Department of Physics

Physics 503 - Graduate Laboratory (0-6-3)
Term 132

Prompt gamma activation analysis

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Abstract

Hydrogen and carbon concentrations were measured in four crude oil samples via 14 MeV neutron inelastic scattering using a LaBr3:Ce detector. Despite its intrinsic activity, the LaBr3:Ce detector performed well in detecting the hydrogen and carbon elements. Water and benzene samples were used for the detector energy calibration to identify the carbon and hydrogen peaks. The carbon to the hydrogen (C/H) element ratio was calculated from the experimental data for each oil sample. Samples with the higher (C/H) ratio are defined as poorer quality (less liquidity) and samples with the lower (C/H) ratio have better quality. In our samples the measured (C/H) ratio varies from 1.403 to 1.847.

1- Introduction

Prompt Gamma Activation Analysis (PGAA) is ideally suited for in-situ elemental analysis of bulk samples in various industrial, environmental, medical and other applications [1]. Radiative neutron capture, or (n, γ)-reaction is the most important reaction for PGAA. Once a nucleus absorbs a neutron, a compound nucleus is formed with excitation energy equals the binding energy in addition to the kinetic energy of the neutron. The compound nucleus needs about 10^{-16} s to decay as shown in figure 1. The nucleus reaches its ground state, typically in 10^{-9} - 10^{-12} s, by emitting 2 to 4 gamma rays in a cascade. Gamma rays (γ) are called prompt, if their decay times following the capture, are much shorter than the resolving time of the detection system, which typically is in the range of 10 ns to 10 μs [2]. Radioactive decay radiation (typically ß-decays and electron capture followed by γ-rays) with a given half-life will also be emitted when the ground state reached after the de-excitation is not stable. The delayed gamma radiation produced in this way based on neutron activation analysis (NAA), but several nuclides can be analyzed in PGAA using their decay radiation too. If the ground state of the daughter nucleus is stable, the process ends here (Figure 1).

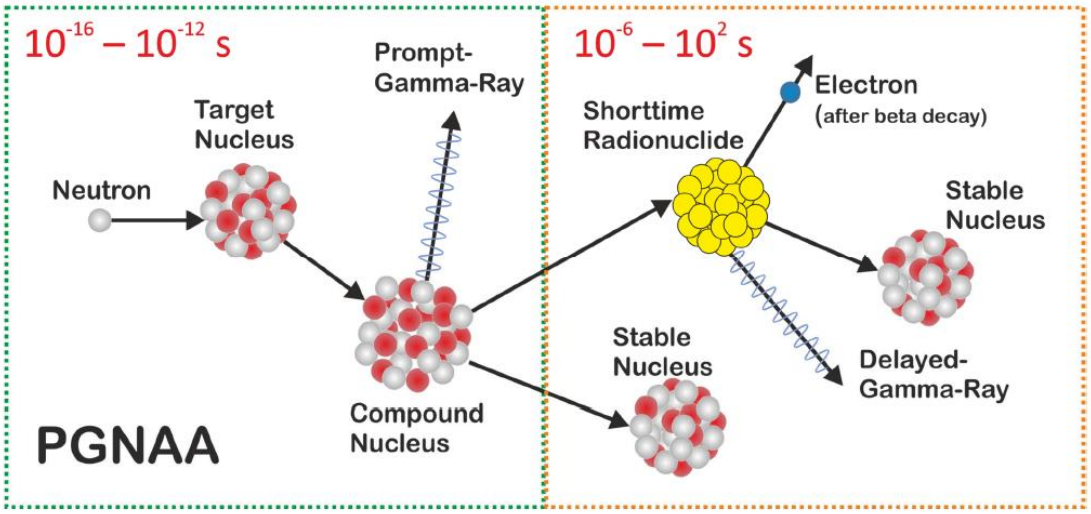

Figure 1: Schematic diagram illustrates nuclear decay products, prompt and delayed gamma ray.

In general, the inelastic scattering of neutrons or reaction can happen only when the neutron energy is above the energy of the first excited state of the scattering nucleus, i.e. it is a threshold reaction. A neutron generator is, usually, used as a neutron source, in which deuterons are accelerated at a few hundred keV onto a deuterium or tritium target (or vice versa), with a yield of about 10^{-5} neutrons/deuteron [2]. The neutron energy from the fusion reaction $^{2}H + {}^{2}H \rightarrow {}^{3}He + n$, is about 2.5 MeV, and from $^{2}H + {}^{3}H \rightarrow {}^{4}He + n$ it is about 14 MeV.

In the PGNAA technique, fast neutrons were used to irradiate a material. Some of the fast neutrons are moderated by the material in an external moderator. These neutrons interact with the material through neutron inelastic scattering (n, ′nγ) or thermal neutron capture (n_{th}, γ) reactions to produce prompt γ-rays. The elemental composition of the sample can then be determined from the intensity of prompt γ-rays produced, either through neutron inelastic scattering (n, n`γ) or thermal neutron capture (n_{th}, γ) or both [3]. Figure 2 shows gamma rays decay scheme due to inelastic scattering of neutrons from C, N and O elements. Prompt gamma rays emitted by the irradiated samples are counted by gamma ray detector with good energy resolution. The measured intensity of gamma ray is directly proportional to elemental concentration in the sample. The detection sensitivity of a prompt gamma ray measured is strongly depends upon the gamma ray detector performance.

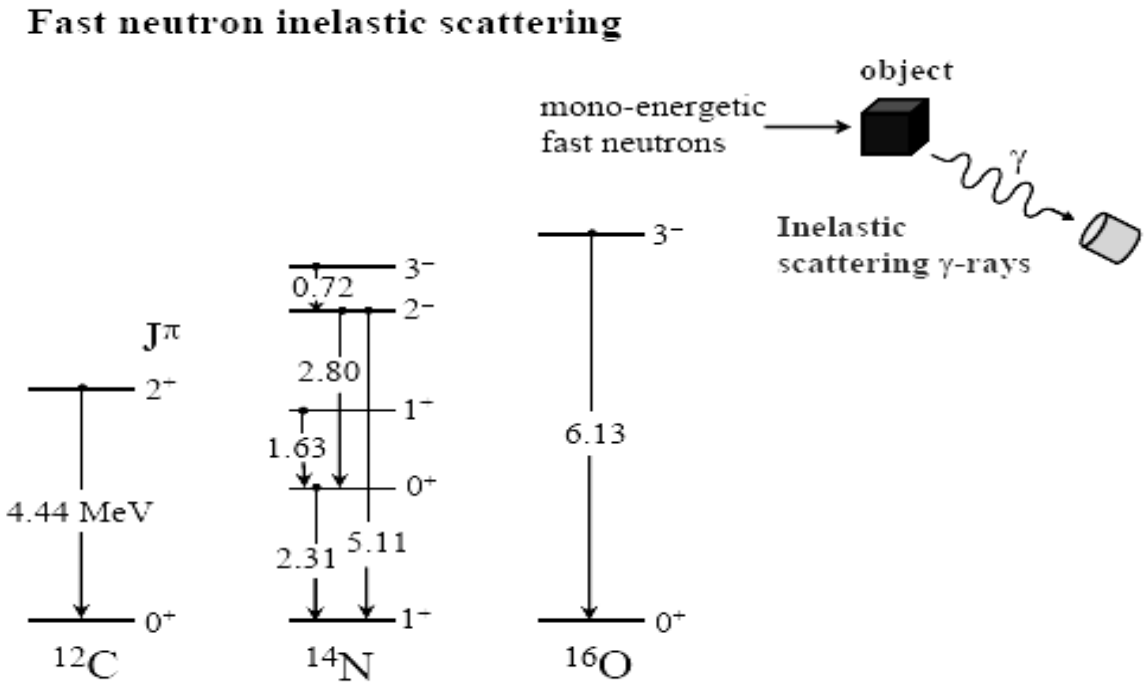

Figure 2: Gamma Ray decay scheme from inelastic scattering of neutrons from C, N and O.

1.1 $LaBr_3$:Ce Gamma ray Detector

Recent developments of radiation hardened lanthanum-halide ($LaBr_3$:Ce and $LaCl_3$:Ce) gamma ray detectors with improved light output, decay time and energy resolution have widened the scope of applications for the Prompt gamma-ray neutron activation analysis (PGNAA) technique [4]. Although lanthanum halide detectors have an intrinsic activity due to radioactive decay of a naturally occurring unstable La isotope, they have nonetheless been successfully employed in high count rates studies because this type of detector can handle higher count rates than the conventional NaI detectors [5]. However, because of their intrinsic activity, lanthanum-halide detectors may not be suitable in low-level counting experiments.

PGAA setup employing lanthanum halide detectors are expected to have better performance than those employing NaI detectors because lanthanum halide detectors have outperformed conventional NaI detector in terms of light decay time, energy resolution and high count-rate handling capabilities. Moreover, LaBr3 has approximately a factor of two improved energy resolution, compare to NaI, with full width at high maximum (FWHM) less than 3% at 662 keV and 30% higher detection efficiency. This detector also has faster decay time of 60 ns and can operate over wide dynamic range of count rate with little variation in the energy resolution [6].

2- Experimental set up

The geometry of the 14 MeV neutron based PGNAA setup used in the present study is shown in Figure 3 [7]. It consists, mainly, of a cylindrical sample (90 mm x 145 mm) placed 7.0 cm away from a tritium target at a 0^o angle with respect to the 14 MeV neutron beam. The gamma ray detector is placed at a center-to-center distance of 19 cm from the sample at an angle of 90^o with respect to the 14 MeV neutron beam. Tungsten blocks are inserted between the neutron target and the gamma ray detector to shield it from the direct beam of 14 MeV neutrons, the detector was also shielded from 14 MeV neutron-induced gamma ray background through massive lead shielding inserted between the detector and the tungsten shield.

Finally, a paraffin structure was built next to the tungsten blocks to shield the detector from room scattered neutrons. The paraffin shield was prepared by mixing lithium carbonate and paraffin wax in equal weight proportions. The paraffin and lead shield was quiet effective in shielding the detector against scattered neutrons and background gamma rays. However, the gamma ray peaks in the background spectrum due to inelastic scattering of 14 MeV neutrons from lead and paraffin shielding were quite pronounced. The collected data was processed using standard NIM electronics modules connected to a personal PC as a multichannel analyzer (MCA). The NIM electronics block of the experiment is shown in figure 4.

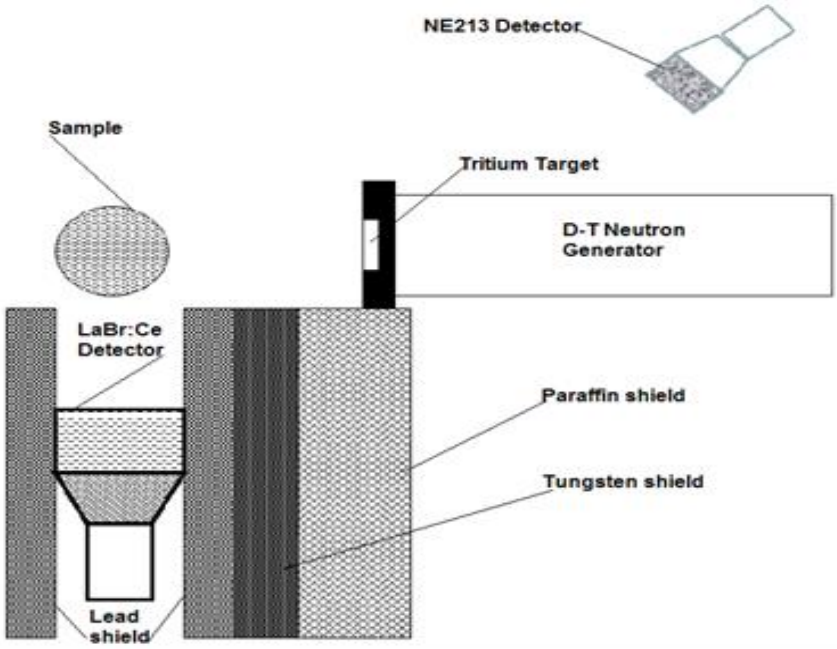

Figure 3: Schematic of 14 MeV neutron-based setup used for measurement of H, C and O concentration in crude samples.

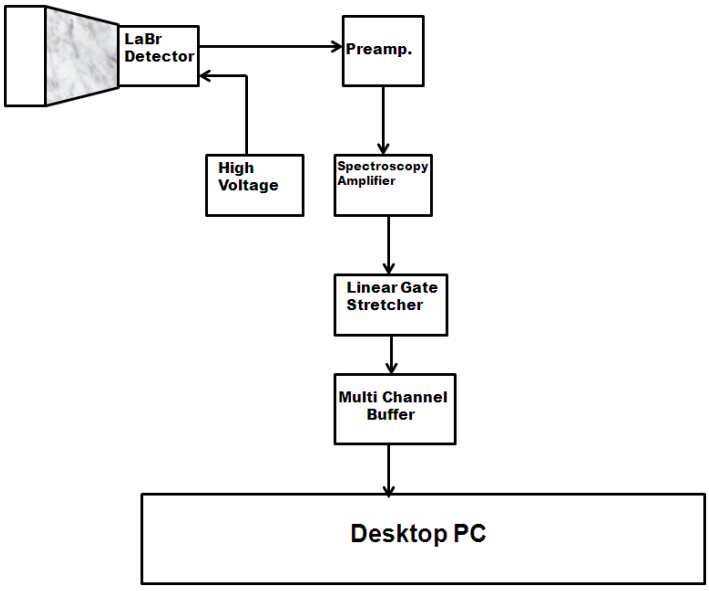

Figure 4: The electronics block diagram used in the present PGAA experiment.

3- Results and Discussions

In the present study, data from a LaBr3:Ce detector for hydrogen and carbon peaks in the respective pulse height spectra of four crude oil, water, and benzene samples were analyzed. The gamma rays yield spectrum as a function of a channel number was generated for each sample.

3.1 Energy Calibration of LaBr3:Ce Detector

The LaBr3:Ce gamma ray detector and its components do not give a direct reading for energy versus intensity, so that the C, H, O, and intrinsic peak's identification is needed in order to calibrate these types of detectors. Two samples, water and benzene, were used in addition to the La source in the detector itself to achieve this purpose.

3.1.1 Intrinsic Spectrum of LaBr3:Ce Detector

The LaBr3:Ce gamma ray detector has 588 volts with positive polarity, and its intrinsic activity was measured, as a reference, using standard NIM electronics modules. The intrinsic peak was founded in channel number 75. Figure 5 shows the pulse height spectrum of the detector itself recorded over a period of 2011 seconds. It shows the 1468 (1436+32) keV gamma line of the detector's intrinsic activity resulting from the sum of the 1436 keV gamma due to beta decay of ^{138}La isotope and the 32 keV X-ray fluorescence peak due to K shell X-ray fluorescence of Ba produced in the electron capture by La [8]. The intrinsic activity rate was determined from the integrated counts under the 1468 keV peak.

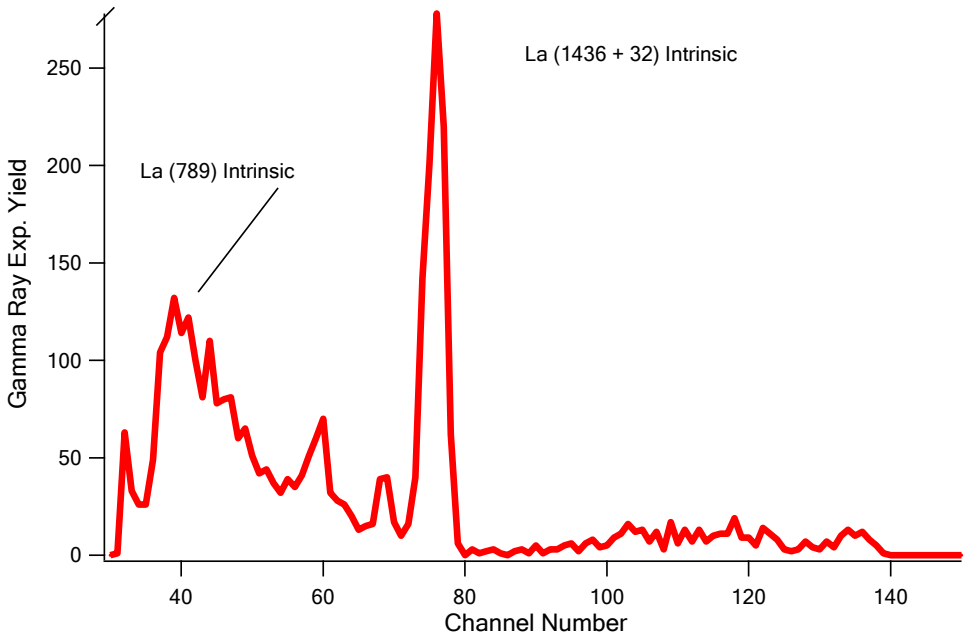

Figure 5: Intrinsic activity spectrum of the cylindrical 76 X 76 mm^2 (diameter X height) LaBr3:Ce gamma ray detector.

In (H_2O), concentration of oxygen is very high comparing to hydrogen and there is zero carbon concentration. On the other hand, there is almost no oxygen concentration in benzene (C_6H_6), while the carbon concentration is very high as shown in Figure 6. Therefore, water can be used to determine the position of oxygen and hydrogen peaks, and benzene can be used to identify the carbon peak.

The spectrum of water shows four peaks: Oxygen full peak, single escape SE, double escape DE, and Hydrogen peaks, respectively from right to left, whereas the benzene spectrum shows one sharp peak refers to Carbon and two small peaks fitted with the oxygen and SE peaks which come from the atmosphere since, there is no oxygen in the benzene sample. The intrinsic peaks appear in both spectrums. The pulse height spectrum of the LaBr3: Ce detector for water and benzene samples is shown in figure 6.

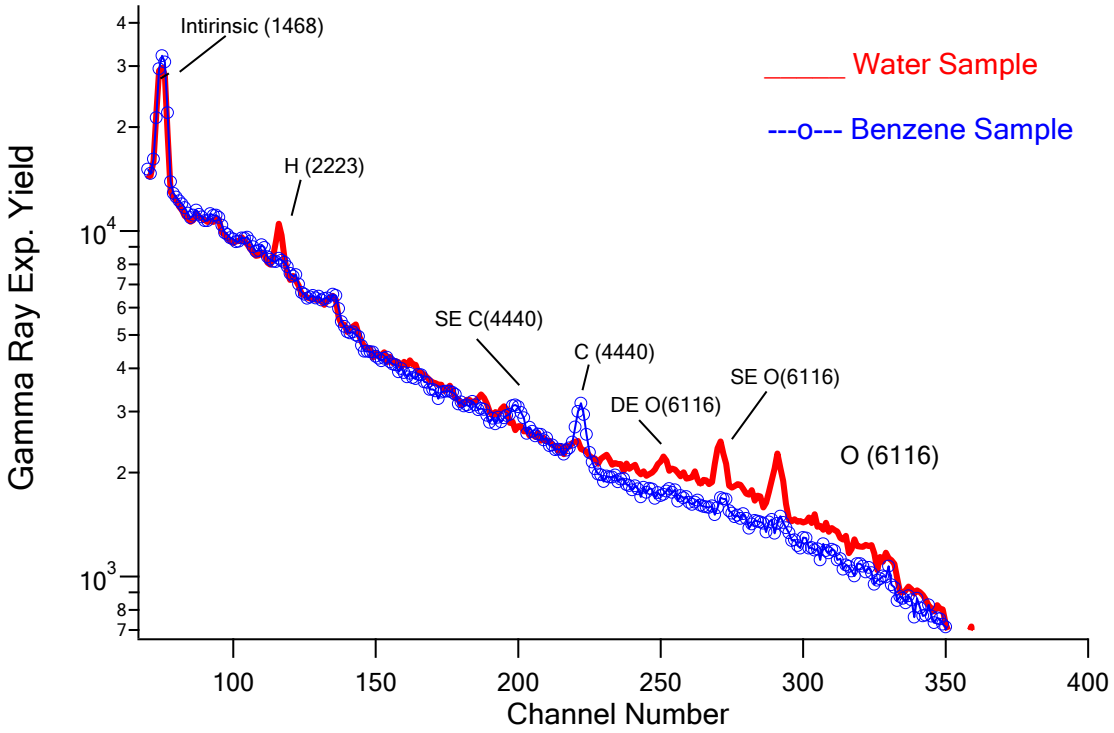

Figure 6: Full prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector for water and Benzene, taken with 14 MeV neutrons PGNAA setup.

The spectra exhibits the full energy peaks along with associated escape peaks. For the 4.44 MeV prompt gamma rays from carbon, the full energy and single escape SE peaks have been detected while for the 6.116 MeV prompt gamma rays of oxygen, the single escape, and double escape DE peaks have been detected along with the full energy peak.

The prompt gamma-ray energies for each main peak correspond to their channel number is shown in table 1.

Table 1: Prompt Gamma ray energy corresponding to each channel number.

Peak	Channel Number	Corresponding Energy (MeV)
Intrinsic	75	1.468
H	118	2.223
C	223	4.440
O	295	6.116

Figure 7 shows the fitted curve of the energy calibration for the LaBr3:Ce detector. The calibration equation is shown on the graph where E referred to the prompt gamma energy while C corresponds to the channel number.

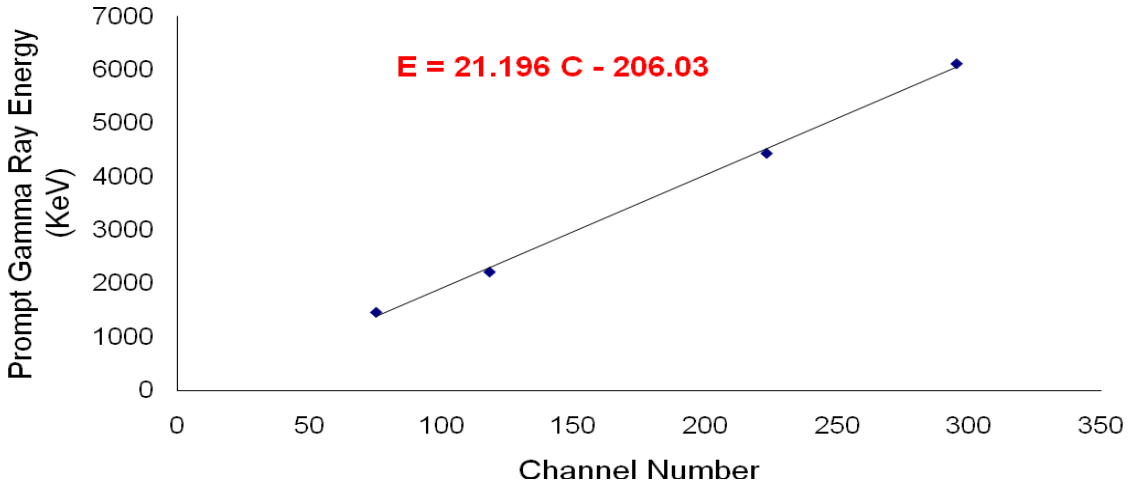

Figure 7: Energy calibration curve of the LaBr3: Ce detector.

The detector background spectrum is also measured without any sample, and the results were shown in Figure 8.

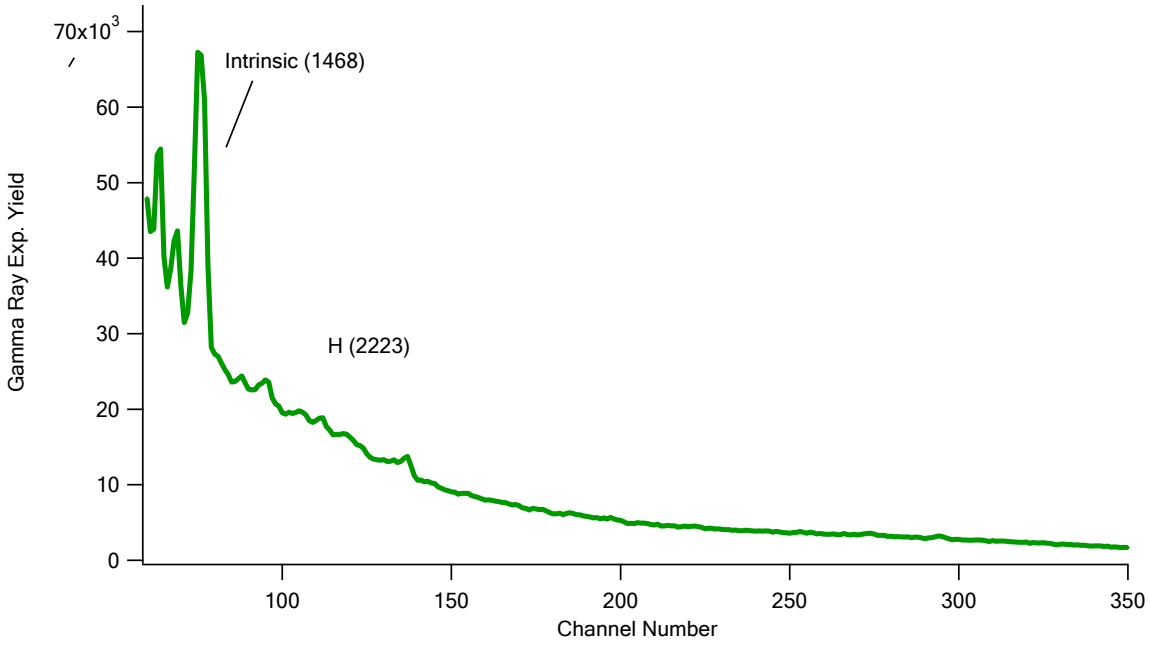

Figure 8**:** Full prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector for the Background, taken with 14 MeV neutrons PGNAA setup.

3.2 Prompt Gamma Analysis of Crude Oil Samples

Carbon and Hydrogen concentrations in four crude oil samples, namely crude 369, crude 486, crude G-26, and crude G-57, were measured using PGAA technique. In order to distinguish the C and H peaks' locations in the crude oil samples, the crude -369 sample was superimposed to the water and benzene full spectrum as shown in figure 9.

The Crude -369 oil sample has a peak of the H peak and a C peak as shown in figure 9.

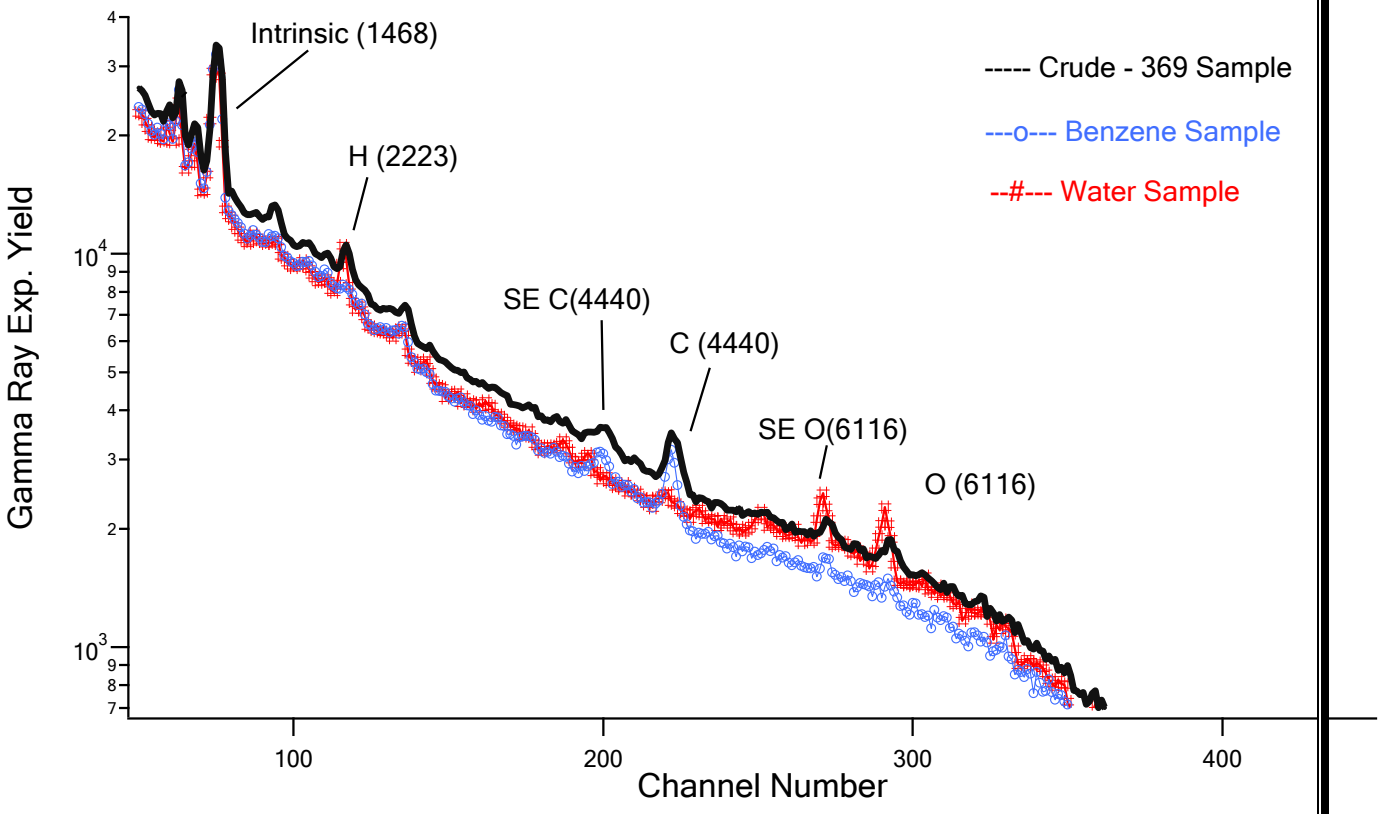

Figure 9: Full prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector for water and Benzene, and Crude – 369 oil sample taken with 14 MeV neutrons PGNAA setup.

Figure 10 shows full prompt gamma-rayspectra for the four crude oil samples superimposed upon the background spectrum.

Each sample has different concentrations of both carbon and hydrogen. The height of peaks is also varying from sample to another one.

Carbon to the Hydrogen (C/H) element concentration ratio determines the quality of the crude oil. The (C/H) element ratio for the samples was calculated from the experimental data for the samples. The background (figure 8) has been superimposed upon the four samples spectra (figure 10). Background has been subtracted from the full spectra in order to get the (C/H) element ratio.

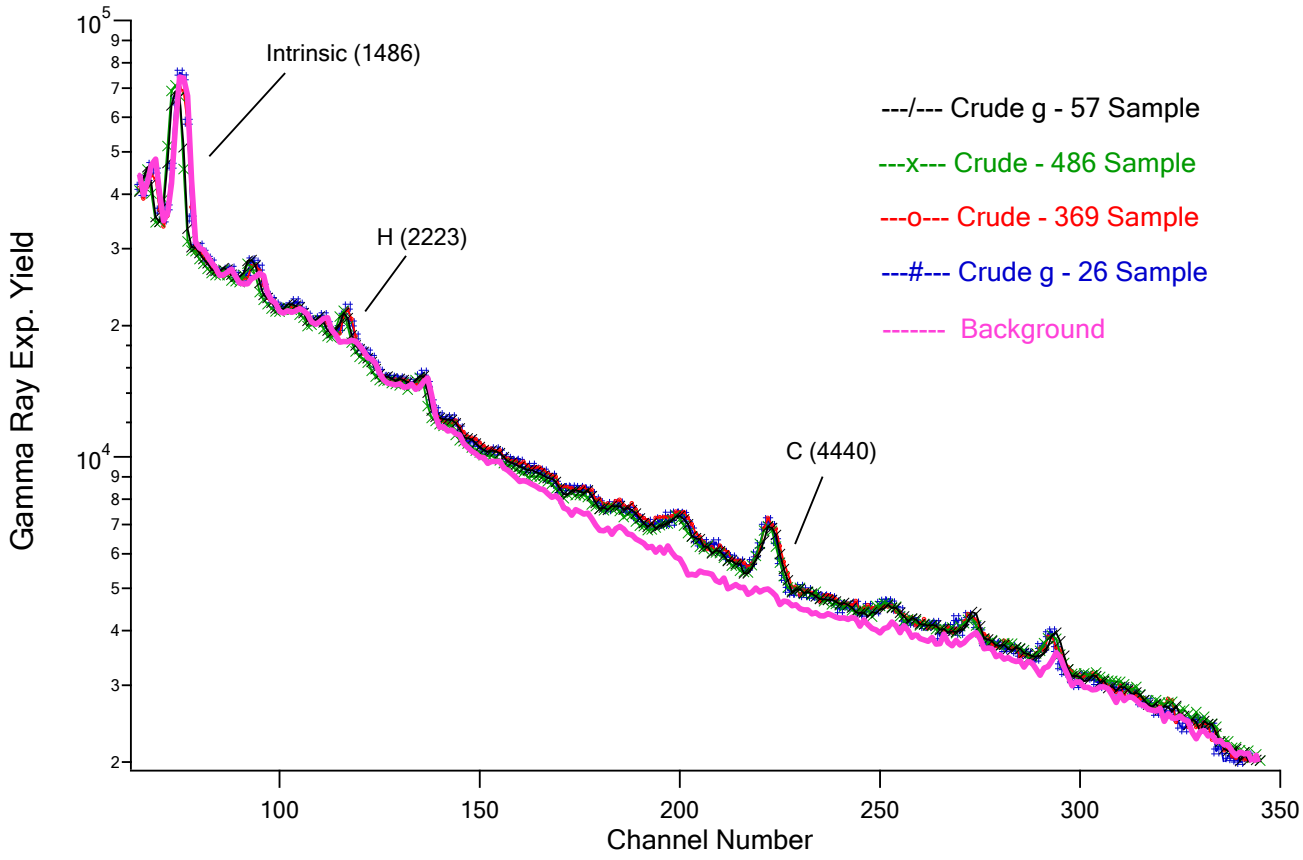

Figure 10: Full prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector of four crude oil samples plus the background spectrumtaken with 14 MeV neutrons PGNAA setup.

The H peaks of the four samples after subtracting the background are shown in figure 11, which display only an expanded part of graph 10 in order to examine purely the H peaks for the analysis purpose.

Similarly, another expanded part from figure 10 shows the carbon peaks only after subtracting the background are displayed in figure 12.

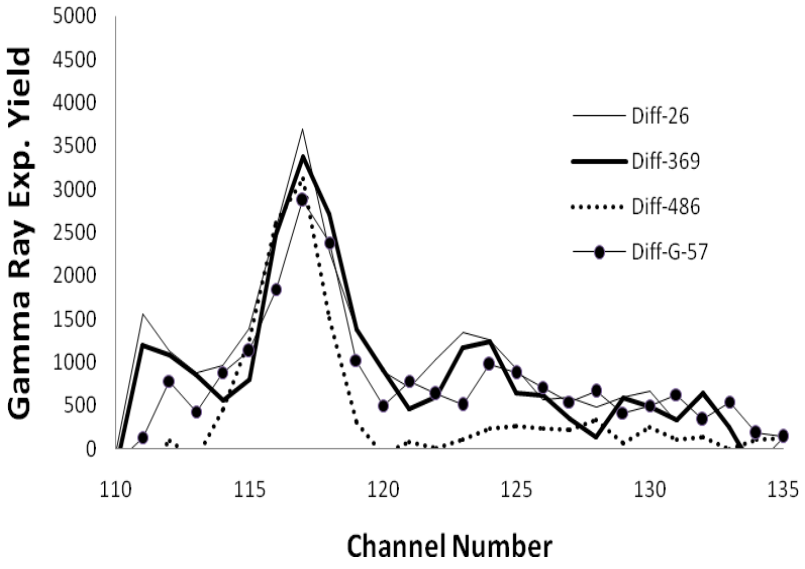

Figure 11: Prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector taken with 14 MeV neutrons PGNAA setup shows the hydrogen peaks for four crude oil samples after subtracting the background spectrum.

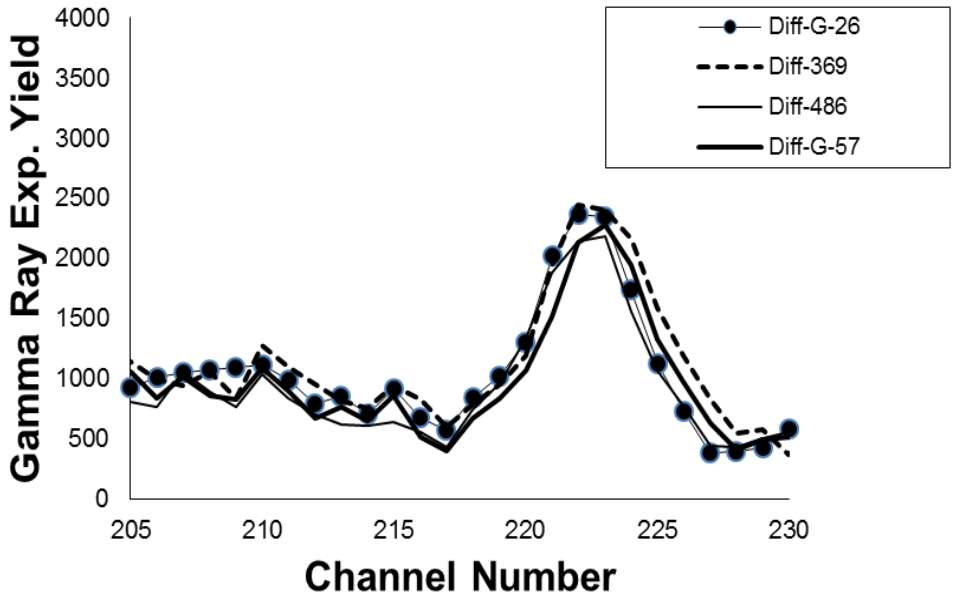

Figure 12: Prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector taken with 14 MeV neutrons PGNAA setup shows the carbon peaks for four ARAMCO crude oil samples after subtracting the background spectrum.

Table 2 summaries the detailed full spectra measurements for all crude oil samples. The counting time for all samples was 2011 seconds. The net counts for all samples were normalized to the same sample value. The (C/H) element ratio for the four samples is also shown.

Table 2: Summary of full prompt gamma ray spectra of $LaBr_3$:Ce gamma ray detector of four crude oil samples taken with 14 MeV neutrons PGNAA setup.

Sample Name	The Net C counts	The Net H counts	(C/H) Ratio
Crude-369	17967 ±3%	10639 ± 3%	1.689
Crude G-26	18427± 4%	13131 ± 4%	1.403
Crude G-57	17478 ± 3%	10539 ± 3%	1.658
Crude - 486	16869 ± 3%	9135 ± 3%	1.847

Samples with the higher (C/H) ratio like Crude - 486 has poorer quality (less liquidity), while the lower (C/H) ratio like in Crude G-26 has a better quality. The ratio is useful as a preliminary indication of the hydrogen quantity needed to convert the hydrocarbon to a gas and/or liquid [9].

Conclusions

The LaBr3:Ce gamma ray detector was used for detection of high energy prompt gamma-rays of four crude oil, water and benzene samples using a 14 MeV neutron-based PGNAA setup. Hydrogen and carbon concentration in bulk samples was determined via 2.222, and 4.440 MeV gamma rays respectively for four crude oil, water and benzene samples. The experiment shows that, in spite of its intrinsic activity, the LaBr3:Ce gamma ray detector has an excellent performance for the detection of hydrogen and carbon in crude oil, water and benzene samples. The concentration of hydrogen and carbon was experimentallymeasured, and then the carbon to the hydrogen (C/H) material ratio was calculated for each sample. Sample with the higher (C/H) ratio, like Crude – 486, has poorer quality (less liquidity), while the lower (C/H) ratio sample, like in Crude G – 26, has a better quality. The ratio is useful as a preliminary indication of the hydrogen quantity needed to convert the hydrocarbon to a gas and/or liquid.

Acknowledgement

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