جامعة الملك فهد للبترول والمعادن
King Fahd University of Petroleum & Minerals

Physics Dept.

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PROJECT

"Measurement of Carbon Concentration in Bulk Hydrocarbon Samples"

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Abstract

In this experiment, PGNAA technique was used with $LaCl_3$:Ce gamma-ray detector to measure carbon concentration in : Propanol (C_3H_8O), Porpanic-Acid ($C_3H_6O_2$), Butyl-Alkhol ($CH_3(CH_2)_3OH$ and Formic-Acid (HCOOH) samples. Water and benzene samples were used for peak identification and energy calibration of the $LaCl_3$:Ce gamma-ray detector based on PGNAA setup. It is found that, yield of prompt gamma ray increases linearly with increasing in the concentration of carbon in the sample, showing excellent performance of the $LaCl_3$:Ce gamma-ray detector in detection of carbon in bulk sample using 14 MeV inelastic scattering.

1. Introduction

Prompt gamma-ray neutron activation analysis (PGNAA) is ideally suitable for non-destructive in-situ measurements of elemental concentration of bulk samples [1]. Applications of prompt gamma-ray neutron activation analysis technique is increasing in various industrial, environmental, homeland security, medical discipline due to continuous development in gamma ray detection capability such as improved energy resolution and detection efficiency[2]. Experimental requirements differ from one application to another; each type of application requires specialized gamma-ray detectors, neutron source energy, intensity, etc. application involving detection of low energy prompt gamma-rays requires gamma detectors with excellent energy resolution while applications involving detection of elements with low neutron capture or scattering cross section requires intense neutron source [1]. Recently, lanthanide-halide $LaBr_3$:Ce and $LaCl_3$:Ce gamma-ray detectors are available in large crystal sizes and they exhibit improved energy resolution and faster light decay time compared to conventional NaI and BGO gamma-ray detectors [3]. In this work, PGNAA technique with $LaCl_3$:Ce gamma-ray detector were used to measure carbon concentration in: Propanol (C_3H_8O), Porpanic-Acid ($C_3H_6O_2$), Butyl-Alkhol ($CH_3(CH_2)_3OH$ and Formic-Acid (HCOOH) sample, concentration calibration for carbon was also obtained. Water and benzene samples were used for peak identification and energy calibration of the $LaCl_3$:Ce gamma-ray detector based on PGNAA setup.

2. PGNAA Technique

PGNAA was developed for detection of concealed explosives in airline luggage during the late 1980s and is now being investigated and tasted for new applications. These applications include detection of drugs in passenger luggage, detection of explosives in bottles, and detection of buried land mines and unexploded ordnance. For explosive, the PGNAA is based on the 10.8 MeV capture gamma ray from nitrogen, whose high density is uniquely characteristic of modern high explosives. In Neutron Activation Analysis (NAA), neutrons are used to bombard a simple, and then neutrons interact with atoms in the sample to create new radioactive isotopes. As these radioactive

nuclides decay, they emit gamma rays whose energies characteristic for each nuclide. By measuring the gamma rays released when these isotopes decay, it is possible to determine which elements are present and their concentration by comparison of intensities of these gamma rays. NAA technique is divided in two branches, with respect to a major experimental parameter; whether nuclear decay products (gamma rays) are measured during neutron irradiation (PGNAA), or at some time after irradiation (DGNAA) as shown in Figure 2.1. In the PGNAA technique, a material is irradiated with fast neutrons, some are moderated by the material in an external moderator. These neutrons interact with the material through neutron inelastic scattering $(n, n'\gamma)$ which this experiment based on, or thermal neutron capture (n_{th}, γ) or both. Since we are going to detect carbon, Figure 2.2 shows prompt gamma rays energies due to inelastic scattering of neutrons from C, N and O elements. To sum up, in PGNAA technique, the method is to excite prompt gamma either by thermal capture of thermal neutrons or by inelastic scattering of fast neutrons which used for nuclei like C, O and N having very small thermal capture.

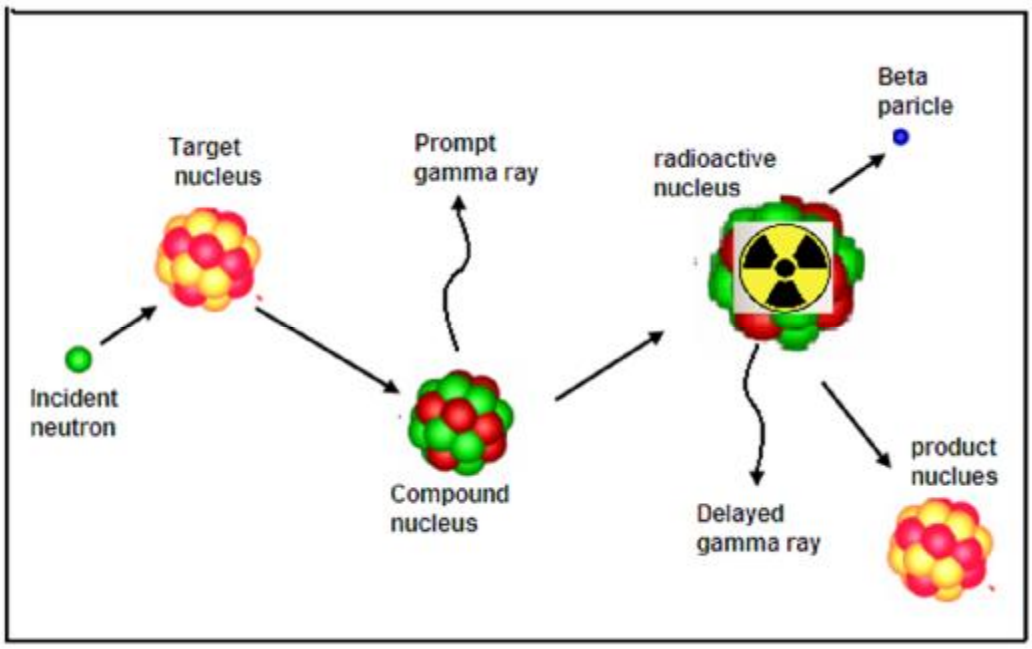

Figure.2.1: Schematic diagram illustrates nuclear decay products, prompt and delayed gamma ray

but there are some advantages in choosing the prompt gamma rays produced by thermal neutron capture for elemental analysis. The prompt gamma rays, due to thermal neutron

capture have generally higher energy as compared to those from neutron inelastic scattering. Accordingly, it is easier to detect higher energy thermal capture gamma rays. Prompt gamma rays neutron activation analysis (PGNAA) is non-destructive technique and is widely used for identification and quantification of elements in bulk gas, liquid, or solid samples. For detection of drugs, the use of capture gamma ray signals from both hydrogen and chlorine (from hydrochloride drug salt) have been investigated, and specific set of features based on these gamma ray signals selected for detection algorithm. The technique is ideally suited for determination of elements concentration such as carbon, boron, chlorine, cadmium [4].

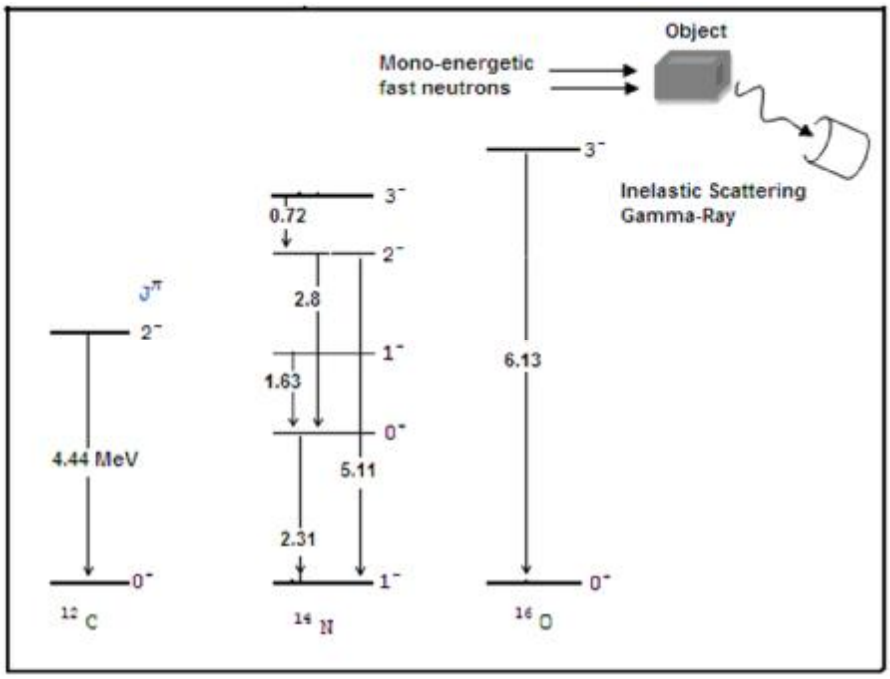

Figure.2.2: Gamma ray decay scheme from inelastic scattering of neutrons from C, N and O

3. Experimental setup

The response of the detector was measured for high energy prompt gamma rays using the 14 MeV neutron-based PGNAA setup shown in Fig. 3. [5]. The setup consisted of a 9.0 cm Φ x 14.0 cm cylindrical plastic container filled with the sample material and placed 7.0 cm in front of the water-cooled tritium target, at 0o angle with respect to the 14 MeV neutron beam. The gamma ray detector was placed at a center-to-center distance of 11.9 cm from the sample, at an angle of 90 degrees with respect to the 14 MeV neutron beam. Tungsten blocks were inserted between the tritium target beam line and the gamma ray detector to shield it from direct beam of 14 MeV neutrons. Furthermore, the detector was shielded from the 14 MeV neutron-induced gamma ray background by lead blocks inserted between the detector and the tungsten shield.

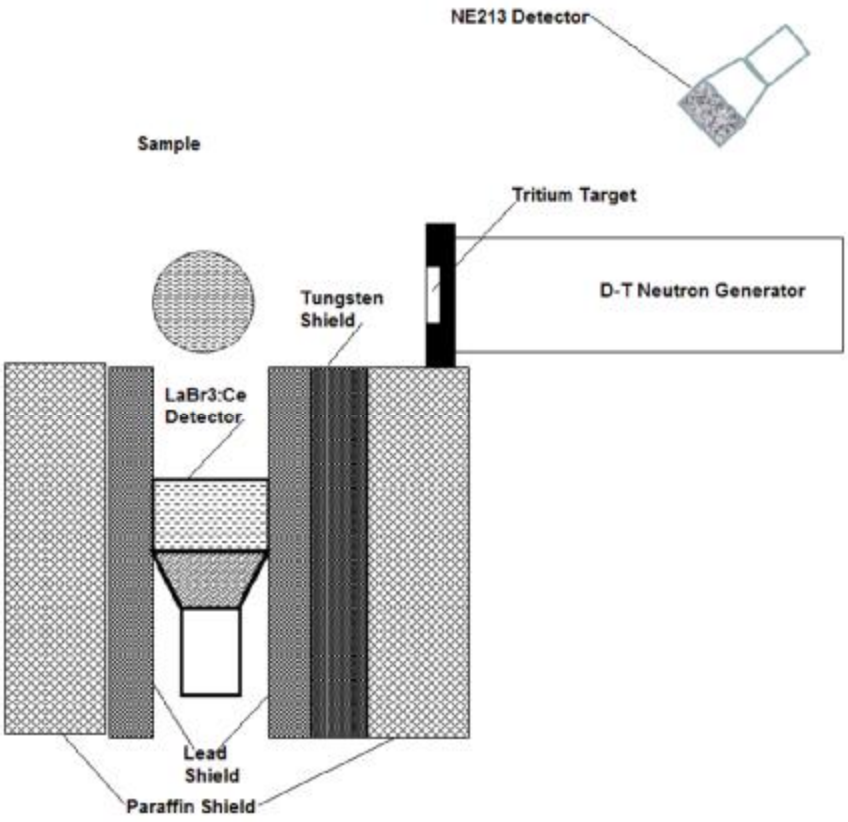

Fig.3: Schematic of 14 MeV neutron-based setup used for measurement of H, C and O concentration in bulk samples.

Finally, a paraffin shield was built around the detector enclosing the lead and tungsten blocks, to shield it from room-scattered neutrons. The paraffin shield was prepared by mixing lithium carbonate and paraffin wax in equal weight proportions. Although the paraffin and lead shields were effective in shielding the detector against scattered neutrons and background gamma rays, the gamma ray peaks due to the inelastic scattering of 14 MeV neutrons from lead were quite pronounced in the gamma ray detector pulse height spectrum.

A pulsed beam of 14 MeV neutrons was produced via the T(d,n) reaction using a pulsed deuteron beam with 200 ns width and a frequency of 31 kHz. The typical pulsed beam current of the accelerator was 60 μA dc beam current. As shown in Fig. 3, the fast neutron flux from the tritium target was monitored using a 7.6 cm Φ x 7.6 cm cylindrical NE213 fast neutron detector, placed at a distance of 1.8 m from the target and at a backward angle of 130 degrees with respect to the beam axis on the opposite side of the LaBr3:Ce detector gamma-ray detector. The 14.8 MeV neutron flux was measured to be 106 n/cm2/s. The prompt gamma-rays spectrum of the $LaCl_3$:Ce was recorded for a preset time.

4. Results and Discussion

4.1 Energy Calibration of $LaCl_3$: Ce Detector

Detectors with its components do not give us a direct reading for energy versus intensity, rather it is a relationship between intensity and channel number so that, before any measurement we need to calibrate our detector and peak identification. For this purpose two samples, water and benzene, were used. Also because our main goal is to detect carbon and measure its concentration, it is useful to determine in advance the position of the carbon and oxygen peaks. Figure 4 shows the Pulse height spectrum of the LaCl: Ce detector for water and Benzene samples.

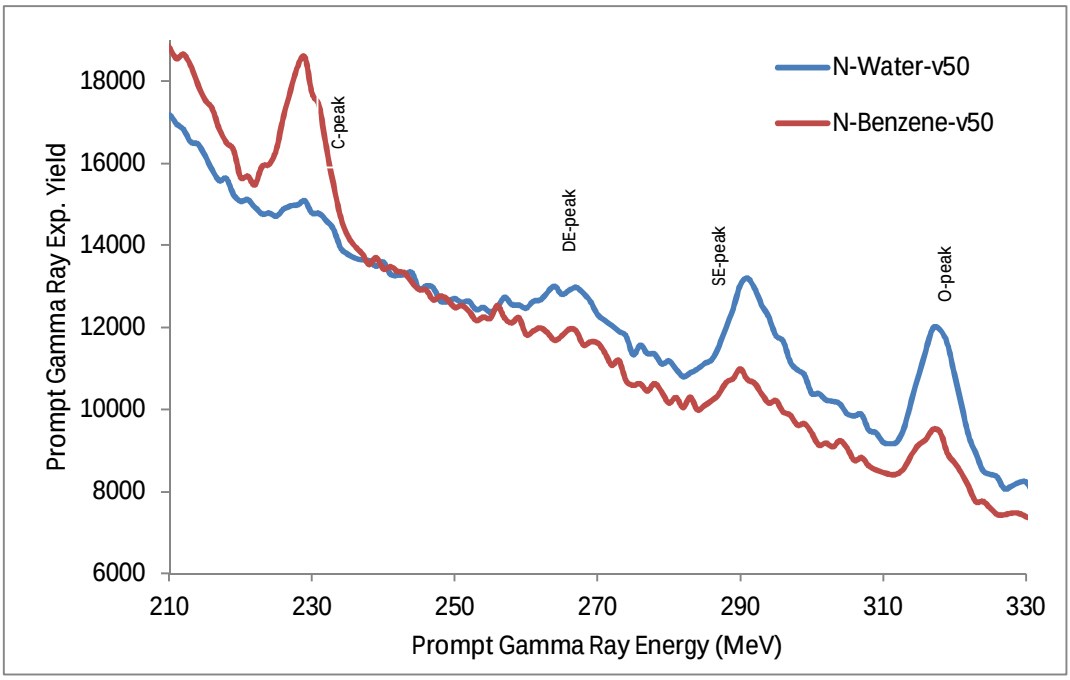

Fig. 4: Pulse height spectrum of the LaCl: Ce detector for water and Benzene.

For water, Oxygen's concentration (H_2O) is very high comparing to hydrogen and it is zero for carbon. In contrast, for benzene (C_6H_6), Oxygen's concentration is zero but it is high for carbon. Therefore, water can be used to determine the position of oxygen and benzene for carbon, the spectrum of water, in the figure above, shows the three peaks: oxygen, single escape and double escape 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 table below shows the Prompt Gamma ray energy corresponding to each channel number.

Peak	Ch#	Energy (MeV)
C	**228**	**4.44**
DE	**264**	**5.1**
SE	**290**	**5.62**
O	**317**	**6.13**

Table 4: Prompt Gamma ray energy corresponding to each channel number.

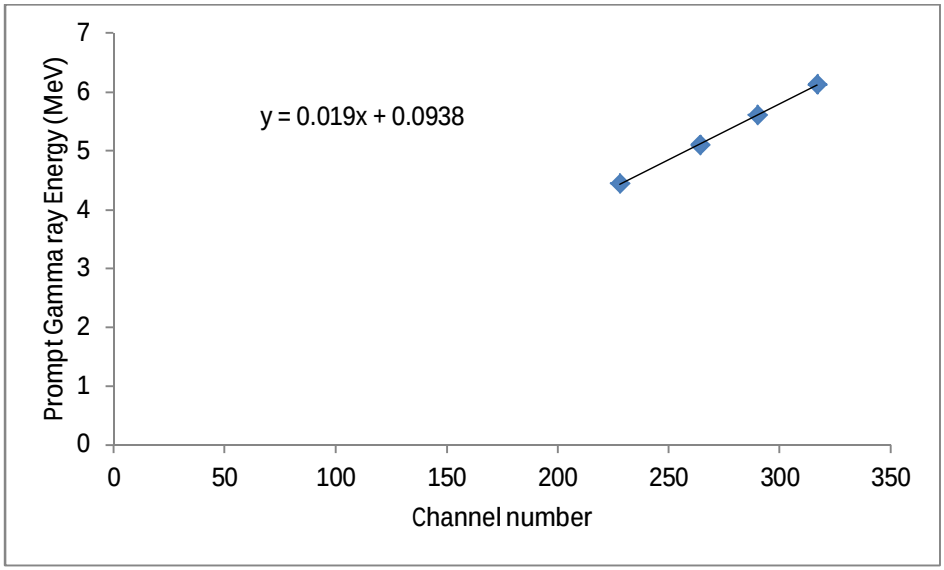

Fig 4.1: Energy calibration curve of the LaCl: Ce detector

Figure 4.1 shows the energy calibration graph for $LaCl_3$:Ce gamma-ray detector. The calibration equation of the detector is given by

$E\gamma$(MeV) = 0.019×X + 0.0938, where X is the channel number.

4.2 Prompt Gamma Analysis of Carbon in Propanol, Propanic-Acid, Butyl-Alkhol, Formic-Acid Samples.

Detecting carbon was obtained in four different samples; Propanol (C_3H_8O), Porpanic-Acid ($C_3H_6O_2$), Butyl-Alkhol ($CH_3(CH_2)_3OH$ and Formic-Acid (HCOOH). Each one has a different concentration of carbon which can be used for calibration. Table 5 shows the Elemental composition of Propanol, Propanic-Acid, Butyl-Alkhol, Formic-Acid which reveals the different concentration of carbon.

Compounds	Chemical Formula	C (wt%)	O (wt%)	H (wt%)
Propanol	**C_3H_8O**	**60**	**26**	**13.3**
Propanic-Acid	**$C_3H_6O_2$**	**48.6**	**43.2**	**8.1**
Butyl-Alkhol	**$CH_3(CH_2)_3OH$**	**64.8**	**21.6**	**13.5**
Formic-Acid	**HCOOH**	**26**	**69.5**	**4.3**

Table 5: Elemental composition of Propanol, Propanic-Acid, Butyl-Alkhol, Formic-Acid

Using LaCl3:Ce gamma-ray detector, the obtained spectrum for all samples is shown in Figure 5.1. It reveals the two original peaks correspond to carbon (4.44 MeV) and oxygen (6.13 MeV) with different intensity depending on the concentration of element in each sample. The other two peaks (5.6, 5.1 MeV) correspond to the single and double escape. Regarding the hydrogen peak (2.2 MeV), it is not appears in the spectrum because it is in the low energy range. In this work we are going to investigate the concentration of carbon versus the Prompt Gamma Ray Exp. Yield. Figure 5.2 shows the Pulse height spectrum of the LaCl: Ce detector for carbon peak in all samples. It is reveals that the higher concentration of carbon in the sample gives the higher prompt gamma ray experiment yield.

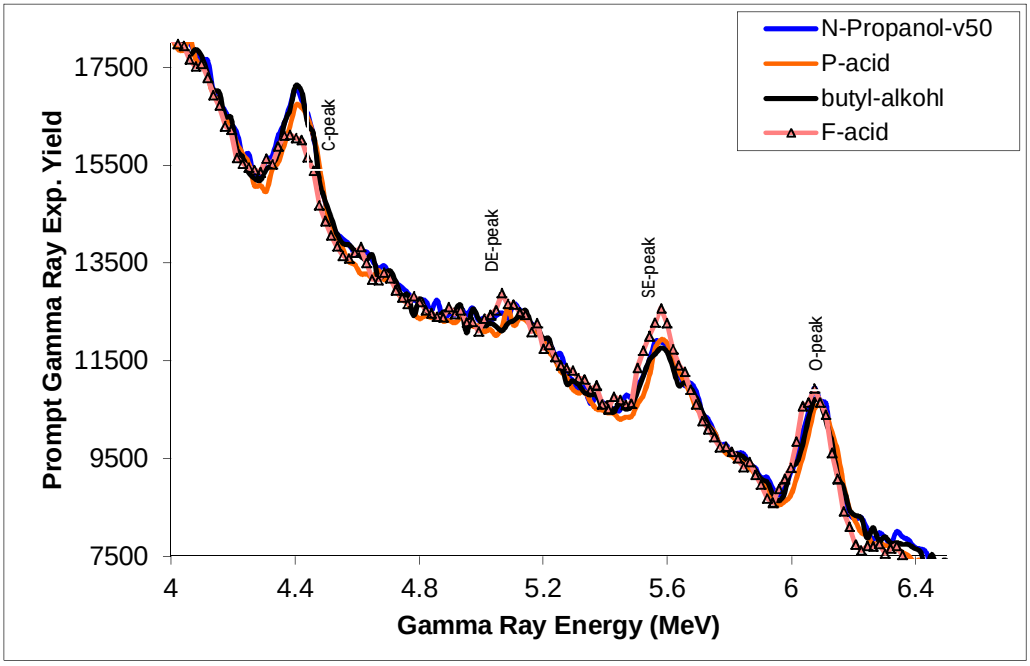

Fig5.1: Pulse height spectrum of the LaCl: Ce detector for O, C, SE and DE peaks in samples

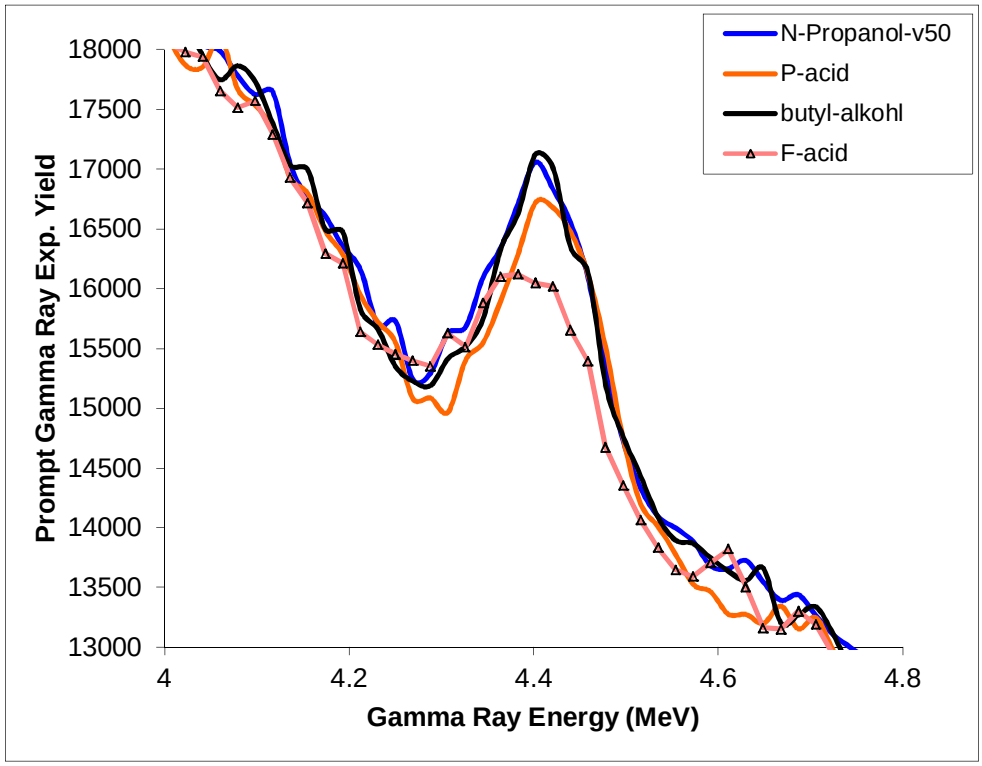

Fig5.2: : Pulse height spectrum of the LaCl: Ce detector for carbon peak in all samples

Figure 5.3 shows the yield of 44.4 MeV prompt Gamma Ray of carbon from four samples plotted as a function of carbon concentration. It reveals that yield of prompt gamma ray increases linearly with increase in the concentration of carbon in the sample

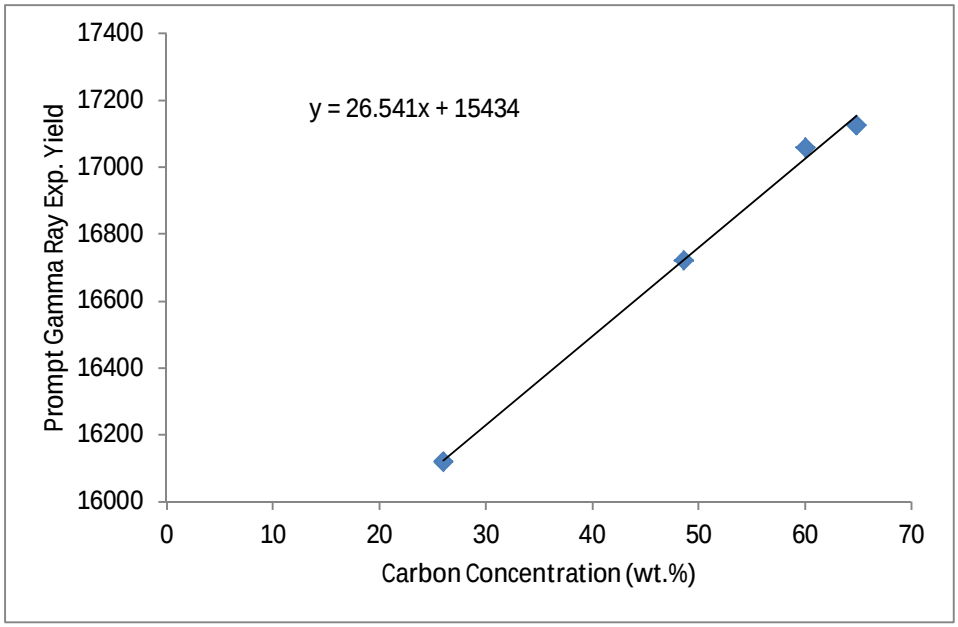

Fig5.3: Yield of 44.4 MeV prompt Gamma Ray of carbon from four samples

5. Conclusion

In this experiment, PGNAA technique was used with $LaCl_3$:Ce gamma-ray detector to measure carbon concentration in : Propanol (C_3H_8O), Porpanic-Acid ($C_3H_6O_2$), Butyl-Alkhol ($CH_3(CH_2)_3OH$ and Formic-Acid (HCOOH) samples. Water and benzene samples were used for peak identification and energy calibration of the $LaCl_3$:Ce gamma-ray detector based on PGNAA setup. It is found that, yield of prompt gamma ray increases linearly with increasing in the concentration of carbon in the sample, showing excellent performance of the $LaCl_3$:Ce gamma-ray detector in detection of carbon in bulk sample using 14 MeV inelastic scattering. Due to the energy peak of chlorine (Cl) 6.11 MeV, which is close to oxygen's peak 6.3 MeV, makes the measurements of oxygen's concentration not accurate result in the interference of the two peaks.

6. References

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