Asphalt and Paraffin Scale Deposit Measurement by Neutron Back Diffusion Using ^{252}Cf and ^{241}Am-Be Sources

Samir Abdul-Majid & Waleed AbulFaraj
Faculty of Engineering, King Abdulaziz University
P.O. Box 80204, Jeddah 21589
Saudi Arabia

Abstract

Hydrocarbon deposits, often found in oil or polymer industries, are very difficult to detect by the usual inspection techniques of gamma radiography or x-ray radiography methods. The density and atomic number of pipe walls are much higher than that of the scale, therefore, most of the attenuation of gamma rays or x-rays, takes place in the iron, and only a negligible fraction in the scale. This makes the contrast in the radiography film very poor. Moreover accessibility is needed on both sides of the pipe or reservoir. The ultrasonic method cannot function on hot or unprepared surfaces.

In this method neutrons from ^{241}Am-Be or ^{252}Cf sources are allowed to interact with carbon steel pipes containing either asphalt or paraffin scale. Fast neutrons penetrate the pipe wall without significant interaction; they are scattered elastically with H and C atoms in the scale and hereby slow-down. Some of the slowed down neutrons diffuse backward and are measured by a BF_3 slow neutron detector.

Counts taken with thickness in the BF_3 counter for asphalt scale were linear for both the neutron sources. When using paraffin, counts were linear with ^{241}Am-Be neutrons but approached saturation with ^{252}Cf neutrons. This is attributed to the fact that ^{241}Am-Be neutrons has harder spectrum compared to ^{252}Cf neutrons, and can penetrate into deeper layers. With weaker spectrum slowed down back diffused neutrons are absorbed or scattered in the scale before reaching the detector.

Because sensitivity depends on reduction of background counts, several attempts were made to reduce these counts by using cadmium, boron or boric acid shield around the detector. It was found that 2.5 cm of boron powder around the detector reduced background counts significantly. It was possible to measures scale thickness down to a fraction of one mm.

Introduction

Pipe corrosion and scale deposits can cause large economic losses for industrial plants, such as refineries or petrochemical plants. One such loss would be the shutdown of a plant due to pipe blockage by scale accumulation and the resulting reduction in flow. Another is the shutdown for routine inspection and replacement of certain pipes, although pipes may still have life time, in order to avoid future failure. The early identification of deposits inside pipes or tanks during operation will reduce maintenance cost by minimizing unnecessary pipe replacement and plant shutdown for inspection.

Hydrocarbon deposits, often found in oil refineries or polymer producing plants, are very difficult to defect by existing methods of radiography or ultrasonic techniques on-line, even if flow is stopped. New unconventional methods for detecting organic scale are needed.

In order to understand the reason for failure of gamma or x-ray radiography we define two parameters, A and D. The parameter A shows the amount of attenuation of radiation in steel pipe of 1 cm thickness, given by:

$$A= I/I_0 = \exp(-\mu_{ai})$$

Where I and I_0 are attenuated and incident beams and μ_{ai} is the linear attenuation coefficient of iron. The parameter D provides information on the fraction of absorption of radiation in organic scale of thickness x,. This will show if this value is high enough to give detectable change in photographic film. This parameter is given by:

$$D = (1 - e^{-\mu_{as} x}) e^{-\mu_{ai}}$$

Where μ_{as} is the linear attenuation coefficient of organic scale.

The results of these calculations are shown in Table 1. At 100 keV, most commonly used value in most industrial x-ray applications, the iron wall of the pipe absorb most of the incident energy (92.8%) and only about 7% is left for scale. The fractions of absorption of the incident beam in 1 mm and in 1 cm of scale are 0.14% and 1% respectively. As 1 cm of scale is considered high thickness, it is very clear that the 1% change in attenuation will not be detected by radiographic film.

At 0.4 MeV which is the average gamma ray energy emitted from ^{192}Ir, a common gamma source in industrial radiography, iron absorbs 0.513 of incident beam in 1 cm leaving 0.487 for scale. At scale thicknesses of 1 mm and 1cm the fractions of incident beam absorption in scale are 0.5%, and 5% respectively. Although this kind of attenuation is better than previous case, the contrast will still be poor. At 1.25 MeV, the average energy of ^{60}Co, the fractions of incident beam absorbed in 1 mm and 1 cm of scale are 0.4% and 4% respectively. Again the contrast will be poor.

It can, therefore be concluded that the use of x-ray or gamma ray attenuation at different energies is not going to give detectable contrast, mainly because of the scale low atomic number and low gamma attenuation coefficient.

The ultrasonic method can work only if the outer surface of pipe is well prepared, clean and polished. The method does not work on pipes with insulator. Moreover the pipe wall temperature needs to be low enough in order for medium needed between the pipe and the ultrasonic device not melt down.

In neutron moderation and back diffusion method (Fig. 1), fast neutrons emitted from the source penetrate the iron pipe without significant absorption since iron has a small absorption cross section for fast neutrons. They interact elastically with hydrogen and carbon atoms of the organic scale and are hereby slowed down. Some of the slowed down neutrons will diffuse backward and are detected by the BF_3 neutron detector. This is a slow neutron detector that respond poorly to fast neutrons and strongly to low energy ones. The boron cross section to fast neutron is low (1 b at about 1MeV), and it is high to slow ones (σ_a is about 2000 b at thermal neutrons). Count increases with amount of scale, and the greater the thickness of the scale, the more moderation and backscattering of the slow neutrons will be. Different energy gamma rays are emitted due to the absorption of slow neutrons, but this can be discriminated against by the counting system. In this technique the results of the inspection are readily available and accessibility is needed from only one side of the object to be examined. The method needs no actual contact with the pipe wall and they can be applied successfully on hot, unprepared surfaces and insulated pipes.

Other studies on organic scale deposition and measurements by neutron moderation and back-diffusion were made Abdul-Majid et al., [1-3] who also used other radiation interaction methods for the same purpose [4-7]. Ribeiro et al investigated the obstruction of crude oil carrying pipelines by paraffin deposits. [8]. San-Miguel [9] studied the effect of corrosion inhibitors on wax deposition. Gunarathne and Keatch [10] studied the dissolution of mineral deposits in petroleum pipelines.

On-line inspection of scale and corrosion was studied by Fujine et al, [11] who used video image processing system for real-time neutron radiography. Dalichow et al [12] studied crack depth measurements in stainless steel pipes while Arney et al studied cement-lined pipes for water lubricated transport of heavy oil [13].

Materials and Methods

In this technique three different organic materials have been studied as deposit materials; these are:

1. Asphalt [$C_{89} H_{104}S_3N_20_2)_2$]: Careful observation of actual organic scale was found to have very close properties to asphalt. The ductility test performed on asphalt showed that it was very hard and not elastic. The softening point test was performed by heating an asphalt sample up to 85 ^{0}C.

2. Paraffin ($C_{25}H_{52}$): This is a common oil product material likely to be found as deposit in oil refineries.
3. Polyethylene $(CH_2)_n$: This type of scale is found inside polyethylene chemical reactor following a process known as "sheeting".

All scale materials have density of about 1 g/cm³.

The pipes used in the measurement were made of carbon steel. Aluminum pipes each has 1 mm wall thickness been used to confine the scale inside each iron pipe. Carbon steel consists mainly of iron, with little percentage of manganese, silicon, copper, aluminum, chromium and traces of other elements.

The experimental arrangement is described elsewhere [1]. The detector was a BF_3, gas filled, slow neutron proportional counter (LND Inc., model 202A, USA). The associated electronic components includes a power supply (type 2000) a preamplifier (type 1406) an amplifier (type 2012), and an 8192 multi- channel analyzer (pc with special electronic card); all made by Canberra, USA.

The neutron sources were either ^{241}Am-Be or ^{252}Cf. The activity of the ^{241}Am in the ^{241}Am-Be neutron source was 3 Ci (1.11×10^{11} Bq) with accuracy within ±6%. The neutron emission rate was 6.6×10^{6} n/s with a tolerance of ±10%. The associated gamma exposure rate for the bare source was about 7.5 mR/h, at 1m, while the total neutron dose rate was 6.6 mrem/h at 1m. ^{241}Am has a half-life of 433 years and decays by emitting alpha-particles of 5.4 MeV followed by Gamma rays of 60 keV. Several gamma rays are also emitted following a neutron emission due to de-excitation of the Be nucleus. The ^{252}Cf decays with 2.65 year half-life, with neutron emission rate of about 10^{9} n/s. It is alpha emitter with only 3% spontaneous fission. Several fission and capture gamma rays are emitted following the decay process.

Results

Bare Detector

Data on back diffused neutrons with paraffin scale for 16 cm diameter pipe for 5 minutes is shown in Fig. 2 using ^{241}Am-Be source and in Fig 3 using ^{252}Cf. Clearly ^{252}Cf give much higher counts. On the other hand, counts approach saturation with thickness with ^{252}Cf faster than that of ^{241}Am-Be. Because of the softer spectrum of ^{252}Cf, the self absorption of incident and back diffused neutrons is higher. The sensitivity (counts/mm) decreases with thickness for ^{252}Cf source faster than for ^{241}Am-Be source.

In order to increase neutron flux at the scale, water reflector of about 10 cm thickness was put behind the source. Although neutron reflector increased counts, it also increased background making the use of reflector not so useful. Increase of background would increase the minimum detectable scale thickness.

Reduction of Background

In order to increase sensitivity, background counts (counts without scale) due to interaction with surrounding materials, needed to be reduced. Cadmium is known for its high absorption cross section for slow neutrons. The BF_3 detector was wrapped with Cd and was put at 50 cm from the neutron source. The counts for 2 min as a function of Cd thicknesses around the detector are given in Fig. 4. Counts dropped about 7 folds in the first mm of cadmium. No significant drop in counts was observed with the additional Cd layers. Part of neutrons from the source and surrounding having energy of more than 0.4 eV can still penetrate Cd and make interactions with the BF_3 gas and produce counts.

Boron is another element of high cross section for slow neutrons. The detector was surrounded with boron that was encapsulated within a box made of aluminum of about 1 mm wall thickness. Counts in the BF_3 counter with 0.5 cm of boron at 50 cm between the source and detector dropped from 7250 to 190 for two minutes counting time. Counts in the BF_3 counter as functions of boron thickness are given in Table 2 for ^{241}Am-Be and ^{252}Cf. At 2 cm almost all counts in the detector vanishes, and further thickness bring background counts to disappear. Boric acid was also tried as a shield around the detector but was no so effective and thick layer would be needed to reduce background to the desired value.

One possible design was a vertically positioned BF_3 detector surrounded by 2.5 cm boron with an opening of one cm facing the pipe. Most of the counts will be from neutrons coming from scale side while scattered neutrons from surrounding are absorbed (Fig. 5). Counts with asphalt scale are shown in Fig. 6. It can be observed that background is very small relative to signal from scale. The average slope (sensitivity) of the graph is about 750 founts/cm min.

The minimum detection limit (MDL), assuming 95% confidence level is given by [14, 15].

$$\text{MDL} = 2.71 + 4.653\ \sigma_B$$

where σ_B is the standard deviation of the background counts. Taking zero scale thickness counts as a mean counts for background and assuming Poisson distribution, σ_B is 20.4, and MDL is 97 in 2 min.

The slope of the curve is 75/mm.min. Assuming the error associated with base line subtraction to be about the same, it can be concluded that minimum detection thickness is 1 mm in two min. Counts for 10 minutes will bring the minimum detectable thickness to around 0.3 mm at 95% confidence level. This is much better sensitivity compared to our previous work [1].

Discussion and Conclusion

Neutron moderation and back diffusion method is very fast compared to gamma radiography method that fails to detect organic scale. Radioactive source used are millions of times less in activities. It is therefore much safer.

The work was performed using, mainly, laboratory equipment. For field work it is possible to design a compact, easy to carry portable system. Simpler electronic components are needed because, essentially, there are only one or two peaks that needed evaluation. A system based on single channel analyzer with proper window would be sufficient. A remote system should be designed to transfer neutron sources from its container to the proper location in the set-up. In this case the exposure can be negligible.

The ^{241}Am-Be of 450 years half life has cost advantages over the ^{252}Cf source of about 2.6 years half-life which also has softer spectrum. The sensitivity, the slope of counts – thickness curve remain almost constant with ^{241}Am-Be neutrons for thickness under consideration, while it reduces with thickness with ^{252}Cf neutrons.

The actual organic scale of crude oil is very similar in nature to asphalt or paraffin. In pipes carrying different oil products, scale may differ depending on type of oil passing through the pipe. This depends on location in the plant where measures take place. Scale at a specific location usually has same characteristics. Accordingly calibration of counting system should be made at specific locations. The system was more sensitive to paraffin for oil carrying pipes. In order to have a system that is sensitive to small scale thickness change or can see only small portion of the pipe it is important to reduce background. The use of Cd around the detector had reduced background counts significantly in the first mm. Boron was more effective shield against neutrons coming from other than the source.

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Table 1. Attenuation Parameters for organic scale of thickness x inside 1 cm wall thickness iron pipe at 0.1 MeV, 0.4 MeV and 1.25 MeV.

	0.1 MeV		0.4 MeV		1.25 MeV	
X	A	D%	A	D%	A	D %
0.1	0.98	0.14	0.989	0.53	0.9937	0.4
0.2	0.967	0.237	0.969	1	0.987	0.8
0.3	0.95	0.36	0.969	1.5	0.981	1.2
0.4	0.935	0.468	0.958	2	0.975	1.6
0.5	0.912	0.63	0.948	2.5	0.969	1.6
0.6	0.905	0.68	0.938	3.0	0.963	1.6
0.7	0.889	0.8	0.938	3.5	0.957	2.4
0.8	0. 875	0.9	0.919	3.9	0.951	3.2
0.9	0.86	1	0.909	4.4	0.945	3.6
1.0	0.85	1.1	0.9	4.8	0.939	4

Table 2 : Neutron counts for 2 min in BF_3 counter held vertically surrounded by boron at several boron thicknesses around the detector, the source to detector distance was 50 cm.

Boron Thickness (cm)	**^{252}Cf Source (Counts)**	**^{241}Am-Be Source (Counts)**
0	**10300**	**10300**
2	**82**	**27**
3	**10**	**15**
4	**0**	**14**
5	**0**	**0**

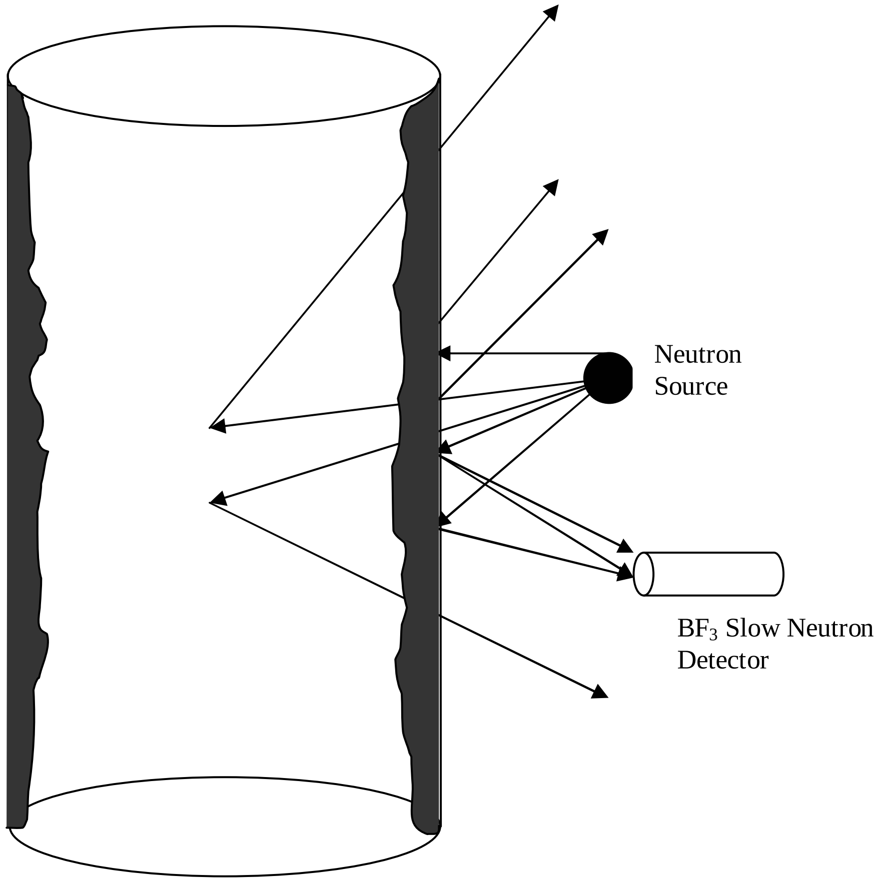

Fig.1 Neutron back scattering and back diffusion

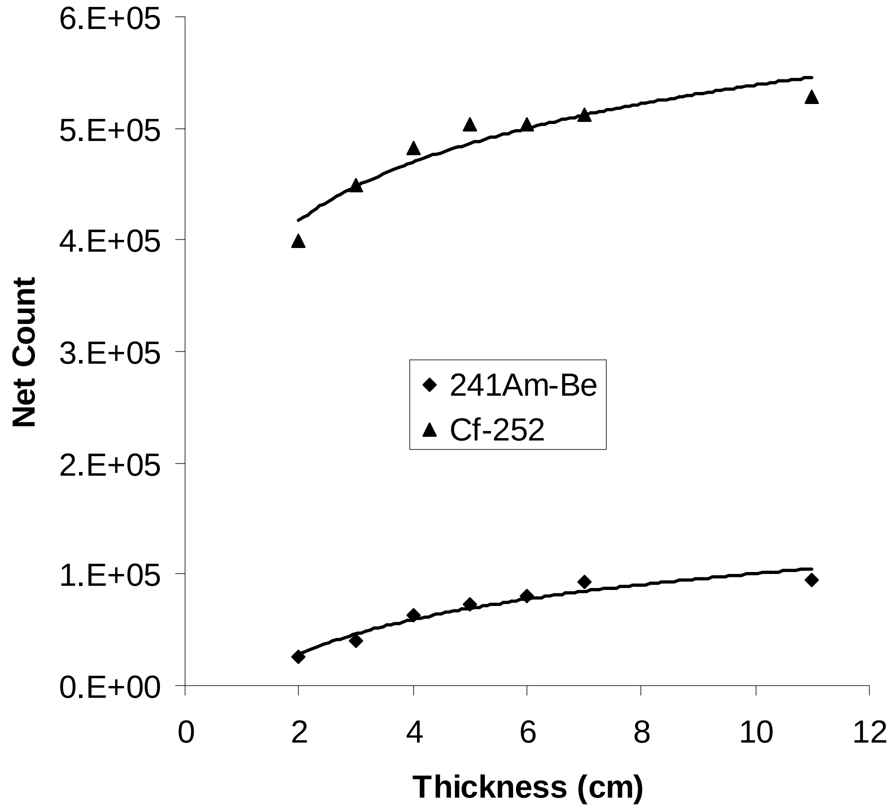

Fig. 2 : Counts for 5 minutes of back diffused neutrons vs paraffin scale thicknesses for ^{241}Am-Be and ^{252}Cf sources; the pipe wall thickness was 4 mm.

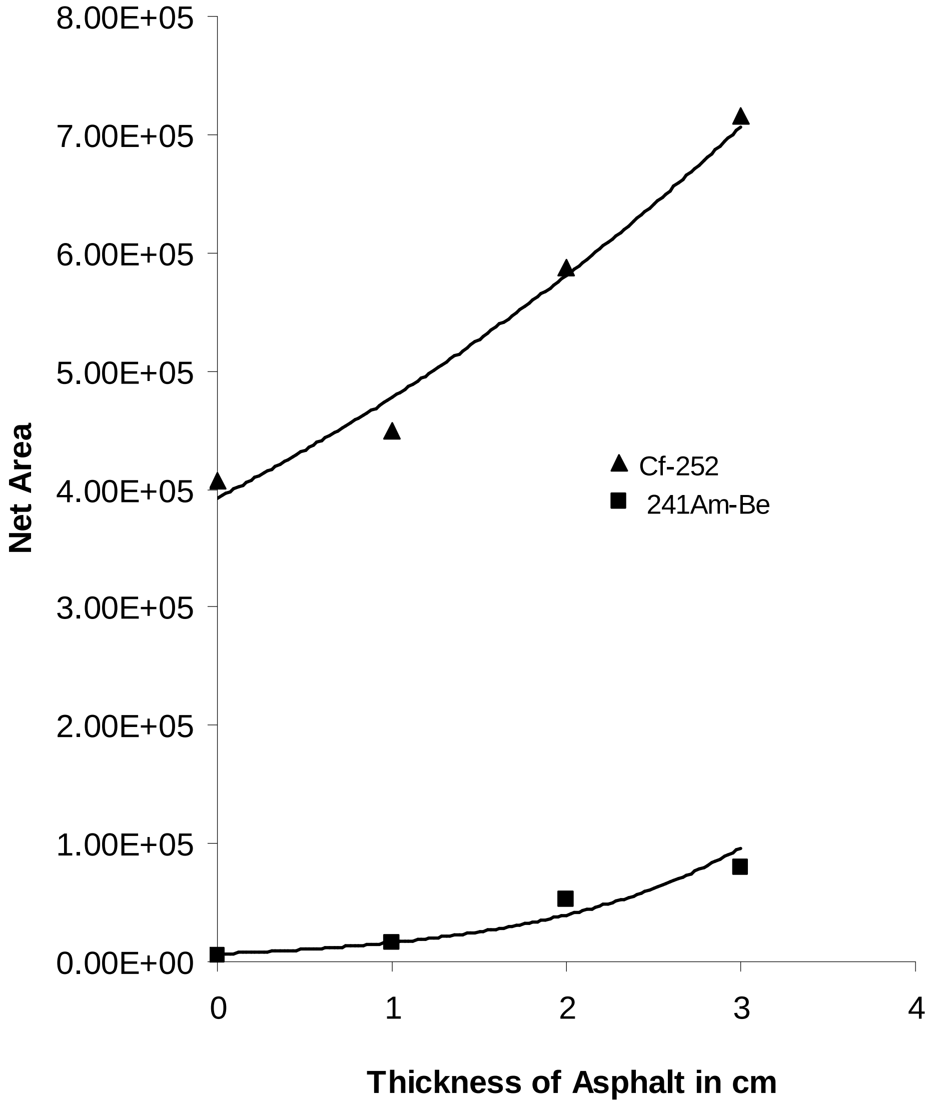

Fig. 3 : Counts for 5 minutes of back diffused neutron vs asphalt scale thickness using ^{241} Am-Be source; the pipe wall thickness was 4 mm.

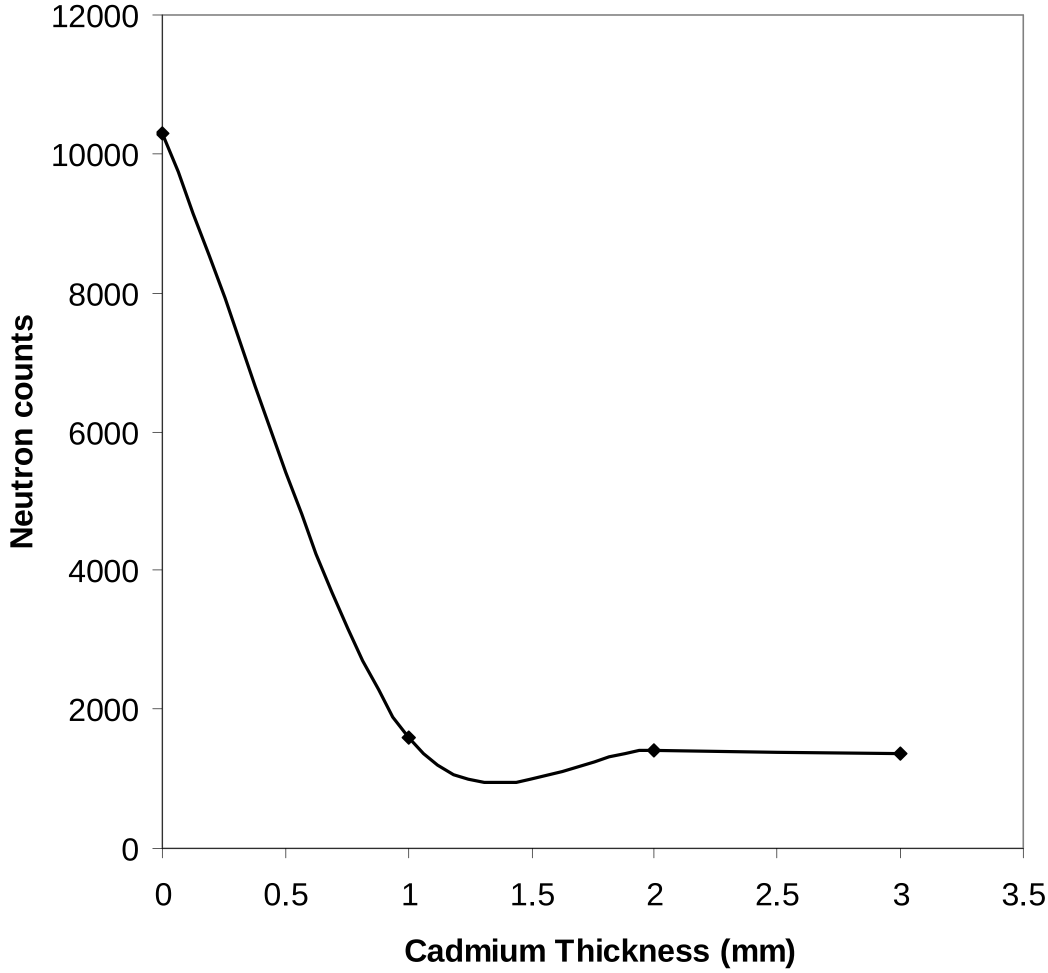

Fig. 4 : Neutron Counts for 2 min from ^{252}Cf source for different thickness of Cadmium around the BF_3 detector

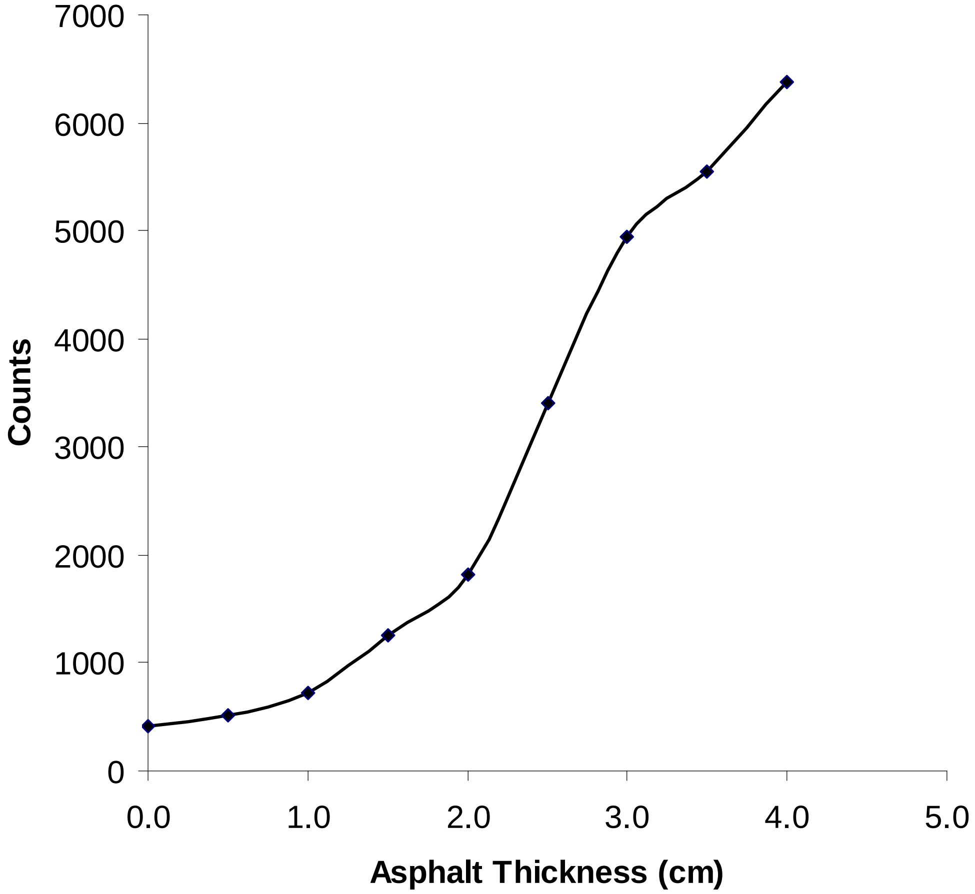

Fig. 6 :Counts for 2 min in vertically positioned BF_3 detector surrounded by 2.5 cm thickness of boron with 1 cm opening at different asphalt scale thicknesses, using ^{252}Cf source.

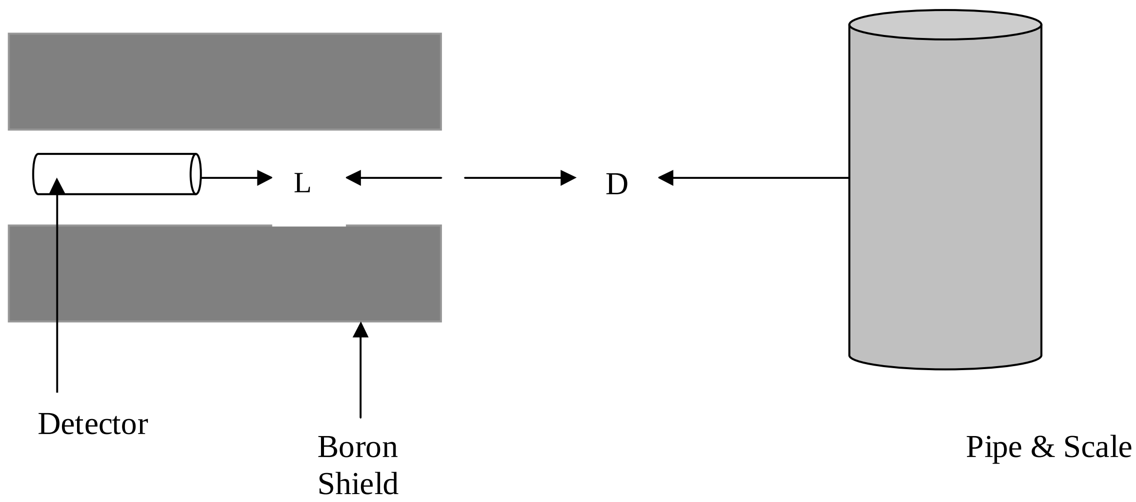

Fig. 7: Geometry for collimation measurements

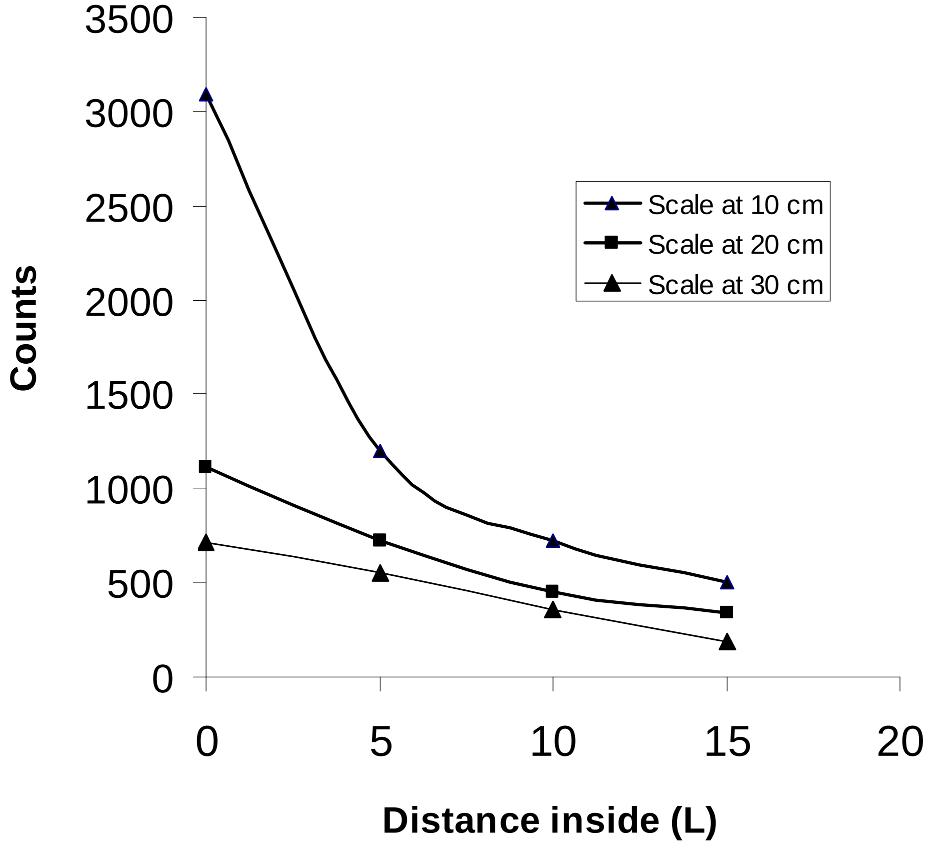

Fig. 8 : Counts in BF_3 Counter at differerent distances L inside boron shield and different paraffin scale distances D of 10, 20 and 30 cm from the detection system.

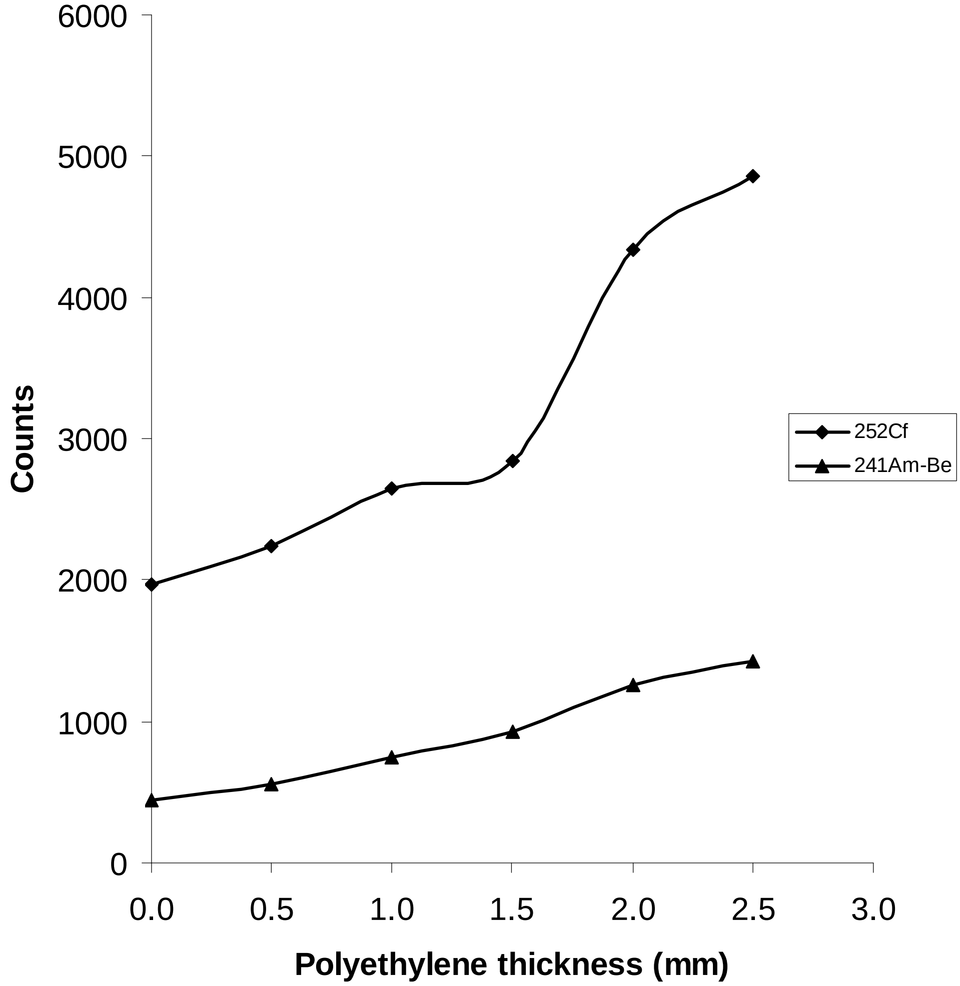

Fig. 9: Neutron counts for 10 min in BF_3 counter held horizontally and surrounded by 2.5 cm boron at 2 cm away from polyethylene scale inside iron pipe, using ^{252}Cf and ^{241}Am-Be sources.