



DETERMINATION OF THE HYDROGEN CONTENT OF OIL SAMPLES FROM NIGERIA USING AN Am-Be NEUTRON SOURCE.

1

S.A. JONAH^{1*}, I.I. ZAKARI² and S.B. ELEGBA¹

1 Centre for Energy Research and Training, Ahmadu Bello University, P.M.B. 1014, Zaria, Nigeria. E-mail: jonah@abu.edu.ng, fax: 234 62 242227

2 Ahmadu Bello University, Zaria, Nigeria.

* Chief Scientific Investigator

ABSTRACT

A 5 Ci Am-Be neutron source-based facility, which utilises the principles of thermal neutron reflection technique in combination with foil activation method, has been used to determine the total hydrogen content of commercial oil samples from Nigeria. With an established detection limit of 0.25 H w% for oil matrix of volume 600-ml, the total hydrogen contents of the samples were found to be in the range of 11.11-14.22 H w%. The facility is economical and suitable for the determination of moisture in solid samples. A brief description of the on-going projects and future plans concerning the CRP are enumerated.

INTRODUCTION

A knowledge of total hydrogen content of bulk materials is necessary in several areas of science and technology. In Nigeria, there is a growing interest in the mining industries where knowledge of the hydrogen/moisture contents of minerals is needed for plant quality control. Presently the industrial method of assaying hydrogen in different materials is destructive and entails long hours of analysis. Since in most cases, hydrogen needs to be measured in bulk media, the use of a nuclear analytical technique based on moderation of fast neutrons has been recommended [1]. The thermal neutron reflection technique which was developed by Buczko *et al.*, [2] and used by a number of investigators [3], [4], [5], [6] and [7] was adapted for this work.

A set-up based on a 5 Ci Am-Be neutron source which utilises activation foil as the neutron probe was developed for the determination of total hydrogen content in commercial oil samples from Nigeria.

EXPERIMENTAL

The experimental set-up shown in Figure 1 consists of a 5 Ci Am-Be neutron source embedded in a cylindrical paraffin block of diameter 30 cm and of height 30 cm. The extended paraffin block serves as a moderator and provides personnel shielding against radiation hazards. In order to increase the signal-to-background response, a 1-mm thick

¹ Part of this work is to be published in Appl. Rad. & Isotopes

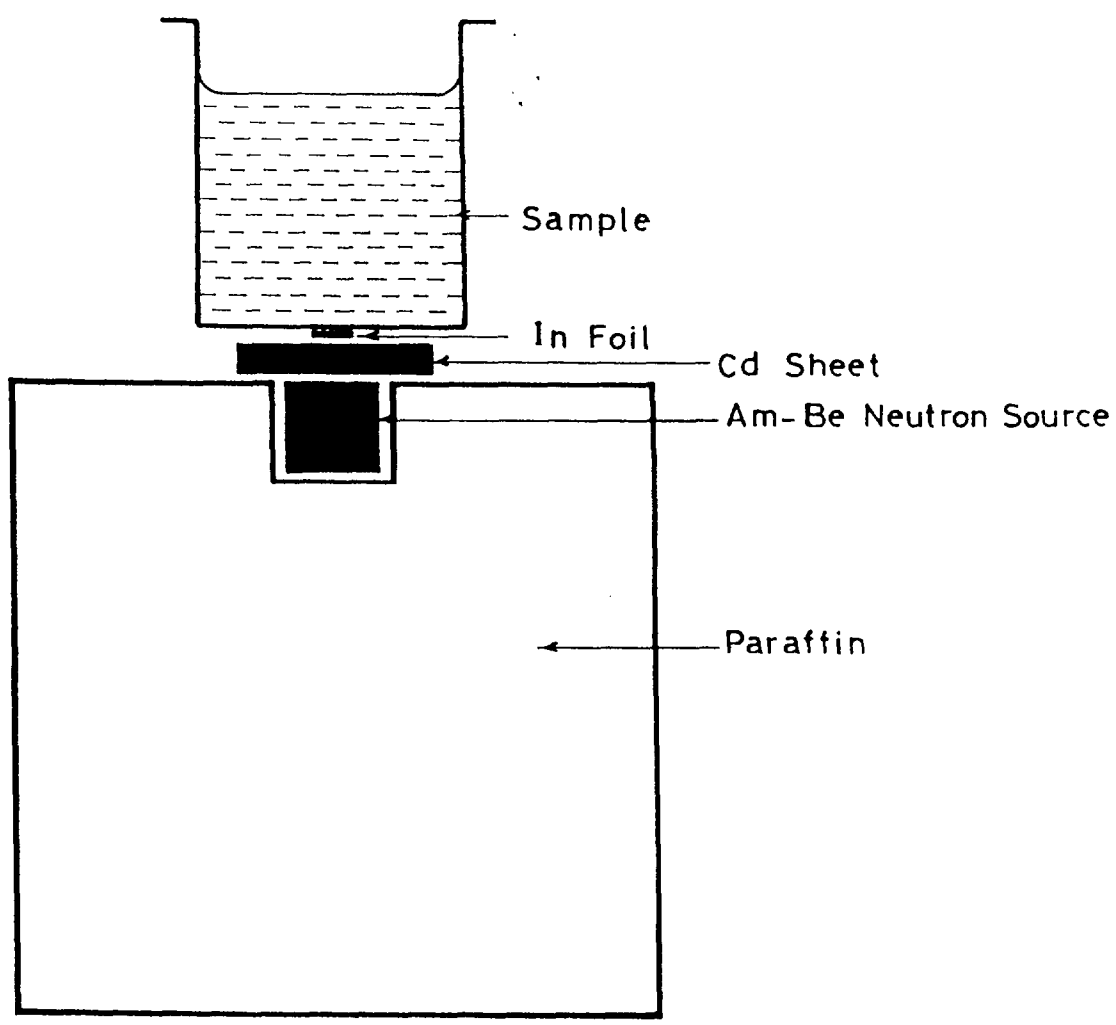


FIG. 1 The experimental setup used for the determination of H content of the oil samples

arrangement the thermal neutron probe and the 5×5 cm cadmium sheet already fastened to the bottom of the container are placed directly on the source in a reproducible geometry. Relevant nuclear data for the high-purity indium foil of dimensions 15×10×1-mm, are given in Table 1.

TABLE 1. Nuclear data of the thermal neutron flux detector.

Reaction	Half-life (min)	Cross section (b)	Gamma Energy (keV)	Gamma Intensity (%)
$^{115}\text{In}(n,\gamma)^{116m}\text{In}$	54.12	16.20	416.90	29.20
			1097.30	56.20
			1293.50	84.40

For a given mass of indium foil, the relative excess specific activity induced in the detector foil in the presence of a sample is related to the neutron reflection parameter (η) as shown below:

$$\eta = (A - A_0)/\rho \cdot A_0 \quad (1)$$

Where, ρ is the physical density of the sample, while A and A_0 are the specific activities of thermal neutrons induced in the detector foil with and without the sample respectively. The sample is exposed to fast neutrons from the Am-Be source for one hour and the resulting intensity of thermal neutrons from the samples is monitored by an indium detector foil via the $^{115}\text{In}(n,\gamma)^{116m}\text{In}$ reaction.

Radioactivity measurements of the detector foils are carried out for 30 minutes by a gamma-ray spectrometric system after a cooling time of 20 minutes. The radioactivity measuring system consists of a 7.6×7.6 cm NaI(Tl) crystal scintillator detector coupled to a computer-based multi-channel analyser.

Hydrocarbons in liquid form containing well-known hydrogen and carbon contents were used as standards to determine the calibration line shown in Figure 2. The measured (η) values for the standards are presented in Table 2.

TABLE 2. Data of standard samples used to construct the calibration line.

Standard	ρ (g/cm ³)	η	H (w%)
C ₆ H ₆ BENZENE	0.879	3.74 ± 0.02	7.692
C ₇ H ₈ TOLUENE	0.866	4.11 ± 0.02	8.696
(CH ₃) ₂ CO ACETONE	0.792	4.33 ± 0.02	10.344
H ₂ O WATER	1.000	4.60 ± 0.02	11.110
C ₂ H ₅ OH ETHANOL	0.790	4.99 ± 0.03	13.043
C ₄ H ₁₀ O BUTYL- ALCOHOL	0.789	5.11 ± 0.03	13.514
C ₇ H ₁₆ n-HEPTANE	0.684	5.70 ± 0.03	16.000

RESULTS AND DISCUSSION

The solid line in Figure 2 for the determination of total hydrogen content was derived by fitting equation 2 below.

$$\eta = \eta_0 + m \cdot H \text{ (w\%)} \quad (2)$$

Where, $\eta_0 = 2.030 \pm 0.092$ represents the effect of the matrix and

$m = 28 \pm 0.012$ is the slope of the calibration line for hydrogen determination.

The total hydrogen content of the samples displayed in Table 3 were deduced using equation 2. These are the average values of the data obtained using the peak analysis reports of the 416.9, 1097.3 and 1293.5 keV gamma lines respectively. Results show that the hydrogen content increases as one moves from fuel oil to lubricants and also as one moves from spent lubricants to fresh lubricant. The hydrogen content of spent lubricants appears to decrease with usage as can be seen in the results indicating the possibility of using this set-up as a quality control facility.

Further measurements currently in progress are designed to standardise the set-up for industrial applications most especially for the determination of moisture content in solid minerals and agricultural products.

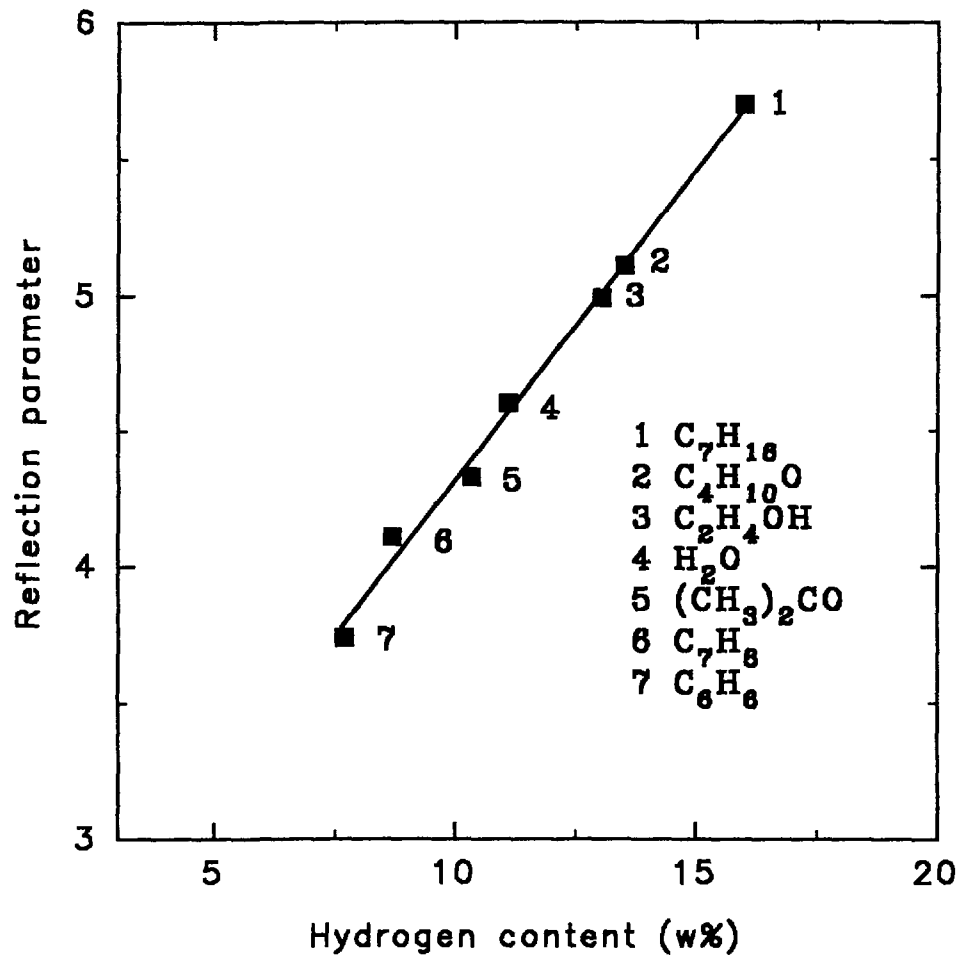


FIG. 2 Calibration line for the determination of total H content in oil samples

TABLE 3. Deduced hydrogen w% in the samples.

Sample	ρ (g/cm ³)	H (w%)
Petrol	0.744	12.30±0.15
Kerosine	0.794	12.26±0.15
Lubricant 1	0.835	14.22±0.17
Lubricant 2	0.867	13.54±0.16
Lubricant 3	0.842	13.38±0.16
Lubricant 4	0.855	12.78±0.15
Lubricant 5	0.864	11.11±0.13
Spent lubricant 2 (5000 km)	0.897	12.62±0.15
Spent lubricant 2 (8000 km)	0.898	11.48±0.14

ON-GOING PROJECTS AND FUTURE PLANS

1. Modification of the set-up shown in FIG. 1 by replacing the indium foil with a Bf_3 counter so as to increase the signal-to-background ratio and decrease the time for analysis.
2. Experimental studies in respect of a prompt-gamma neutron activation analysis (PGNAA) has started with the determination of the fast and thermal neutron flux density distributions inside a 30-cm square panel holder containing powdered coal samples.
3. Acquisition of the MNCP code to improve the PGNAA experimental set-up
4. Testing of the PGNAA facility using coal samples from Nigeria and Niger Republic.
5. Evaluation of the PGNAA facility using results obtained by the XRF technique
6. Development of an in-situ equipment for plant processing and control.

REFERENCES

- [1] S. Semel and S. Helf, *Int. J. Appld. Radiat. & Isot.*, **20**, 229, (1969)
- [2] M. Buczko, Z. Dezso and J. Csikai, *J. Radioanal. Chem.*, **25**, 179, (1975)
- [3] S. M. Al-Jobori, S. Szegedi and Cs. M. Buczko, *Radiochem. Radioanal. Letts.*, **33**, 133, (1978)
- [4] S. Szegedi and Z. Dezso *Radiochem, Radioanal. Lett.* **52**, 343, (1982)
- [5] J. Csikai, *Handbook on Fast Neutron Generators*, CRC Press Inc., Boca Raton, Florida, (1987)
- [6] M. Yousif Ali, A .M. El-Megrab, S. A. Jonah, Daw May Su, M. Varadi and J. Csikai, *Nuclear Geophysics*, **9**, 203, (1995)
- [7] S. A. Jonah, A. M. El-Megrab, M. Varadi and J. Csikai, *J. Radioanal. & Nucl. Chem.*, **218**, 193 (1997)