FABRICATION OF AN Am-Be BASED THERMAL NEUTRON REFLECTION ANALYSIS FACILITY

BY

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CERTIFICATION

This thesis entitled "**Fabrication of an Am-Be based thermal neutron reflection analysis facility**" by Issu Ibrahim Zachari meets the regulations governing the award of the degree of Master of Science of Ahmadu Bello University, and is approved for its contribution to knowledge and literary presentation.

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ABSTRACT

A 5ci Am-Be source based experimental set up for the determination of H content and $\frac{0+C}{H}$ atomic ratio in oil or hydrocarbon samples and coal or solid mineral samples has been developed at the Centre for Energy Research and Training (CERT).

It utilizes the thermal neutron reflection technique in which the relative intensity of thermal neutrons reflected by a bulk sample is used to determine its total hydrogen content. Use is made of chemicals of known chemical formula and carbon black of known carbon content to obtain the calibration line which establishes the relation between the hydrogen content, $\frac{0+C}{H}$ atomic ratio and the reflected thermal neutrons.

The fabricated set up was used to analyze edible and non-edible oil samples. The samples quality was relatively graded in terms of the hydrogen content and $\frac{0+C}{H}$ atomic ratio. The detection limit of 0.25 H w% was obtained for hydrogen.

The adaptation of this technique for quality control of petroleum products was established by the analysis of used and un-used lubricants. The facility is economical and recommended for use in the determination of moisture content of solid minerals and building materials.

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CHAPTER ONE

1.0 INTRODUCTION

1.1 GENERAL INTRODUCTION

The subject of nuclear physics and indeed nuclear technology as it is today can be said to have begun with the discovery of the neutron. Soon after its discovery by James Chadwick in 1932, and even before its properties were fully understood, the neutron has been used to probe the structure of the nucleus via the neutron-nuclei interactions [Marion and Fowler, (1960), Willis,(1973)].

The use of neutrons for analytical purposes had led to the development of very powerful methods for quantitative determination of elements and isotopes in various fields of endeavour (Rakovic, 1970). It is a valuable tool for exploring the structure of the nucleus because of its characteristic electrical neutrality, possession of magnetic moment and a wide range of energy from the "cold" region (E_n < 0.025eV) to the spallation region (E_n > 100 MeV).

The adaptation of analytical techniques based on neutrons for nuclear data studies and bulk sample analysis can be used to initiate new ideas in basic research and various fields of application.

The fast neutron moderation technique is recommended for

the measurement of moisture content in bulk materials with concentrations under one weight percent [Semel and Helf, (1968), Jonah *et al.*,(1997)].

The development of new methods for the determination of hydrogen content in oil samples which can be used for quality control of finished products is of continuing interest especially for petroleum products used in Nigeria.

1.2 A REVIEW OF PREVIOUS WORK

The thermal neutron reflection technique was first developed for the determination of bitumen content is asphalt concrete (Buczko *et al*, 1975). This was based on the flux abbedo developed in 1933.

Roth, 1966 has also reviewed ten instrumental methods for measuring the moisture content of solids based on the same neutron physical method. The method is recommended because it satisfies the needs of most quality control requirement with regard to specificity, sensitivity, speed of response, ease of operation and cost.

Neutron physical methods based on two different approaches have been used for the measurement of moisture and or hydrogen contents in materials. One is concerned with the transmission of slowed down neutrons and the other with the reflection of slowed down neutrons [Semel and Helf,(1968), Gozani, (1988)].

Subsequently Al-Jobori *et al.*, 1978; Szegedi *et al.*, 1982; Szegedi *et al.*, 1990; Chimoye, 1991; Vourvopolous, 1993 and more recently, Yousif *et al.*, 1995; Jonah *et al.*,(1997).

have used these methods with improved instrumental techniques for the analysis of H_2 in different types of matrices.

A survey of the literature of the work enumerated and the availability of the ^{241}Am-Be source at the centre for Energy Research and Training, Ahmadu Bello University, Zaria motivated the adaption of the technique in this investigation.

1.3 PRESENT WORK

The present work is centered on the fabrication of experimental set-up with minimum flexibility using locally available materials. Unlike the situation with earlier set-ups (Buczko *et al*, 1975 and Jonah *et al*, 1995), the intensity of reflected thermal neutrons was determined via the activation method (Knoll, 1979, Nicholas, 1972) in this investigation.

The set up consists of a steel container housing the source and an aluminium sample container. The source holder is filled with a paraffin wax and a hole bored at the centre to house the ^{241}Am-Be source. The indium foil probe attached to a cadmium sheet is fixed to the sample container, then the sample container placed directly on the source in the paraffin filled source container for irradiation.

Use is made of the available ^{241}Am-Be source as a fast neutron source, the neutron from the source is utilized for the neutron data research and bulk sample analysis. The facility is designed to enable the use of the set-up as a thermal neutron reflection facility.

The facility was tested via the determination of the

total hydrogen content in different categories of oil samples. Furthermore, the determination of (O+C)/H atomic ratio in these samples was also carried out. Successful utilization of the facility lend credence to the possible adaptation of the set up for non-destructive and on-line quality control processes which is the major objective of this work.

CHAPTER TWO

2.0 THEORY AND PROPERTIES OF NEUTRON INTERACTIONS

A thorough understanding of the principles of neutron production and interaction together with a knowledge of the techniques involved in conducting investigations are a pre-requisite to the utilization of neutrons for research and analysis. Consequently, the fundamental properties of the neutron and its classification based on its kinetic energy are outlined. The interactions of neutrons with bulk matter and the techniques of measuring this interaction is discussed. General characteristics of isotopic neutron source used in this work is described. The thermal neutron reflection technique adapted in this investigation is briefly mentioned.

2.1 FUNDAMENTAL PROPERTIES OF NEUTRONS

The fact that neutrons possess magnetic moment could only result from some circulation of charge particles which, therefore necessitates the consideration that for a fraction of time the neutron could exist as a concentrated positive charge surrounded at a relatively large distance by a cloud of equal negative charge [Becon, (1969) and Morrison, (1951)]. Thus the assumption of zero net charge of neutrons could indicate an equal number of positive and negative charges. The neutron is therefore assumed to be entirely neutral.

Experimental results reveal that neutrons, natural or free are unstable, it undergoes beta decay into an electron,

a proton and an antineutrino (Beckurts and Wirtz, 1964).

$$n = P + \beta^{-} + \bar{\nu} \qquad \ldots 2.1$$

with a beta energy E_{β} = 782 kev and a half-life of a few hundred seconds.

Neutrons like electrons exhibit wave properties and obey the Fermi-Dirac statistics. Table 2.1 contain a summary of the basic properties of the neutron.

TABLE 2.1 BASIC PROPERTIES OF THE NEUTRON (From Curtis, 959)

Mass (kg)	Charge (e)	Spin ($\hbar$)	Half-life ($T_{\frac{1}{2}}$) S	Magnetic moment (μ) NM
1.675×10^{-27}	0	$\frac{1}{2}$	641 ± 8	$-1.913148 \pm 6.6 \times 10^{-5}$

2.2 CLASSIFICATION OF NEUTRONS

This is necessary for an understanding of the different types of interaction.

Neutrons are broadly classified into three major groups according to their energies as:

(i) Slow or low energy neutrons (En < 1 kev)

(ii) Intermediate energy neutrons (1 keV < En 500 kev)

(iii) Fast or High energy neutrons (En > 500 kev)

The above grouping are further subdivided in a more detailed classification as shown in Table 2.2.

TABLE 2.2 CLASSES OF NEUTRONS (Thewlis, 1961)

Neutron Type	Energy Ev	Temperature K	Velocity m/s	Wavelength m
Cold	0.001	11.6	4.37×10^{-1}	9.04×10^{-10}
Thermal	0.0253	295.0	2.20×10^{3}	1.80×10^{-10}
Epithermal	11.0	1.16×10^{4}	1.139×10^{4}	2.86×10^{-11}
Resonance	100.0	1.16×10^{6}	1.39×10^{5}	2.86×10^{-12}
Intermediate	10^{4}	1.16×10^{8}	1.39×10^{5}	2.86×10^{-13}
Fast	10^{6}	1.16×10^{12}	1.39×10^{7}	2.86×10^{-14}
High Energy	10^{8}	1.16×10^{12}	1.29×10^{8}	2.79×10^{-15}
Relativistic	10^{10}	1.16×10^{14}	2.99×10^{8}	1.14×10^{-16}

2.3 NEUTRON SOURCES

Free neutrons are hard to find in nature because any freely existing neutron is either absorbed quickly or decays on its own within a relatively short time.

Neutrons are therefore produced artificially by separating them from nuclei, particularly those in which they are loosely bound [Bechurts and Wirtz, (1964), Gordan and Peters, (1988)]. This could be achieved through various nuclear reactions and fission processes.

There are different types of neutron sources based on the type of reaction leading to the production of the neutron. The sources vary widely in physical sizes and complexity as do the intensities of the neutron beams they produce (Bacon, 1969, Leo, 1987). Most neutron producing devices can be classified under one of the following:

(i) Isotopic Neutron Sources

(ii) Neutron Generators

(iii) Nuclear Reactors.

The isotopic neutron source was used in this work, because of its portability and relatively low radiation hazard.

2.31 ISOTOPIC NEUTRON SOURCE

This is the simplest source of neutron production. It consists of a target material mixed with a naturally decaying radioactive component which supplies the bombarding radiation used in the release of neutrons. Such sources are usually small in size (approximating to a point source) and produce strong, almost isotopic beam of neutrons which is nearly constant in time.

Isotopic or radioactive neutron sources are in three categories depending on the type of reaction involved such as:

(a) Radioactive alpha-neutron sources

(b) Radioactive photo-neutron sources, and

(c) Spontaneous fission sources.

These radioactive neutron sources are differentiated by the nature of the target materials and by the radioactive nuclei producing the bombarding radiation. Fission sources produce neutrons by the natural or spontaneous emission of neutrons by some heavy nuclei like $^{252}_{Cf}$. A photoneutron source is an assembly of a gamma emitter (e.g. $^{124}_{Sb}$) and a suitable target material (e.g. beryllium or deuterium). When the nucleus of the target material absorbs gamma radiation from the emitter, neutron production through a (γ, n)

reaction takes place if, the incident gamma energy (3 MeV) exceeds the binding energy of a neutron in the target nucleus (1.665 MeV or 2.225 MeV) (Beckurts and Writz, 1964, Leo, 1987). The reactions are:

$$^{9}_{4}Be + \gamma = {}^{8}_{4}Be + {}^{1}_{0}n - 1.665MeV$$

$$^{2}_{1}H + \gamma = {}^{1}_{1}H + {}^{1}_{0}n - 2.225MeV$$

The radioactive alpha-neutron source can similarly produce neutrons using the reaction that led to the discovery of the neutron. This source is explained further below.

Each of the neutron sources mentioned above have its advantages and disadvantages and specific field of use, but generally the major drawback of an isotopic neutron source include the low yield of neutrons, broad neutron spectra and the inability to be turned-off at will.

2.32 RADIOACTIVE (α,n) SOURCES

The reaction that led to the discovery of the neutron is the most common reaction used for its production, i.e.

$$^{9}_{4}Be + {}^{4}_{2}He = {}^{12}_{6}C + {}^{1}_{0}n + 5.71MeV \qquad \ldots 2.31$$

A beryllium target was bombarded by α-particles produced by naturally occurring radioactive elements such as polonium, radium, plutonium and americium. Neutrons are produced with the resulting daughter atom (12 C) and an energy difference of 5.71 MeV released.

The (α,n) source is made by mixing the suitable target (Beryllium) with a strong alpha emitter in a suitable proportion. Beryllium is the most yielding target material because it has a low (α,n) reaction threshold. Others elements like boron, flourine and lithium have relatively low yields. For activation analysis purpose, Be is mostly suited, the other targets are usually used for special purposes such as stimulating a fission process (Leo, 1987).

The general characteristics of each (α,n) source type which makes it desirable or otherwise when considering the choice of radioactive (α,n) source are largely determined by the properties of the alpha-emitter involved. Some of them are summarized in Table 2.3 below.

TABLE 2.3 PROPERTIES OF THE α^- EMITTERS OF RADIOACTIVE Be (α,n) SOURCES (IAEA ISOTOPIC, 1988)

Nuclide	Half Life	Alpha Energy (MeV)	Yield Per 10^6 Alpha (ns^{-1})	Yield (nsCi)	Yield ($ns^{-1}g^{-1}$)	Average Neutron Energy MeV	τ-Dose at 1 m for 10^{10} n/s in Gy/n
^{238}Pu	89Y	5.50	-	$2.8x10^6$	$4.5x10^7$	4.0	< 0.01
^{239}Pu	24.36Y	5.14	65	$1.6x10^6$	$1.2x10^5$	4.59	< 0.01
^{210}Po	138d	5.30	73	$2.5x10^6$	$1.1x10^{10}$	4.54	<0.001
^{241}Am	458Y	5.48	82	$2.2x10^6$	$6.5x10^8$	4.46	0.01
^{244}Cm	18.1Y	5.79	100	$3.0x10^6$	$2.5x10^8$	4.31	< 0.01
^{242}Cm	163d	6.10	118	$4.0x10^4$	$x10^{10}$	4.16	< 0.01
^{226}Ra	1620Y	7.69-4.77	502	$1.1x10^7$	$15x10^7$	3.94	0.5
^{227}Ac	22Y	7.36-5.65	702	$1.5x10^7$	$1.7x10^9$	3.87	0.07

2.4 INTERACTION OF NEUTRON WITH MATTER

The interaction of neutrons with matter can lead to a variety of phenomena, depending on the energy of the incident neutron and on whether the interaction is with a nucleus or with a bulk matter (Nicholas, 1972; Knoll, 1979).

The interactions with nuclei are used for probing the structure of the nucleus whereby important parameters such as cross sections are determined. However the interactions with bulk media are used for quantitative and qualitative assay of such media.

The experimental work of interest in this investigation is based on the latter type of interaction.

2.41 SLOWING DOWN OF NEUTRONS IN BULK MATTER

Interaction of neutrons with materials in which encounters are made with many nuclei leads to a number of physical effects that are used in nuclear analytical techniques. In this type of interaction, the neutrons interacts with a nucleus, but both particles reappear after the reaction. A scattering collision is indicated as an (n,n) reaction. Scattering may be elastic or inelastic. Elastic, when the total kinetic energy of the two colliding particles is conserved, and the kinetic energy is simply redistributed between the two particles. In inelastic scattering part of the kinetic energy is given to the nucleus as an excitation energy. After the collision, the excited nucleus will decay by emitting one or more gamma rays, thus emitting a neutron with kinetic energy less than that of the incident (absorbed) neutron and the residual nucleus also left in an excited state. Scattering reactions are therefore responsible for neutron's slowing down. For energies greater than 1 MeV, first inelastic collision takes place then as the energy is reduced elastic collision is obtained. A clear understanding of the principles and technique of neutron interaction with bulk matter is via the theory of slowing down (Nicholas, 1972; Beckuts and Wirtz, 1964).

Neutrons produced in nuclear reactions have energies far above the thermal energy range. When such fast neutrons collide with the atoms of a scattering medium, loss of energy occurs simultaneously with the diffusion process. The target atom can be assumed to be free and at rest before the

collision as long as the energy of the neutron is greater than 1ev (Beckuts and Wirtz, 1964).

Considering slowing down in space independent case, i.e. an infinite medium with uniformly distributed source of energy and taking into account only elastic collisions with free atoms that are initially at rest, the principles and technique of slowing down of neutrons in bulk samples is analyzed by considering neutron of mass M_1 and of initial energy E_1, which collides with a free atomic nucleus of mass A initially at rest. The dynamic of the collision in both laboratory system and centre of mass system is presented diagrammatically in figures 2.1a and 2.1b respectively. The relationship between the velocities in both frames of reference is illustrated in figure 2.1c below, where

V_d is the velocity of the neutron after collision in C-system

V_m is velocity of the centre of mass (C-system)

V_1 is the velocity of the neutron before the collision

V_l is the velocity of the neutron after collision in the lab- system

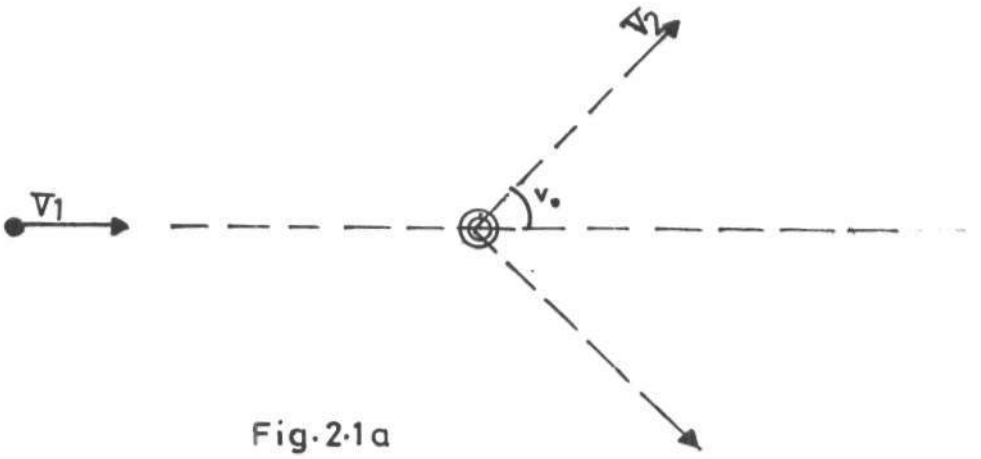
w_0 is the scattering angle in the lab-system

Ψ is the scattering angle in the centre of mass system.

From the diagrams and using the cosine formula, the equations obtained are:

$$\vec{V}_l^2 = \vec{V}_d^2 + \vec{V}_m^2 + 2\vec{V}_m \cdot \vec{V}_m \cos w_o \qquad \ldots 2.10$$

and



Fig·2·1a

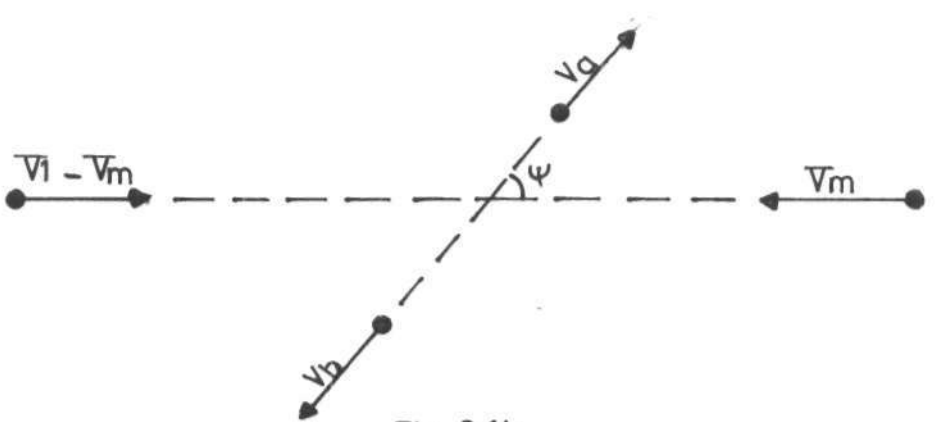


Fig. 2·1b

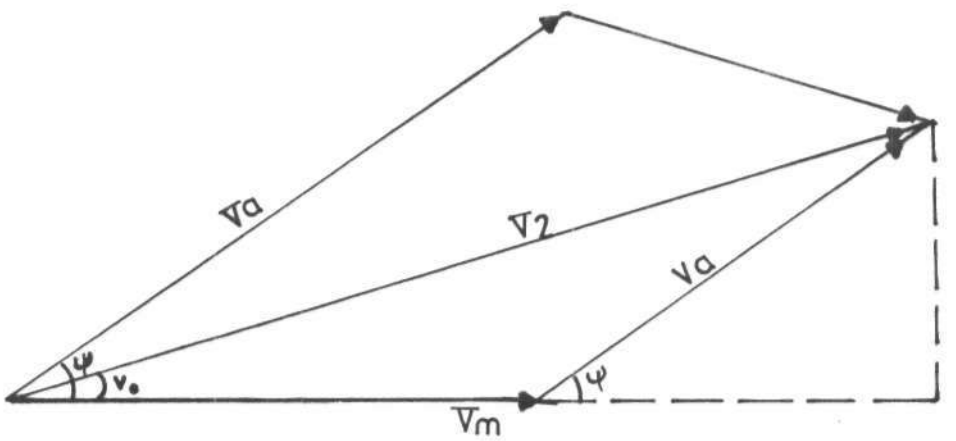


Fig. 2·1c

Fig·2·1: Dynamics of neutron collisions in ⓐ Laboratory system ⓑ Centre of mass system and ⓒ the relationship between the velocities in both systems

$$\vec{V}_2 \cdot \cos\alpha = V_2 \cos\phi + \vec{V}_m \qquad \ldots\ 2.11$$

The velocity of the centre of mass is obtained from the fact that the total momentum in the centre of mass system is equal to that of the laboratory system, using the theorem of momentum conservation. The equation obtained is thus:

$$(A+M_1)V_m = M_1 V_1 \qquad \ldots\ (2.12)$$

and given that $M_1 = 1$ (i.e. mass of neutron = 1)

We have $V_m = V_1/A+1$ ... 2.13

However the velocity of neutron in the centre of mass system before collision is given as:

$$V_1 - V_m = \frac{A}{A+1} V_1 \qquad \ldots\ 2.14$$

Furthermore in the centre of mass system, the momenta of the neutron and the nucleus are equal and oppositely directed before collision. After the collision they must still be equal and oppositely directed. Since the kinetic energy in the centre of mass system is also conserved during an elastic collision, the velocity of the neutron and that of the atomic nucleus must be same as the pre-collision velocities, to have the expression

$$V_m = \frac{A}{A+1} V_1 \qquad \ldots\ 2.15$$

The kinetic energy of the neutron after collision is given as

$$E_2=\frac{1}{2}M_1V_2^2 \qquad \ldots 2.16$$

From equations 2.9 and substituting equation 2.12 and 2.14 the expression obtained is

$$V_2^2=\frac{A^2}{(A+1)^2}V_1^2+V_1^2\frac{1}{(A+1)^2}+2\frac{AV_1^2}{(A+1)^2}Cos\Psi_o \qquad \ldots 2.17$$

Also from equation 2.17 we obtain the expression

$$V_2^2=V_1^2\frac{(A^2+2Acos\Psi_o+1)}{(A+1)^2} \qquad \ldots 2.18$$

$$\frac{M_1}{2}V_2^2=\frac{M_1}{2}V_1^2\frac{(A^2+2Acos\Psi_o+1)}{(A+1)^2} \qquad \ldots 2.19$$

$$E_2=E_1\frac{(A^2+2Acos\Psi_o+1)}{(A+1)^2} \qquad \ldots 2.20$$

where E_1 is the neutron energy before collision

E_2 is the neutron energy after collision

and Ψ_0 is the scattering angle on the centre of mass system.

The scattering angle in the laboratory system is given by

$$\cos\psi = \frac{A\cos\psi_0 + 1}{(A^2 + 2A\cos\psi_0 + 1)^{1/2}} \qquad \ldots 2.21$$

Next, introducing the auxiliary quantity

$$\alpha = \frac{(A-1)^2}{(A+1)^2} \qquad \ldots 2.22$$

The energy equation 2.20 becomes

$$\frac{E_2}{E_1} = 1/2\,[(1+\alpha)\cos\psi_0] \qquad \ldots 2.23$$

The maximum energy loss occurs in a head-on-collision i.e. collision for which $\psi_0 = 180°$ => $\cos\psi_0 = -1$

$$\Rightarrow E_2 = \alpha E_1 \qquad \ldots 2.24$$

The energy cannot fall below αE_1 as a result of a single elastic collision. For hydrogen $\alpha=0$ and $(E_2)_{min}=0$

For large A $\alpha \approx 1 - 4/A+2$ obtained from the expression below

The energy loss per collision i.e. $\Delta E = E_1 - E_2$ becomes

For a head-on-collision $\psi_0 = 180°$

$$E_2 = 4A/(A+1)^2 E_1 \qquad \ldots 2.26$$

thus for A=1 implies $\alpha = 0$ and $\Delta E = 0$ as the case for

hydrogen.

However for large A values equations 2.21 reduced to

$$\alpha = 1 - 4/A + 2 \qquad \ldots 2.27$$

The geometrical average of the ratio E_1/E_2 which is infact equal to ln E_1/E_2 defines a constant and is given as

$$\zeta = \ln E_1 - \ln E_2 \qquad \ldots 2.28$$

The last term of this equation leads to a physical interpretation on the average logarithmic energy decrement per collision. The maximum loss of energy is given as

$$E_3 = \frac{(A-1)^2}{(A+1)^2} E_1 \qquad \ldots 2.29$$

The scattering is isotropic in the centre of mass system,

$$\therefore \zeta = 1 + \frac{(A+1)^2}{2A} \ln \frac{(A-1)}{(A+1)} \qquad \ldots 2.30$$

ζ = 1 for hydrogen and for large A it can be approximated as

$$\zeta = \frac{2}{A + 2/3} \qquad \ldots 2.31$$

The quantity ζ makes it possible to estimate the average number of collision (η) necessary to moderate a neutron with incident energy E_1 to E_2 and it is given as

$$\eta = \frac{\ln(E_1/E_2)}{\zeta} \qquad \ldots 2.32$$

The value of η obtained in this form is only approximate since thermalisation effect at low energies (<0.025eV) and non isotropic scattering in the C-system at higher energies have not been taken into account. Interest is only on our working energy range.

CHAPTER THREE

3.0 MATERIALS AND METHODS

The choice of a particular type of experimental technique out of the several techniques available for investigating neutron interactions depends on the parameter of the neutron interaction under investigation.

The knowledge of total hydrogen content and (O+C)/H atomic ratio in hydrocarbon is a very important means of establishing their structure and purity. Since oil is build primarily from carbon and hydrogen, the quality of crude oil, their by-products and edible oils can be determined from the knowledge of these quantities.

Interest here is on the fabrication of a facility that can be used to measure the hydrogen/moisture content in bulk samples using the neutron physical method. Advantage is taken of the large thermal neutron scattering cross section of hydrogen (Semel and Helf, 1968).

3.1 THE NEUTRON PHYSICAL METHODS

Among all the methods considered for the determination of hydrogen/moisture, the interaction of neutrons offers the greatest potential for satisfying the quality control requirements. There are two available methods that measure the thermal neutrons resulting from the neutron interaction.

One of the methods is concerned with the transmitted thermal neutrons and the other method is concerned with the reflected thermal neutrons by the sample.

The thermal neutron transmission approach has been successfully applied in the determination of hydrogen in hydrocarbons and other organic liquids (Mott and Rhodes, 1966; Mustapha, 1992). The sensitivity of this method can be improved upon by the use of more intense neutron sources such as a high flux thermal neutron beam from the nuclear reactor or a neutron generator. Such facilities are considered to be economically impractical for in-plant quality control. The method of reflection of moderated neutrons for which a high sensitivity has been achieved was used for this work.

3.2 THE THERMAL NEUTRON REFLECTION TECHNIQUE

Generally, neutrons produced by a laboratory neutron source have energies far above the thermal region. Fast neutrons, on passing through materials of high scattering and stopping power are slowed down through elastic and inelastic processes, until they have equilibrium velocity with the atoms in the medium (Curtis, 1959). Practically some neutrons will be lost during the process, either from the boundaries of the finite medium or through the capture process or both, thereby giving rise to thermal neutrons. The elastic scattering process dominates the thermalisation procedure for hydrocarbons.

The escape of thermalised neutrons can be as a result of reflection from a material for which the intensity depends on its hydrogen content. In the thermalisation process, the kinetic energy of the colliding neutron is practically

transferred to the target nuclei. The average energy loss per collision is given as

$$\Delta\bar{E} = E_1\left(\frac{1-\alpha}{2}\right) \qquad \ldots 3.1$$

where ΔE is the mean energy loss per collision

E_1 is the energy of the incident neutron

and

$$\alpha = \left[\frac{A-1}{A+1}\right]^2 \qquad \ldots 3.2$$

as defined in equation 2.22. where A is the mass number of the nuclei.

From the above equations 3.1 and 3.2 it is evident that the intensity of the thermal neutrons strongly depends on the mass number of the medium.

Thus the fractional loss in energy when a neutron collides with an atom is greatest for the hydrogen atom. Consequently when a beam of fast neutrons is passed through Hydrocarbon materials of varying density, the intensity of the thermal neutrons detected is mostly a result of the density of the hydrogen content in the sample and as such a relationship would be observed between the hydrogen content and the measured thermal neutron intensity.

The most widely used method of detecting the thermal neutrons is by use of BF_3 counter via the direct counting method (Jonah,et al., 1997). However in this work activation detector foil was adopted as the neutron probe. Hence high

purity (>99%) indium foil was used as the neutron probe.

Radioactivity measurement was carried out by a 7.6cm x 7.6cm NaI(Tl) detector. The detector was connected to a computer based multichannel analyser as displayed by the block diagram, of Fig. 3.1.

For a given foil mass, considering a constant geometry, the specific activity is calculated from the measured activity divided by the mass of the foil probe for the sample as A_{si} and the background as A_{s0}, the relative excess specific activity induced in the foil $(AS_i - AS_0)/AS_0$ normalised by the physical density ρi of the sample is related to the neutron reflection parameter, denoted by η and given below as

$$\eta = \frac{AS_i - AS_0}{\rho_i \; AS_0} \qquad \ldots 3.3 \text{ (Jonah, 1995)}$$

where AS_i and AS_0 are the specific activity with and without the sample. From an understanding of the fact that the specific activity induced in the foil probe is a function of the thermal neutron intensity which in turn is a function of the Hydrogen content of the sample. The value of η for a given geometry of the sample depends on its hydrogen content.

The nuclear data of the residual radionuclide for Indium used in this work is shown in Table 3.0

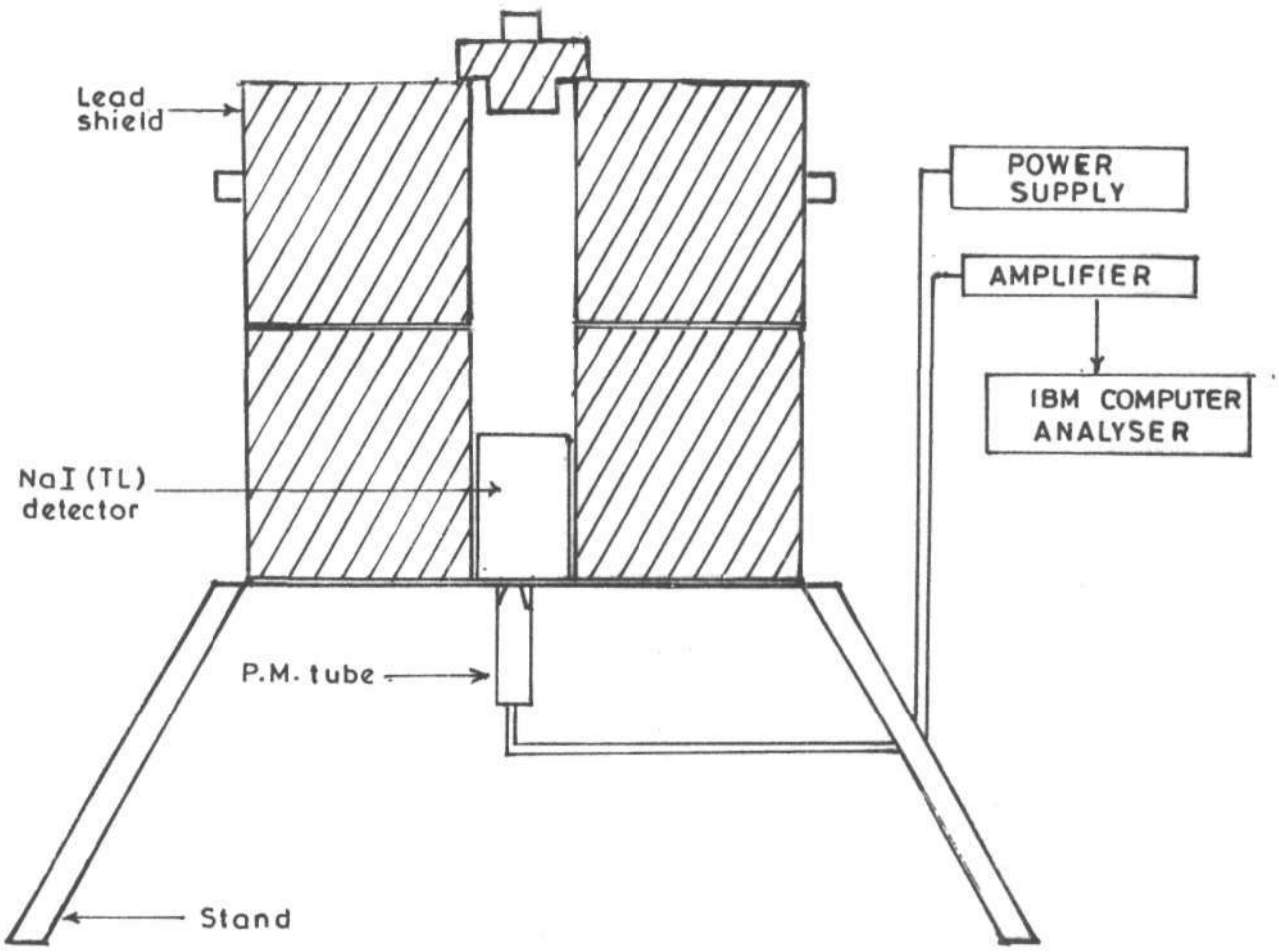


FIG. 3.1: GEOMETRICAL SET-UP OF NaI(Tl) DETECTOR AT CERT ABU [illegible]

TABLE 3.0 NUCLEAR DATA A OF NEUTRON FLUX MONITOR REACTION

REACTION	HALF-LIFE	E_γ (KeV)	I_γ (%)	σ (mb)
$^{115}In(n,\gamma)^{116}In$	54.12 min	416.86 1097.30 1293.54	29.20 56.20 84.40	1.62×10^5

3.3 FABRICATION OF THE FACILITY

The set up shown in Fig. 3.2 was adapted from an earlier experimental arrangement the "*BETATRON*" and was fabricated using locally available materials.

The major components of the facility includes a radioisotope fast neutron source, a suitable radioactive indicator or probe, a cadmium sheet and paraffin wax. The sample container and the source container were fabricated using aluminium and steel sheets respectively.

The design of the facility took into cognisance the source strength personnel and environmental protection from irradiation. The set up makes use of a 5ci Am-Be source which emits $1.29 \times 10^7 ns^{-1}$ ± 10%. Its active zone consists of a compact mixture of Americium oxide and beryllium metal, doubly encapsulated in stainless steel and sealed by argon arc. Welding. It has a cylindrical outer dimension of 30mm diameter x 48mm overall length.

The source container is cylindrical in shape and is of 30cm in diameter and 30cm in height. Since hydrogen is to be measured, aluminium is used to construct the sample container so as to avoid errors in detection of $(O+C)/H$ atomic ratio of the samples.

The sample holder constructed from a 1.5mm thick aluminium sheet is also cylindrical in shape and is of 10cm in diameter and 15cm in height. This is because interest is in bulk samples and the container was designed to take appropriate volume of liquid and solid samples.

Other requirements of the facility to shield personnel and detector from the radiation were considered. As such the source container was built big enough to moderate neutron, as well as to serve as a shield. Concrete wall was also constructed technically to block any stray neutron from the environment. In addition the use of NAA technique for counting allows the detection to be done with the detector in a secured environment for the personnel.

The facility has the neutron source embedded in the paraffin wax moderator and with the sample container placed directly on the source in a reproducible geometry. The indium foils are fastened on to the bottom of the sample holder and covered by a 1cm thick cadmium sheet to cut off the thermal neutrons emanating from the moderator. Consequently, with this arrangement it is ensured that only the thermal neutrons from the sample are reaching the detector.

Since this is a temporary set up, the experiments were carried out behind concrete block technically made into a shield of 1m thickness and of height 1.8m. the integrity of the shield was tested by the use of neutron and gamma detectors at various positions and heights above the floor of the laboratory while the set up was in place, and found

to be adequate for the work. A block diagram of this set up together with the source travel container in which the source is kept when not in use is shown in Fig. 3.3.

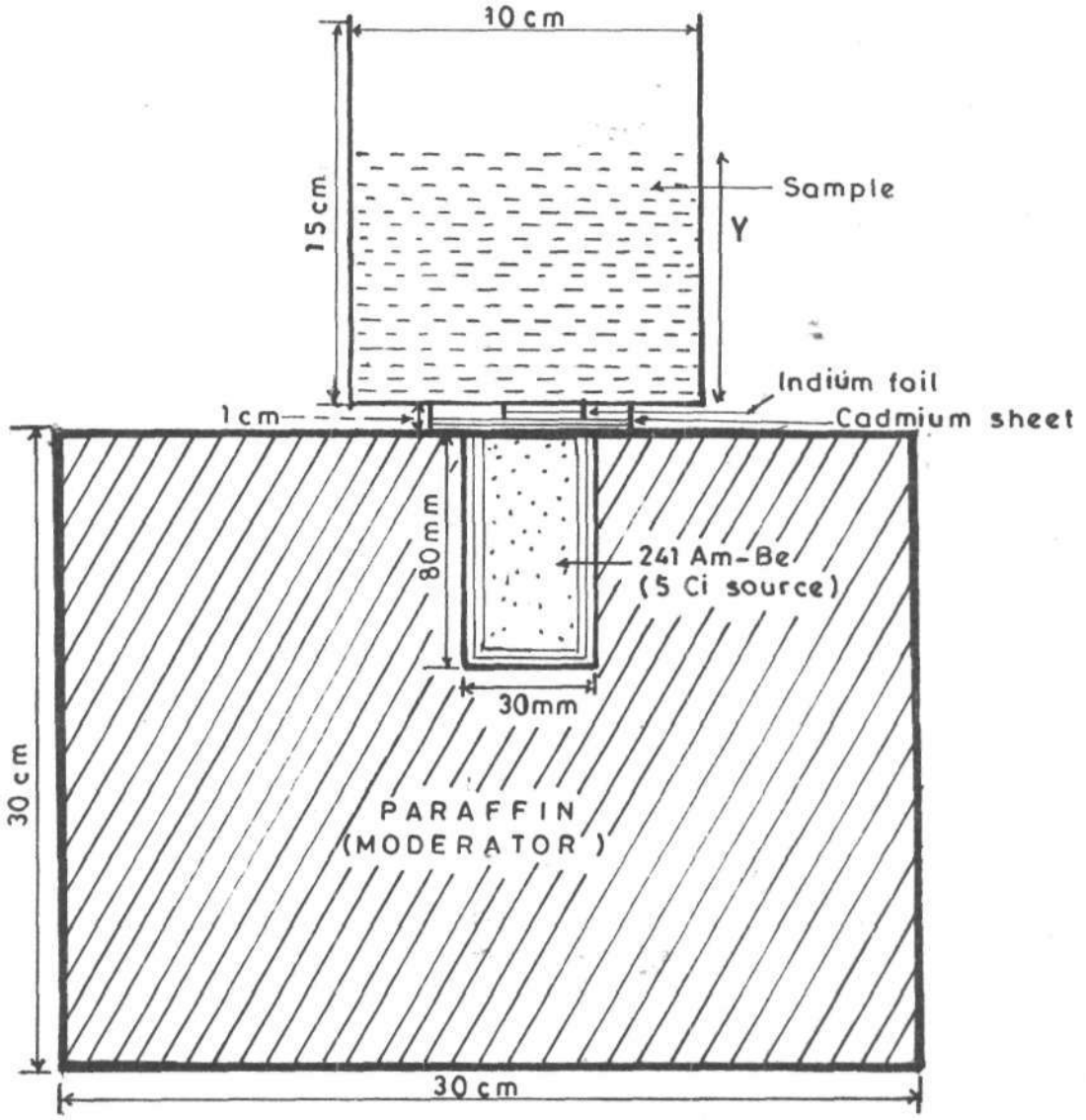
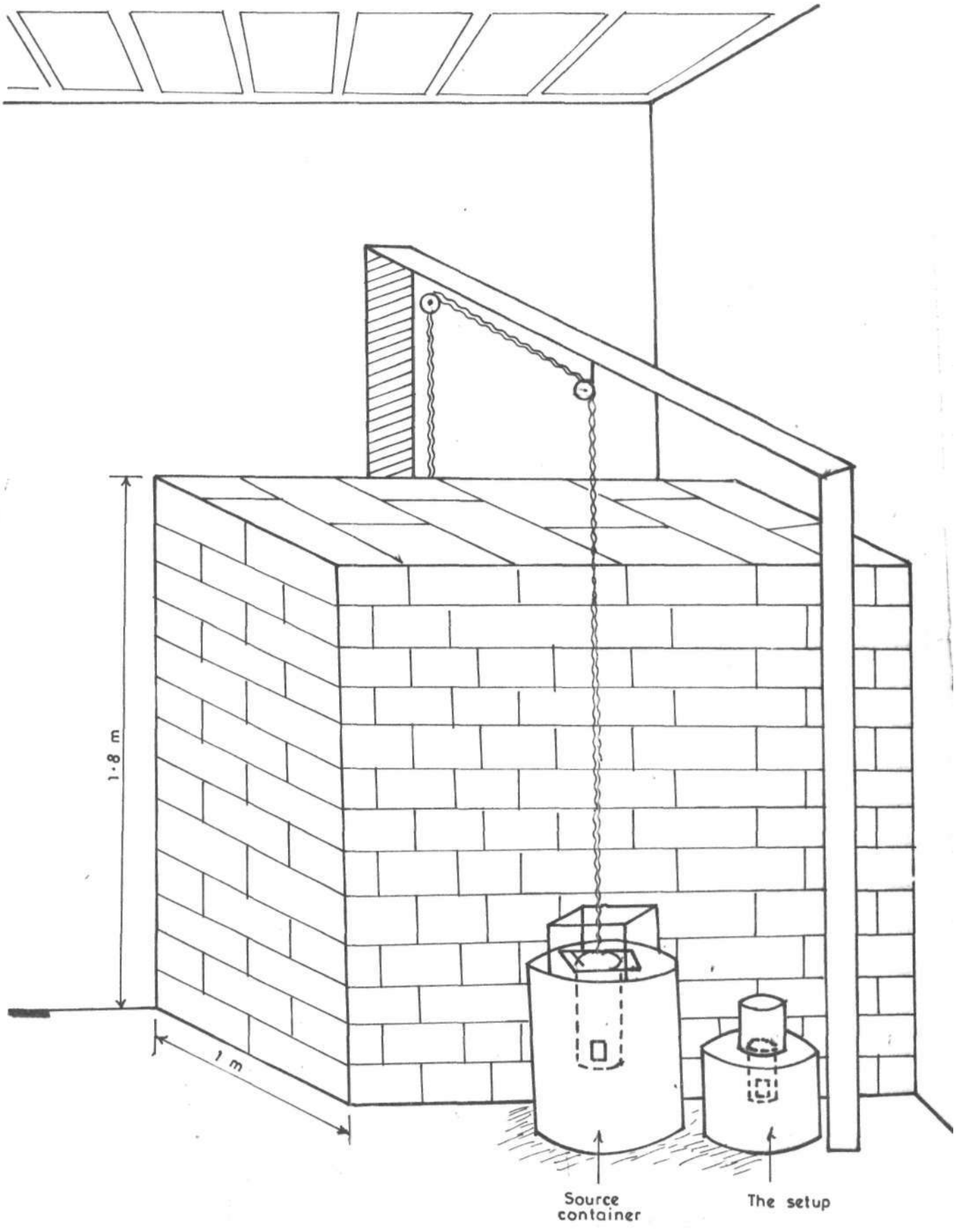


FIG.3.2: THE FACILITY FOR THE DETERMINATION OF H AND O+C/H IN OIL SAMPLES BASED ON THERMAL NEUTRON REFLECTION TECHNIQUE



FIG·3·3: A BLOCK DIAGRAM OF FACILITY AS SEEN IN THE LAB.

CHAPTER FOUR

4.0 DATA MEASUREMENTS AND RESULT

Data measurements were carried out based on the induced activity in the respective indium foil monitors. The radioactivity of the indium foil was measured using a NaI(Tl) detector which has a resolution of 7% for the 662 KeV peak of ^{137}CS. The peak area analysis of the gamma ray spectra was performed by the analyser while the activity was calculated by the IBM computer. The peak reports of interest were then read out. The calculations of specific activity and the resulting neutron reflections parameter (η) of equation 3.3 was then obtained for each of the standards and sample; The η values of the samples is termed the reduced data on their tables.

4.1 EXPERIMENTAL PROCEDURES

In the set up displayed in Fig. 3.2, the beam of fast neutrons, thermalised and reflected by the sample is related to the hydrogen content of the samples as expressed in equation 3.3.

The sample is exposed to fast neutrons for one hour so as to induce appreciable activity in the indium foil and allowing for a cooling time of 20 minutes for the activities of nuclides with short half-lives to decay.

The radioactivity measurement of the indium foil is then carried out for 30 minutes, for the 3 peaks with high branching ratios. By making use of the timing procedures

mentioned above for each of the experiments, the effect of total background activity induced was measured by placing an empty sample container with and without a cadmium sheet on the source. The results obtained are displayed in Table 4.1.

Procedurally, the next experiment was carried out to determine the optimal volume of sample corresponding to the sample height at which the neutron reflection parameter reaches saturation. For this investigation water, ethanol and sand were used to establish the saturation curve. The measured data are shown in Table 4.2.

Hydrocarbons in liquid form and carbon black powder containing well known hydrogen and carbon contents were used as standards to determine the calibration line. In order to ascertain the purity of the carbon powder, the carbon content was determined by the "ashing" method. The powder was ashed at a temperature of 1200^0C and the carbon content found to be 93%. The measured η value of the standards using $600cm^3$ sample are presented in Table 4.3.

The samples of interest in this work are the edible and inedible oils. The non-edible oils include fresh and used lubricants of different grades as well as petrol, diesel, kerosene and pressure fuel. Evidently the quality of a lubricant at different points of use was tested in order to ascertain the ability of the physical method as a quality control technique. Results of the measured data are displayed in Table 4.4 for the variety of edible oil. Similarly the result of the measured data for non-edible oil is shown in Table 4.5

TABLE 4.1 MEASURED BACKGROUND DATA WITH AND WITHOUT Cd SHIELD

SAMPLE	Specific Activity	PEAK READINGS		
		417KeV	1097KeV	1293KeV
Empty samples container without Cd sheet	AS_c	36.05	35.61	45.31
Empty container with Cd sheet	AS_0	6.63	5.8	7.74

TABLE 4.2 MEASURED DATA OF EXCESS COUNT RATE AS A FUNCTION OF VOLUME/HEIGHT FOR DIFFERENT MATERIALS

Volume/Height of sample	SAND ^{n} value ρ = 1.28 g/cm^3	WATER η value ρ= 1.000 g/cm^3	ETHANOL η value ρ= 0.7897g/cm^3
157cm^3 = 2xm	0.75	2.42	3.16
314cm^3 = 4cm	0.77	3.52	3.96
417cm^3 = 6cm	0.79	3.86	4.76
628cm^3 = 8cm	0.98	4.76	4.99
785cm^3 = 10cm	0.85	4.66	4.81
942.5cm^3= 12cm	0.94	4.56	4.80
1099.6c^3= 14cm	0.91	4.53	4.82

TABLE 4.3 MEASURED DATA FOR ESTABLISHING THE CALIBRATION LINE

STANDARD SAMPLES (HYDROCARBONS)

Sample Formula	Density ρ_i g/cm^3	H w%	(O+C)/H w%	Reduced Data η values	
				417 KeV	1293KeV
Toluene C_7H_8	0.8650	8.696	10.5	3.7	4.11
Benzene C_6H_6	0.8790	7.692	12.0	3.62	3.74
Acetone C_3H_6O	0.7920	10.344	8.67	4.2	4.33
n-Butanol $C_4H_{10}O$	0.7890	13.514	6.4	4.98	5.11
Ethanol C_2H_6O	0.7897	13.043	6.67	4.72	4.99
Water H_2O	1.000	11.11	8.0	4.48	4.60
Heptane C_7H_{16}	0.6840	16.00	5.25	5.58	5.7
C + O w% H_2O	0.3716	0.00	-	1.67	1.86
C + 5 w% H_2O	0.4273	0.55	178	1.75	2.18
C + 10 w% H_2O	0.4276	1.100	89	1.89	2.26
C + 15 w% H_2O	0.834	1.660	58	2.11	2.34
C + 20 w% H_2O	0.4950	2.220	43	2.29	2.48

TABLE 4.4 MEASURED DATA FOR DETERMINATION OF H CONTENT AND 0+C/HATOMIC RATIO IN EDIBLE OILS

Samples	Density ρ_i g/cm^3	Reduced Data η		Calculated H w%		Value (0+C)/H W%	
		417 KeV	1293 KeV	417 KeV	1293 KeV	417 KeV	1293 KeV
Cotton seed oil	0.86414	4.00	4.2	9.56	9.55	9.12	9.40
Ground-nut oil	0.85646	3.90	4.121	9.15	9.21	9.55	9.87
Palm oil	0.85003	4.04	4.25	9.73	9.77	9.03	9.23
Corn oil	0.86655	3.86	4.10	8.97	9.12	9.74	9.97
Vegetable oil	0.86974	3.80	3.964	8.74	8.54	10.03	10.83

TABLE 4.5 MEASURED DATA FOR DETERMINATION OF H AND (O+C)/H

CONTENTS OF LUBRICANTS USED AND UN-USED

Samples	Density ρg/cm³	Reduced Data η		Calculated Values			
				H w %		(O+C)/H w %	
		417 KeV	1293 KeV	417 KeV	1293 KeV	417 KeV	1293 KeV
Petrol	0.74436	4.75	4.76	12.65	11.95	6.77	7.34
Kerosene	0.79350	4.79	4.87	12.81	12.42	6.68	7.05
Diesel	0.84005	4.86	4.92	13.10	12.64	6.51	6.91
Low Pressure Fuel	0.90666	4.86	4.98	13.18	12.89	6.46	6.74
Lub. 1:	0.8348	4.986	5.43	13.61	14.82	6.26	5.74
Lub. 2:	0.8665	4.93	5.142	13.48	13.59	6.38	6.36
Lub. 3:	0.84156	4.90	5.12	13.26	13.49	6.42	6.40
Lub. 4:	0.85537	4.80	4.93	12.85	12.68	6.64	6.88
Lub. 5:	0.8638	4.40	4.54	11.21	11.01	7.71	8.17
Spent Lub 2. 800km (505)	0.8980	4.55	4.57	11.82	11.14	7.26	7.98
Spent Lub. 2 700km (505)	0.89648	4.63	4.84	12.15	12.29	7.06	7.13
Spent Lub. 2 700km (504,52)	0.90186	4.67	4.86	12.32	12.38	6.96	7.08
Spent Lub. 2 5,500km (504,53)	0.89544	4.76	4.90	12.69	12.55	6.73	6.98
Spent Lub. 4 Unspecified (Honda Accord)	0.90122	4.51	4.62	11.66	11.35	7.37	7.67
Spent Lub. 2 Unspecified (504,53)	0.89327	4.58	4.69	11.95	11.65	7.26	7.56

4.2 DATA ANALYSIS AND RESULTS

The measured data were based on the three most prominent gamma energy peaks in the decay of ^{116m}In radionuclide. A typical example of the energy peaks obtained from the decay of ^{116m}In radionuclide based on the measuring system is shown in Fig. 4.1

Results of the experiment displayed in Table 4.2 shows that 1mm thick cadmium sheet drastically cuts off the background activity from the paraffin moderator thereby ensuring that only the thermal neutrons from the sample are reaching the detector foil.

A plot of excess count rate (ψ) versus the sample height Fig. 4.2 shows that irrespective of the sample matrix, a moderating thickness of 8-10cm is sufficient to achieve saturation of the thermal neutron flux. This gives the optimum volume of the sample to be used as 600cm^3. Furthermore, results of the measured data for the calibration lines were plotted and are shown in Figs. 4.3 and 4.4 for hydrogen determination and(O+C)H ratio respectively. In Fig. 4.3, the calibration for the determination of total hydrogen content was fitted by the expression given below.

$$\psi = \psi_0 + m\,H\ (w\ \%) \qquad \ldots\ldots\ldots\ldots 4.1$$

where ψ is the excess count rate measured

ψ_0 = 1.96957 representing the matrix effect

m = 0.23351 is the slope of the calibration line.

and H(w %) is hydrogen content to be obtained.

Figure 4.1 – The r-spectrum of irradiated samples

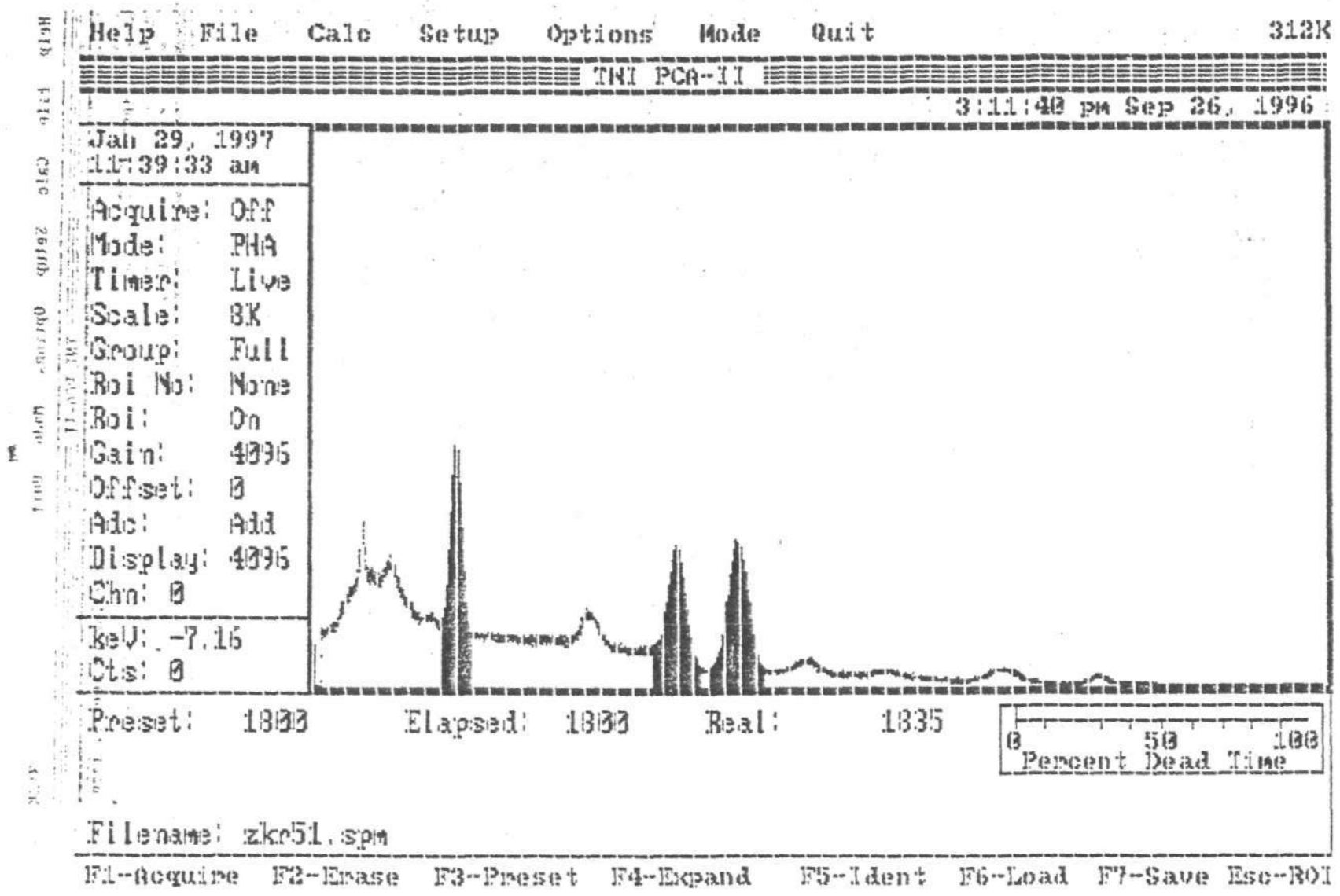


Figure 4.2 - A plot of excess count rate () versus sample height for saturation curve

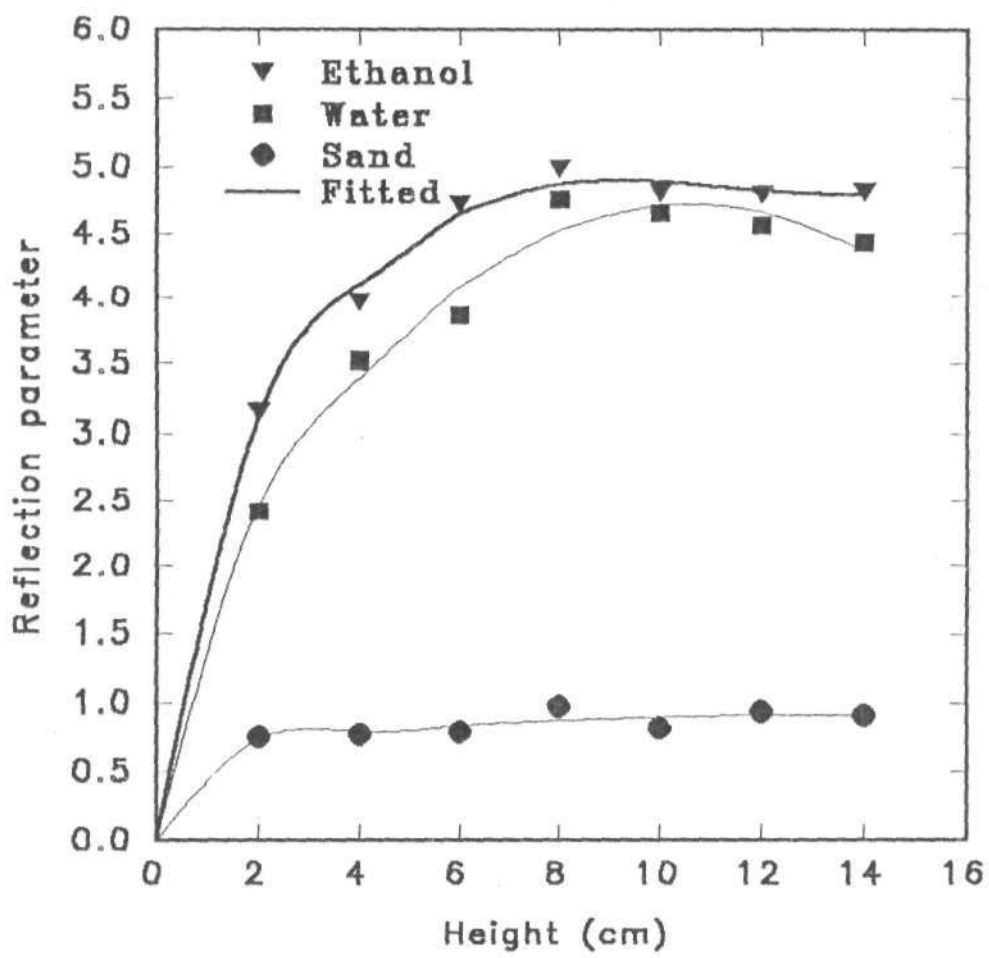


Figure 4.3 - The calibration line for H content

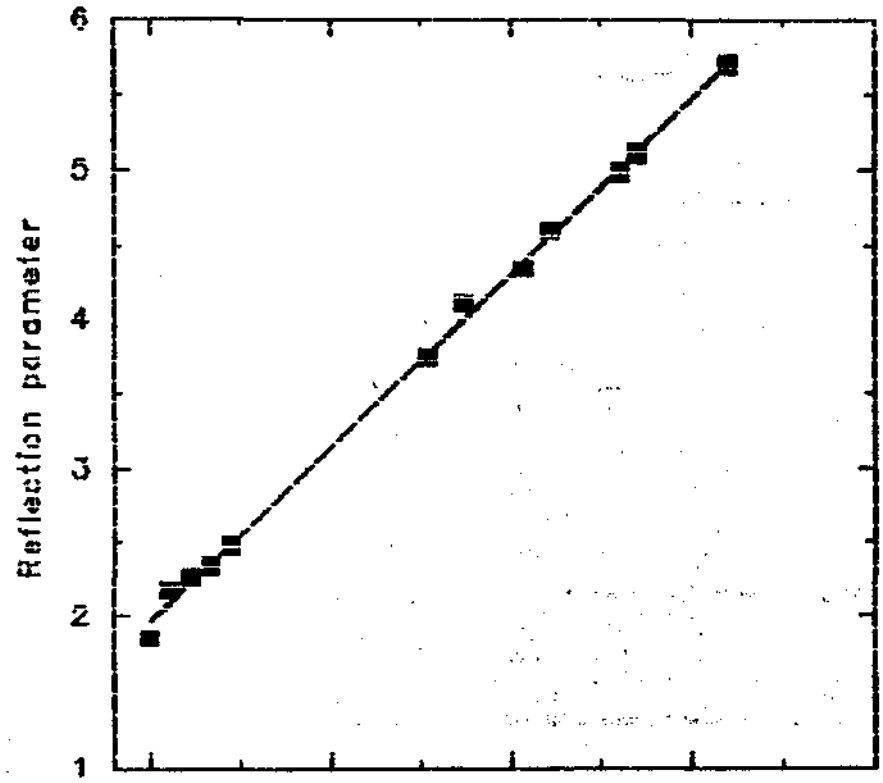
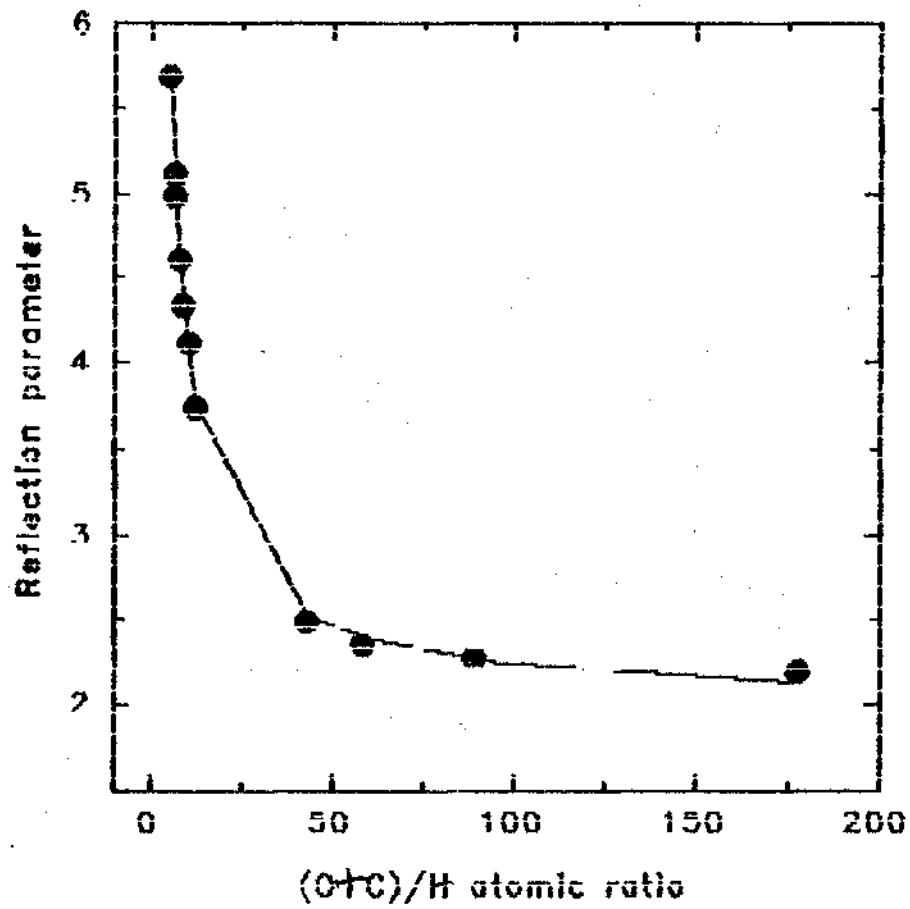


Figure 4.4 – The calibration curve for (O+C)/H atomic Ratio



Similarly, the calibration line for the determination of (O+C)/H atomic ratio shown in Fig. 4.4 was fitted by the equation below:

$$\eta = a + bx^{-1} + Cx^{-2} \quad \ldots\ldots\ldots\ldots\ldots 4.2$$

where η is the measured excess count rate

$a = 1.99817$

$b = 22.78523$

$c = -17.89546$

are the fitting parameters and x represent the (O+C)/H atomic ratio.

The fitting parameters for the calibration lines were obtained for the 417KeV and 1293KeV lines, in order to check the reliability of the results. As can be seen in Tables 4.4 and 4.5 results obtained using the two peaks are in good agreement.

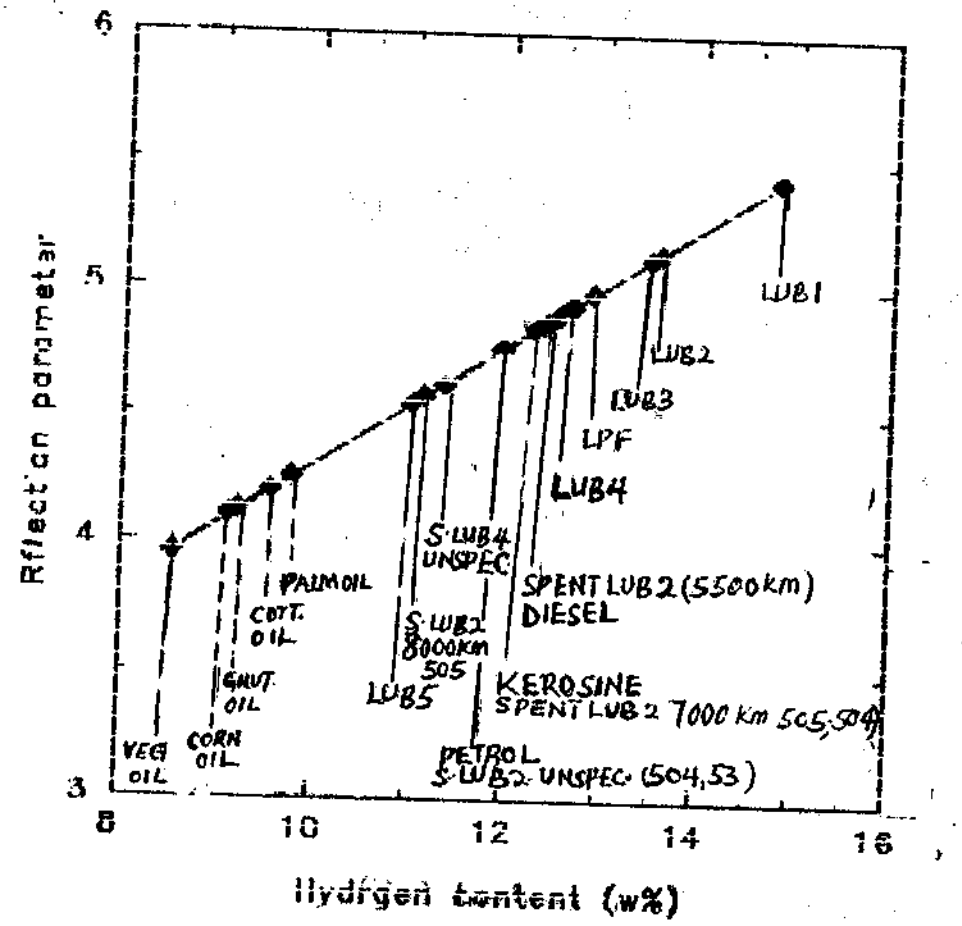
Results of the total hydrogen content in both edible and inedible oils are displayed in Fig. 4.5. The values of total hydrogen content in edible oil are lower than in non-edible oils, while the values of the (O+C)/H are higher in edible oil than in non edible oil samples.

Similarly the behaviour of petroleum products with respect to their hydrogen content at different stages of the cracking of crude oil is highlighted in Table 4.5. The hydrogen content increases as one moves down from fuel oils to lubricants, while the (O+C)/H atomic ratio decreases.

The use of total hydrogen content as a quality control measure for lubricants was tested via the results of measured hydrogen content in used lubricants. As displayed in Table

4.5, it can be seen that the hydrogen content in lubricants decreases with usage and with the age of vehicles.

Figure 4.5 - Display of results for edible and inedible oils



CHAPTER FIVE

5.0 DISCUSSION, CONCLUSION AND RECOMMENDATION

It is evident from the analysis that hydrogen and (O+C)/H atomic ratio in oils can serve as a measure of quality in bulk samples. Ultimately quality is judged by the discriminating consumer who assesses the performance against cost, as such discussing quality will mean considering certain basic factors that constitute the sample in relation to the analytical technique and measurements. The criteria for quality of edible and non edible oil is therefore briefly discussed, the oil samples therefore were graded using the thermal neutron reflection technique.

5.1 DISCUSSION AND CONCLUSION

The available literature reveals that the quality of fats and edible oils is determined by three factors namely:-

(i) Free fatty content

(ii) Moisture content

(iii) Dirt content

Based on the refining methods of these three factors and some added additives, the interest of the consumer in terms of shelf-life, flavour, taste and to some extent added vitamin values, cost and colour can be satisfied. The lower the percentage of these factors the better the quality [Alfred and Patric, (1985), Joseph and Roger (1976)]. These factors are hydrogen dependent. As such, it can be said of the edible oils that the quality is inversely related to

their hydrogen content. Consequently, the measured neutron intensity or the calculated ψ-values is inversely related to the quality of the samples.

Results of the analysis based on the experimental technique confirms that the vegetable oils is of the best quality in agreement with its taste, flavour, colour and cost. Similarly, results of the analysis also reveals palm oil as the least in quality in terms of its hydrogen content. It contains much of the saturated fatty acid content which is characterized by longer carbon chain and large percentage of water soluble fatty acids and therefore easily solidifies, and has low shelf-life.

Furthermore, available literature on lubricants reveals that the number of additives compatible with each other and with the base oil of the lubricant, to serve as corrosion inhibitors, anti-wear agents, friction modifiers and provide the dispersancy required to prevent low temperature sludge, determines the quality and cost of the lubricant [Berkley, (1973); Ranney, (1978)]. These additives are mostly polyamine reaction products, vinyl fatty acid ester copolymers, hydrogenated naphthalene, phenol esters of trimer and dimer acids, etc, which are all characterized with long chain hydrocarbons. Hence the larger the additives the better the quality and the higher the resulting hydrogen content. Therefore, since quality of inedible oil or lubricants is directly related to the hydrogen contents of the samples, implies the intensity of the thermal neutrons measured or the calculated values of the ψ value is directly

related to the quality of the lubricants or fuels.

The analytical physical method findings reveals that lubricant 1 which is the most expensive of the lubricants tested has the highest hydrogen content as such graded as best quality, while lubricant 5 is graded as poor and evident by the fact that its hydrogen content is lower than that of fuel samples as such not a reliable lubricant.

Similarly petrol was found to contain the lowest hydrogen content as a fuel sample and the order in relation to hydrogen content confirms the well known cracking order of crude-oil.

In addition, the investigation performed on used lubricant shows that the hydrogen content in used lubricants tends towards that of fuel oils with usage. Consequently, this investigation reveals that spent lubricants can be used as fuel oil in low combustion engines or burners where the heat of combustion required is relatively low.

5.2 RECOMMENDATION

Further investigations and sophisticated equipment are required to improve upon the facility so as to make it a permanent set up. The set up can be recommended for use as an on-line analyser of bulk materials in industries and ministry of solid minerals. This work is therefore a fore runner in the analysis of hydrogen/moisture content in bulk samples using the non-destructive nuclear technique hitherto not in use in Nigeria.

The adaptation of the neutron reflection method for

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