ATTENUATION AND MODERATION OF NEUTRONS AND ATTENUATION OF ELECTRONS IN A FEW TYPES OF POLYMERS FOR MATERIAL TESTING

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الخلاصـة :

لقد تَـمَّ في هذا البحث دراسة توهين شدة النيوترونات الصادرة من المصدر النيوترني (أمر يشيوم - بريليوم - ٢٤١) في بعض أنواع اللدائن ، حيث إنَّ لهذا المصدر استخدامات مختلفة وخاصة في التصوير الإشعاعي والبحث عن المعادن .

لقد أظهر البولي إثيلين قابلية عالية على توهين النيوترونات في وحدة الوزن ، بينما كان توهين كـلٍّ من البولي فنايل كلورايد (PVC) ، والبولي إيثلين متقارباً في وحدة السُّـمْـك . وقد أُستفيد من خاصية التوهين والتهدئة للنيوترونات في اللدائن عند فحص خواصها كالسُّـمْـك والكثافة ووجود الفجوات والشوائب داخل مادتها . من ناحية ثانية تم دراسة تفاعل الالكترونات الصادرة من النظير بيزموث - ٢٠٧ في بعض اللدائن حيث أظهرت الدراسة إمكانية استخدام طريقة الدراسة في فحص خواص الرقائق البلاستيكية .

ومن ميزات طرق الفحص هذه أنها غير متلفة ، وأن نتائج الفحص يمكن الحصول عليها بسرعة .

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ABSTRACT

Polyethylene, PVC, and other polymers' neutron attenuation was investigated using a ^{241}Am–Be source. This source is widely used in mineral exploration, elemental analysis, radiography, and in other applications. Polyethylene showed the highest density thickness attenuation, while both PVC and polyethylene exhibited close values linear thickness attenuation. Neutron moderation in polymers was also studied. The data obtained from both attenuation and moderation of neutrons can be utilized for polymer material property tests, such as thickness, density and existence of voids and impurities. Data from electron interaction with polymers studies, using ^{207}Bi conversion electron source, showed that the process can be utilized for polymer thin film material property tests. These techniques are noncontact, nondestructive tests, and their results are immediately available.

Keywords: Radiation attenuation, material testing, radiation interaction, nondestructive testing.

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INTRODUCTION

Polymers contain mainly H and C atoms, they are, therefore, excellent attenuators, scatterers, and moderators for fast neutrons. Polyethylene $[(CH_2)_n]$ is considered an attractive neutron shield because of its low effective atomic number, chemical stability, and availability. The neutron attenuation properties of polyethylene, Polyvinyl chloride (PVC) $[(CH_2{-}CH{-}Cl)_n]$ and other polymers were investigated using an ^{241}Am–Be source. This source is widely used in mineral exploration, oil well logging, material analysis, neutron radiography, gauging, and other applications.

Radiation attenuation can also be a useful tool for polymer material testing and quality control. The gamma-ray attenuation and scattering properties of a few types of polymers (for material properties testing) were studied by Abdul-Majid and Abulfaraj [1]. While gamma-ray interaction is affected mainly by material density and atomic number, that for neutrons is affected by properties such as absorption, scattering, and moderation and, therefore, can provide additional useful information. For this reason, neutron moderation was investigated as well. Elements with close gamma-ray attenuation properties can have huge differences in their neutron absorption and scattering.

The simplest and most straightforward attenuation study is that of the narrow beam. The arrangement is such that any single interaction would remove a particle from being detected. For neutrons, this includes, in addition to absorption, the processes of scattering away from the detector, which is associated with reduction in energy. In our experiment, the fast neutron dose equivalent was measured. Neutrons between 1 MeV – 10 MeV have a dose equivalent per unit neutron flux that decreases very slowly with energy increase, which is about 30 times that of neutrons of 10 keV or below. Between 10 keV and 1 MeV the dose equivalent per unit flux increases sharply. Therefore, a neutron that falls below 1 MeV is effectively removed.

The total neutron cross-section for H atoms is generally low, and is constant from 1 eV to about 10 keV and thereafter decreases rapidly with energy increase. A fast neutron undergoing a collision with hydrogen loses on the average 50% of its energy. The cross-section for a second collision is much larger because the energy becomes lower and the mean free path becomes smaller. As a consequence, the second collision might occur not far from the site of the first. A fast neutron that is not removed from the beam in its first collision has a bigger chance of being removed in the second. On collision with a C atom, a fast neutron loses on the average 14% of its initial energy in a single collision. The total cross-section is mainly elastic up to more than 5 MeV. It is constant up to about 1 MeV with clear resonances between 2–7 MeV. PVC contains, in addition to H and C atoms, Cl atoms with an intermediate value of neutron microscopic removal cross-section [2].

No study was found in the literature on the attenuation of neutrons from ^{241}Am–Be source. Detailed monoenergetic fast neutron attenuation data in polyethylene are given in NCRP report No. 38 [3]. Attenuation in a shield composed of polyethylene alone or polyethylene with other materials was studied by Greene and Thomas [4] and Maruyama and Bouts [5] for 14 MeV and 15 MeV neutrons, by Gujrathi and D'Auria [6] for 2.6 MeV neutrons, and by Smathers *et al.* [7], for neutrons created by the Be(d,n) reaction. Yin and Yuxiong [8] studied attenuation in polyethylene of neutrons produced in photonuclear reactions, Yamamoto *et al.* [9], that of those from a D–T neutron source, while Sharma and Hood [10] and McCall and Hootman [11] studied ^{252}Cf neutrons. Other polymers studied for neutron attenuation were water-extended-polyester resin (WEP) [12] and Poly:Boron [13]. However, other polymers did not receive the same attention as polyethylene. PVC for example, is a readily available, inexpensive polymer that can be obtained in powder, pellets, or sheet forms. The Cl atom in the polymer has higher atomic number and higher neutron absorption cross-section than H and C atoms. PVC has higher gamma-ray linear attenuation coefficient and higher density than polyethylene; it can, therefore, be more suitable in combined gamma-ray and neutron fields. In a recent work, Abdul-Majid *et al.* [14] studied neutron and gamma-ray attenuation in polyethylene and PVC loaded mortars, as well as the gamma-ray attenuation in the two polymers in sheet form [1]. PVC mortar and pure PVC showed higher gamma-ray attenuation over polyethylene.

The neutron moderation and back diffusion technique was used before, mainly for the measurement of moisture in soil and in other media [15–18]. A good review of the subject was given by Walker [19]. The process is affected by the material scattering and absorption cross-sections as well as by the average energy loss per collision, which will give the number of collisions needed to thermalize neutrons. The moderation process may, therefore, be described in terms of the moderating ratio $MR = \xi \Sigma_s/\Sigma_a$, where ξ is the logarithmic energy decrement and Σ_s and Σ_a are the macroscopic scattering and absorption cross-sections respectively. Polyethylene has higher moderation ratio over PVC. Impurity atoms, existence of voids or change in density would change the effective value of MR and hence the count rate.

Because of the short range of electrons in matter, the data of electron interaction with polymers will be useful for thin film properties studies. It is known that the change in electron energy and intensity per unit distance traveled occurs with change in density as well as with existence of impurities, particularly high atomic number ones. Both electron and neutron interactions are noncontact and nondestructive techniques and the testing results are immediately available. The method may, therefore, be used for on-line quality control of polymers.

MATERIALS AND METHODS

The set-up used for fast neutron attenuation is shown in Figure 1. The beam defined by the solid angle subtended by the detector and the source, was equal to 0.038 sr. The activity of the ^{241}Am–Be source (supplied by Amersham U.K.) was 1.11×10^5 MBq; it emits about 6.6×10^6 n/s with a tolerance of about 10%. The source is cylindrical in shape encapsulated in a stainless steel capsule with overall dimensions of 2.24 cm diameter by 4.85 cm height. The neutron spectrum of ^{241}Am–Be source has clear peaks at about 1 MeV and 3 MeV and the energy goes up to more than 10 MeV [20]. A neutron remmeter (Model NM2 supplied by Nuclear Enterprises U.S.A.) was utilized for dose rate measurements. Readings were taken from the digital display ranging from 0.01 μSv/hr (0.001 mrem/hr) to 0.1 Sv/hr (10 rem/hr). Above 0.01 Sv/hr the error in digital reading increases progressively reaching a maximum of 10% at 0.1 Sv/hr. An aluminum frame was used as a holding structure because of its low cross-section. The detector background counts due to scattering from the surrounding materials was constant and was subtracted from each reading.

The set-up for neutron moderation and back diffusion is shown in Figure 2. It consisted of the same neutron source, and a BF_3 gas filled proportional counter (type 202A LND Inc. U.S.A.) which responds mainly to slow neutrons. A cadmium sheet was inserted between the detector and the source to absorb slow neutrons coming directly from the source. Only a negligible number of counts due to fast neutrons that pass through the Cd sheet were registered. Some of the neutrons interacting with the polymer diffuse back after being moderated and can be measured by the BF_3 detector.

In electron attenuation experiments the source to detector distance was only few cms. About 3.7×10^4 Bq(1μCi) ^{207}Bi, that emits several conversion electrons, was used. The counting system was a surface barrier detector (type PD-50-24-500, Princeton-Gamma-Tech, U.S.A.) together with 1024 channels multichannel analyzer (series 30,

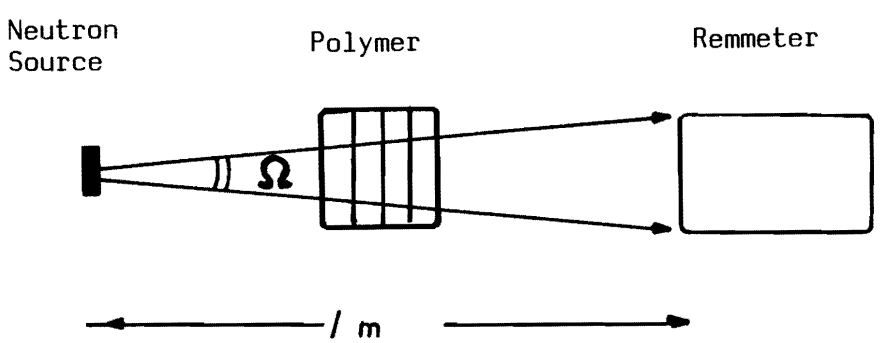

Figure 1. Set-up Geometry for Neutron Attenuation Measurements.

Canberra, U.S.A.) and the rest of nuclear electronics. A thin window G.M. tube (type 7232 LND, Inc. U.S.A.) was also used.

RESULTS AND DISCUSSION

Fast neutron dose rate *versus* polyethylene (PE) and PVC thicknesses in g/cm^2 is shown in Figure 3. Attenuation in polystyrene (PS) [$(CH_2–CH–C_6H_5)_n$], another widely available polymer, was included. Polyethylene has more

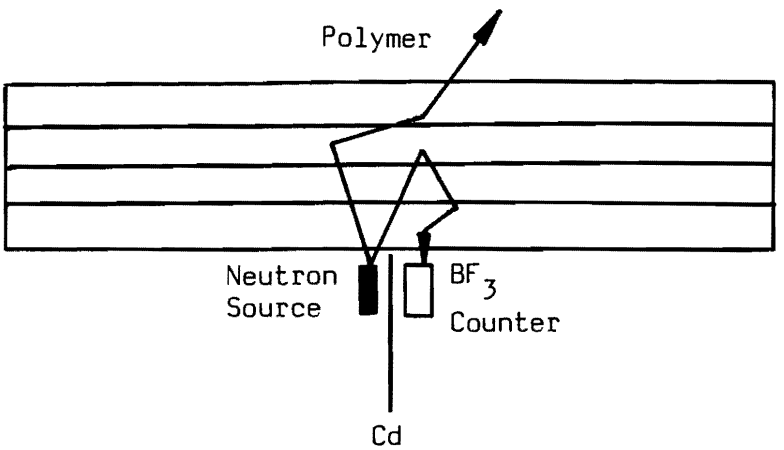

Figure 2. Set-up Geometry for Neutron Moderation and Back Diffusion Measurements.

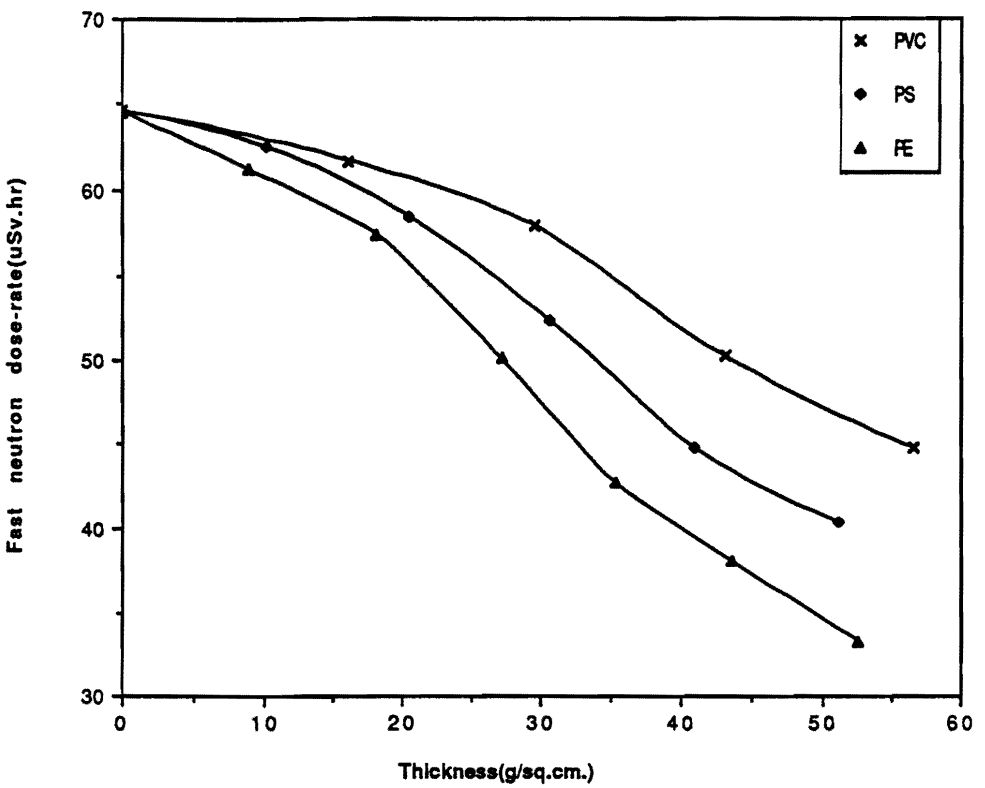

Figure 3. Dose-Rate of Attenuated Neutrons versus *Density Thickness.*

than twice the number of H atoms and about 70% more C atoms per cm^3 than PVC, but PVC has Cl atoms whose number is half that of C, and these have a higher absorption cross-section than H or C atoms. On a linear scale polyethylene will slow down more neutrons than PVC but absorb less. But because the density of polyethylene (0.94 g/cm^3) is much less than that of PVC (1.4 g/cm^3), the difference in attenuation in density thickness is very clear as shown in the figure; polystyrene shows intermediate behavior. As the total cross-section of H and C atoms decreases with energy, it is expected that lower energy neutrons are removed first. After a thickness of more than about 20 g/cm^2 the curves are steeper. At larger thickness those neutrons that undergo small scattering angle may have a chance for a second interaction because their mean free path will be smaller and they will, therefore, be removed.

Fast neutron attenuation in polymers as compared with that of high density heavy elements, where inelastic interaction is mainly responsible for slowing down neutrons, was made in comparison with iron, an element of high removal cross-section for fast neutrons [2]. The attenuation in iron and polyethylene *versus* both linear and density thicknesses are shown in Figure 4. PE shows superior attenuation per density thickness change and clearly better attenuation per linear one. Elastic collision in PE is probably more effective for removing or slowing down fast neutrons than the inelastic and absorption interactions in iron.

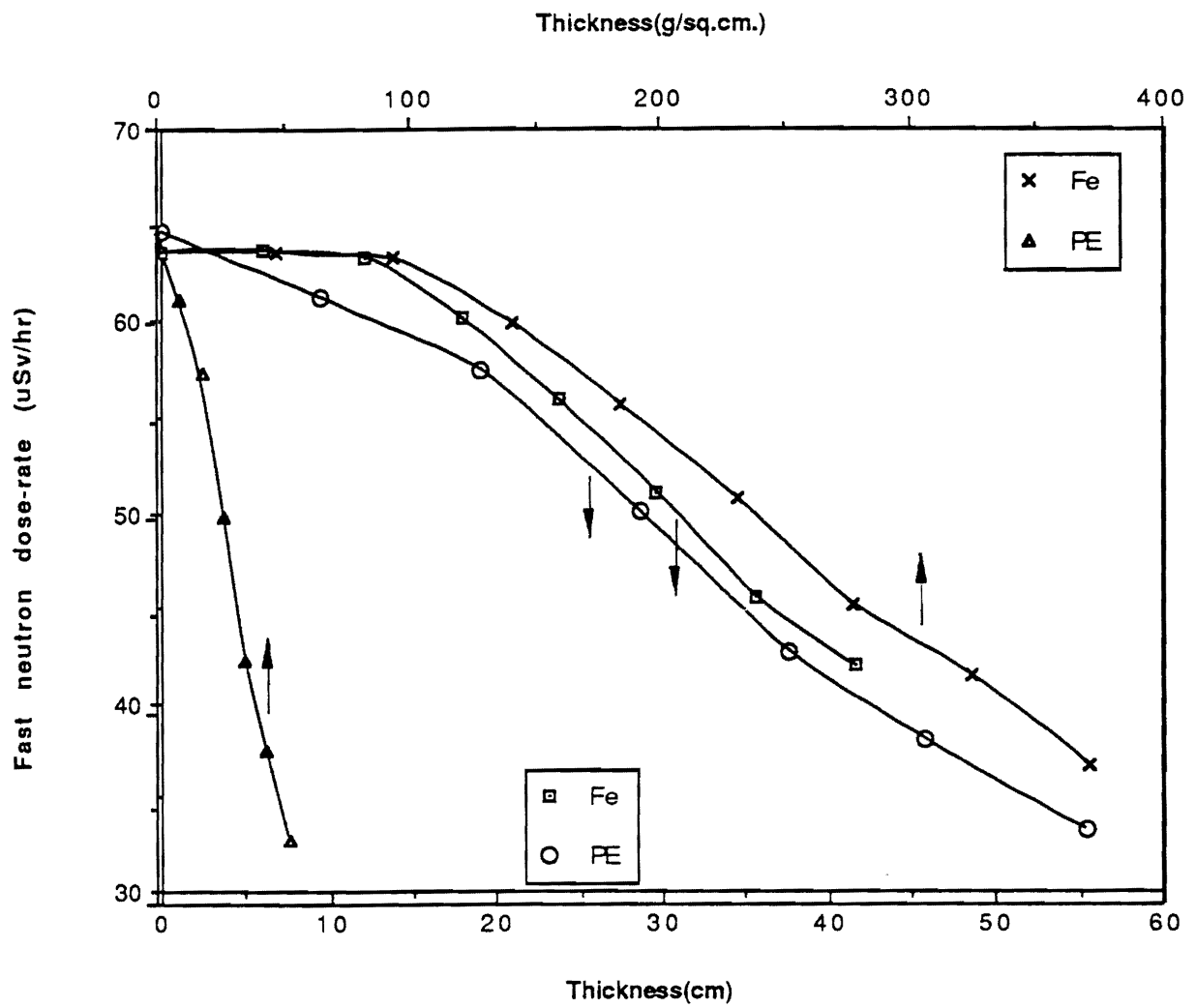

Figure 4. Dose-Rate of Attenuated Neutrons in Both Linear and Density Thickness.

In the neutron moderation and back diffusion experiments shown in Figure 2, polymer samples of about 10 cm diameter were introduced gradually and the corresponding counts for 100 s of slowed down, back diffused neutrons, were registered by the BF_3 counter. The results are shown in Figure 5 for three types of polymers, PE, PVC, and polymethylmethacrylate (PM) $[(CH_2–C–CH_3–OCOCH_3)_n]$ whose density was 1.15 g/cm^3. At the beginning parts of the presented curves the calculated counting sensitivities, given as the count rate per unit activity of the ^{241}Am–Be neutron source per unit thickness change, were approximately 0.9, 0.5, and 0.07 counts/s.Ci.mm for PE, PM, and PVC respectively. Decrease in slope was observed at a thickness of about 50–60 mm because back diffused neutrons will suffer self-absorption and scattering processes on their way back to the detector, which becomes clearer at larger thickness. Furthermore, the solid angle subtended by the sample as the detector decreases with thickness, reducing the fraction of emitted neutrons subject to the interaction process. It was observed experimentally that an increase in sample diameter to about 20 cm doubles the counts in all sample types studied. At larger diameter no significant increase was observed for the thicknesses of interest. These curves can be considered as characteristic curves for these polymers in this specific geometry, where any deviation due to density change, voids, or the existence of impurities would give different counts. The system can further be taken as a thickness measuring one.

The intensity of back diffused neutrons depends mainly on the moderation of neutrons by polymers and would, therefore, be affected by change in thickness and density. For PE whose counting sensitivity was about 0.9 counts/s.Ci.mm, a 10 Ci (3.7×10^{11} Bq) source for a testing time of 10 min will give 5400 counts change

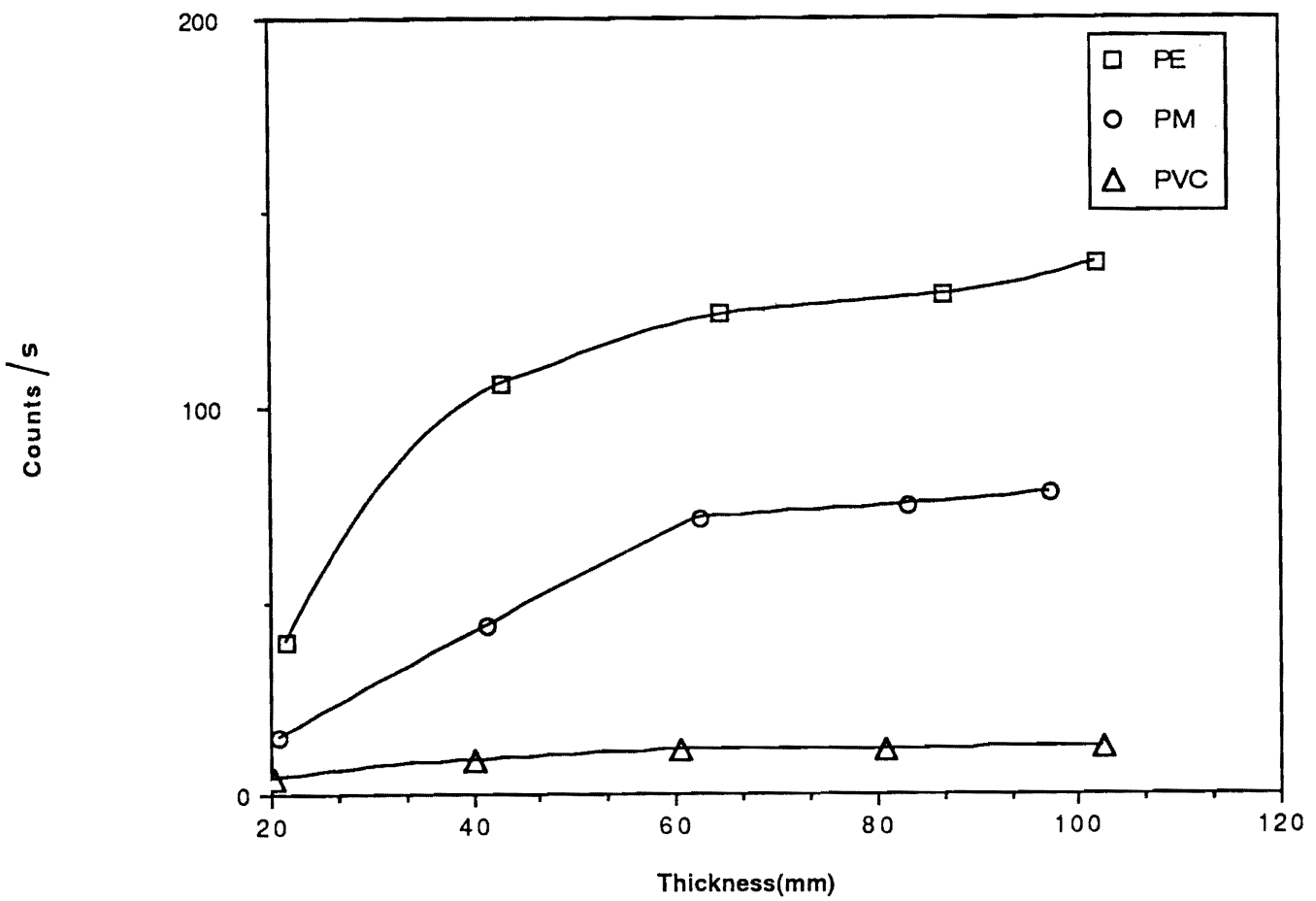

Figure 5. Back Diffused Neutrons Count Rate versus *Thickness.*

per mm change in thickness. The ratio of 2–3 standard deviations to the total counts will be about 3%. Therefore about 0.03 mm change in thickness should be detected. This is slightly better than the change measured by gamma back scattering method [1].

A change in density due to the existence of cracks, voids or any other factor during production would change the macroscopic scattering and absorption cross-sections and hence the intensity of the low energy back diffused neutrons. At a thickness of 6.5 mm the counts registered by the BF_3 counter due to back diffusion from PE were about 125 count/s for the 3 Ci source. Counts for 10 min of 10 Ci (3.7×10^{11} Bq) source will be 2.5×10^5. Assuming again that a count change of 2–3 standard deviations are measurable and that the count rate in the detector is proportional to the total macroscopic cross section of PE, a change of 0.5% in density would be measurable. The density of PE is a very important factor for the end point application of the product. Sensitivity for density change in PVC is an order of magnitude less, while that of PM would be about half that of PE.

It is expected that the existence of impurities, particularly those of high absorption cross section can reasonably be detected. As different additives are usually introduced in polymers to give it certain desirable properties, the technique would be useful for quality control. Considering, for example, the difference in counts between PE and PVC, those of the latter drop one order of magnitude because of the existence of Cl atoms that have intermediate absorption cross section for thermal neutrons (σ_a = 33.2 b at 0.025 eV). Furthermore, the scattering cross-section of PE is an order of magnitude higher than most elements. Therefore, the existence of impurities would also reduce the number of back diffused neutrons in addition to absorption. Considering absorption only and counting time for 10 min of back diffused neutrons from 10 Ci (3.7×10^{11} Bq) source in PE at a thickness of 6.5 mm, then the existence of a few atoms of very high absorption cross-section such as Cd or Sm per million atoms will give a measurable change in counts. A few impurity atoms of intermediate absorption cross section, such as In (σ_a = 193.5 b at 0.025 eV), per 10^5 atoms can be detected, while those of low absorption cross section such as Mn (σ_a = 13 b at 0.025 eV) are detectable at the level of one atom in a thousand atoms of polymer. Impurities in PVC are an order of magnitude less detectable because of the lower intensity of back diffused thermal neutrons due to absorption in Cl. Impurities in PM, and in most other types of polymers, would be detected at an efficiency close to that of PE.

It should be clarified here that this method does not identify the absorbing element. But because it is simple and fast, it may have a role in quality control for early identification of materials at deviant properties. Samples that exhibit noticeable change in counts may be subject to further detailed analysis. As counts are clearly dependent on the type of polymer used for interaction, the technique may be used as a fast method for polymer identification, particularly that many polymers have the same outside appearance.

There is a possibility of doubling the efficiency by using larger samples or by surrounding the detector-source by the sample materials. Using a source that has a softer neutron spectrum, such as ^{252}Cf, would be more useful, where neutrons are moderated within a shorter distance. The ^{252}Cf neutron emission per unit radioactivity is three orders of magnitude greater than that for the ^{241}Am–Be source. The sensitivity would therefore be improved, or counting time would be reduced by the same magnitude.

Polymers are produced in thin films. For thickness measurements a new approach have been used by measuring the change in the energy of the penetrating electrons. In the energy range between 0.5 MeV – 5 MeV, the specific ionization of electrons is almost constant with variation of about 4%. Therefore, electrons lose a constant amount of energy per unit distance traveled. The electron creates 60 ion-pairs per mg/cm^2 in passing through C_2H_4 gas where it loses 26.3 eV to create an ion-pair; the same is expected in polyethylene.

The change in energy of the four monoenergetic electrons of ^{207}Bi *versus* PE thickness is shown in Figure 6, using the surface barrier detector for 80 s counting time. As the thickness increases, peaks widen due to the strong straggling effect of electrons in the medium, but for all measurements taken they were reasonably clear. The count rate *versus* PE thickness using the G–M tube at few cms between the detector and the source are shown in the same figure. The tail that appears at the end of the curve is due mainly to gamma-rays which undergo minimum attenuation by the plastic. Counts *versus* thickness of three types of polymers including PE are shown in Figure 7.

The electron densities of PE, PM, and PVC are 0.537 N_A, 0.620 N_A, and 0.717 N_A, where N_A is Avogadro's number. PVC shows the highest electron attenuation because of its highest electron density, followed by PM and PE.

In these experiments, the system sensitivity for change in energy with thickness was about 200 keV/mm. As a change in energy of about 2 keV is measurable, thickness change of about 0.01 mm can be detected compared to about 0.05 mm by change in intensity method. In the latter method the counting sensitivity for PVC, as an example, was about 30 counts/s.mCi.mm., but can be improved by using a stronger source. Counting time for 10 min of about 100 mCi (3.7×10^9 Bq) source would give 1.8×10^6 counts/mm. Assuming a change of 2–3 standard deviation is detectable, about 0.002 mm thickness change would be measurable. Moreover, few Cl atoms change in the polymer, per thousand atoms is also detectable. Chlorine contributes to more than 30% of the electrons in PVC. As polymers have mainly low atomic number, the existence of higher atomic number impurity atoms would easily be detected. For lead, for example, a well known additive to PVC, which contains about an order of magnitude more electrons per unit volume over most polymers, a few atoms change per 10^4 atoms is measurable. The same will be the case, for example, with iron, copper, or magnesium impurities.

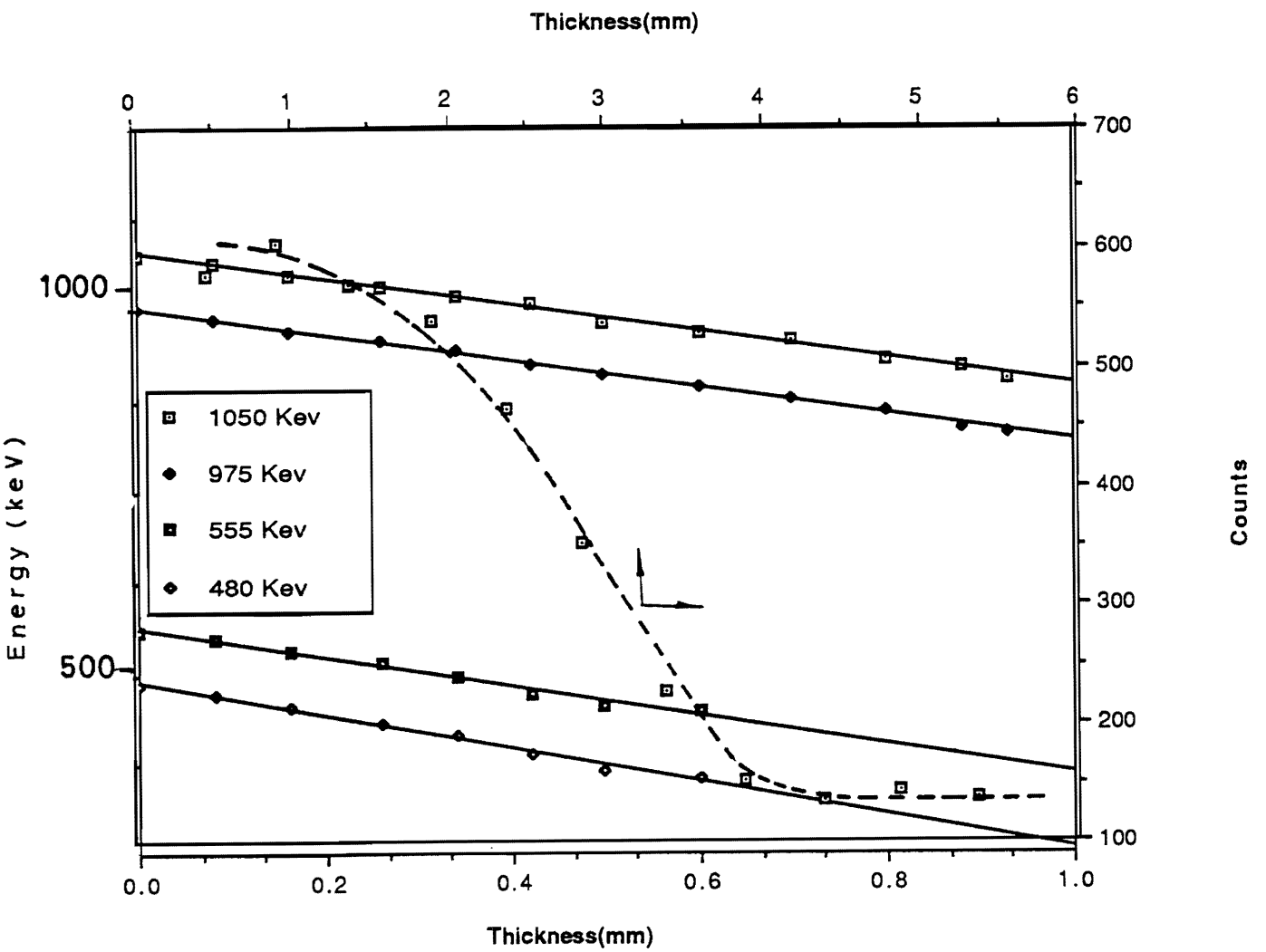

Figure 6. Energy of Four Monoenergetic Electron Peaks (Solid Line) and Counts (Dashed Line) versus *Polyethylene Thickness.*

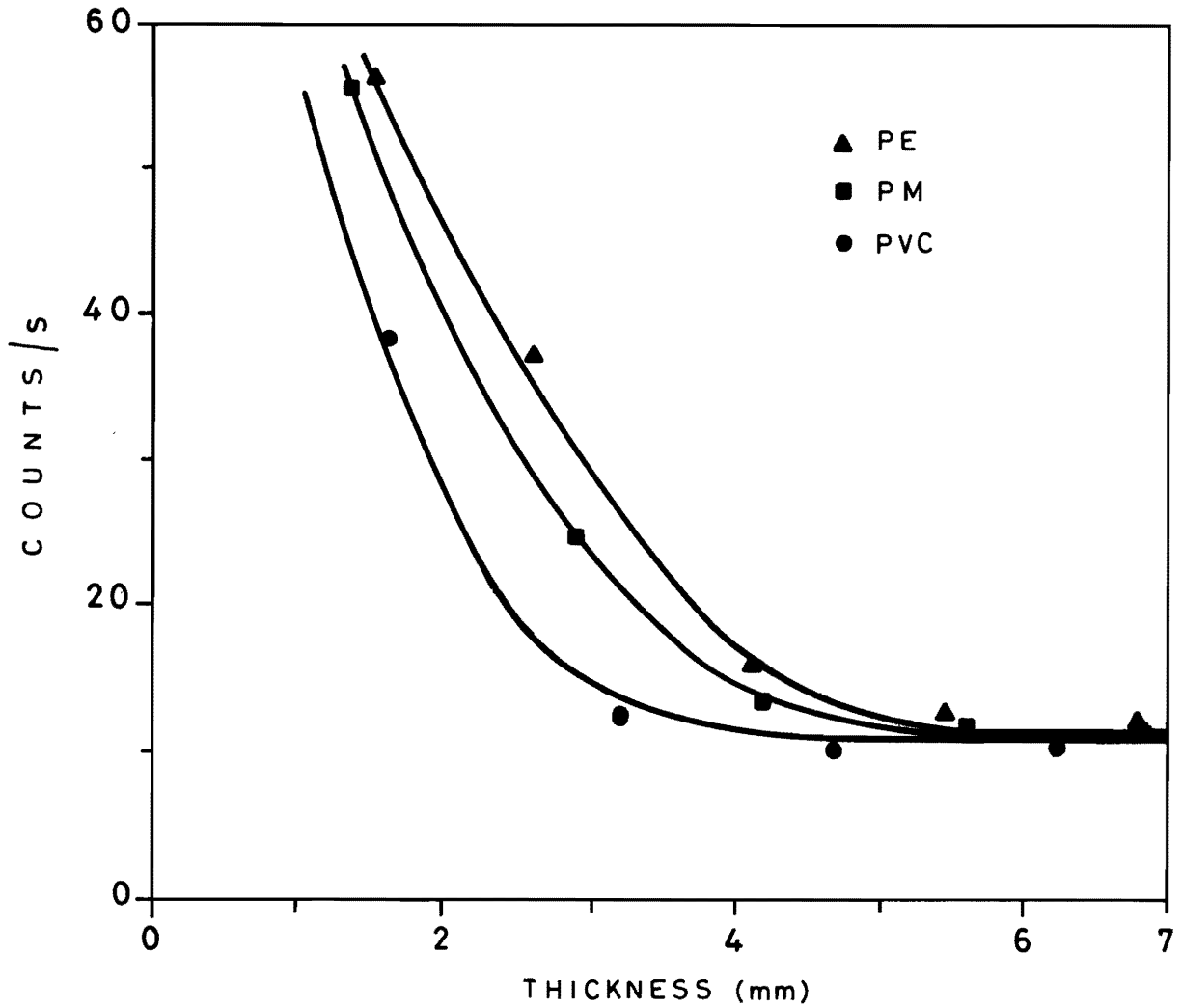

Figure 7. Count Rate versus *Polymer Thickness of Conversion Electrons.*

CONCLUSION

The attenuation of ^{241}Am–Be neutron dose rate shows the superiority of PE in density thickness over other types of polymers. Polyethylene shows much higher neutron attenuation than iron of the same weight while if both have the same linear thickness iron shows also inferior neutron attenuation. Although PVC has less H and C atoms per unit volume than PE, the Cl atom has relatively large absorption peak at intermediate energy. As the two materials have closely similar neutron attenuation characteristics in linear thickness, PVC can be more effective for total radiation dose attenuation because of its higher gamma-ray attenuation where gamma radiation, in most cases, accompanies neutrons.

The attenuation and moderation of neutrons can successfully be used for measuring large thickness with good accuracy. Existence of voids and changes in density, important factors for end point applications of polymers, can be measured in a sensitive way. The system is also sensitive to the existence of certain impurity atoms particularly those of high neutron absorption cross section. This characteristic may be used for controlling impurity atoms intentionally added to polymers to improve some of their properties.

The change in electron energy and the attenuation in its intensity can be used for polymer thin film thickness or density measurements. Although the described system can measure thickness change of the order of 10^{-2} mm, similar systems of higher source strength can measure a change of the order of 10^{-3} mm. The system is particularly sensitive for high atomic number impurity atoms, not necessarily detected by the neutron interaction method.

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