effects on enhanced scales for 0.4 wt. % chlorine concentration in coal sample. Based on the data shown in Figures 2-5 it can be concluded that the the KFUPM accelerator-based PGNAA setup [Naqvi A. A.. et. al., 2006] can be used successfully in determining the concentration of chlorine in the coal sample.

Conclusion

Elemental analysis of coal samples was carried out using the KFUPM accelerator-based PGNAA facility. In this study, the prompt gamma ray yield was measured from pure coal samples and coal samples containing known amount of chloride salt. Inspite of strong interference of chlorine prompt gamma rays with those from the coal sample, the full energy peaks of the prompt gamma ray peak from chlorine were detected. This study has demonstrated the useful application of an accelerator-based PGNAA setup in the elemental analysis of coal samples.

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