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सत्यमेव जयते

भारत सरकार

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गामा-किरण स्पेक्ट्रोमीटर द्वारा विलयन में यूरेनियम के ऑन लाइन निर्धारण हेतु तकनीक का विकास

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सारांश :

विलयन रूप में यूरेनियम के सतत् मॉनीटरन हेतु गामा-किरण स्पेक्ट्रोमीटर पर आधारित एक तकनीक का विकास किया गया । अनुकारित पात्र एवं सहयोगी प्रणाली का अभिकल्पन एवं संविरचन कर लिया गया ताकि क्षमता अंशशोधन वक्र का विकास किया जा सके तथा 185.7 (^{235}U) गामा किरण का प्रयोग करते हुए यूरेनियम के आकलन हेतु संसूचन सीमाओं को प्राप्त किया जा सके । संसूचक ज्यामिती के मानक स्रोत HPGe संसूचक माउंट में प्रणाली का प्रयोग करते हुए इसकी क्षमता की गणना हेतु इस प्रणाली का अंशाकन किया गया । संसूचक प्रणाली की संवेदनशीलता और यूरेनियम के निम्न स्तरीय आकलन हेतु काल गणना का भी स्थापन किया गया । मॉनीटर की संसूचक सीमा विलयन के प्रत्येक लीटर यूरेनियम की ~ 10 mg है । विलयन की घनत्व विभिन्नता सुधारने हेतु एक प्रयोग किया गया जिसमें ^{235}U की 185.7 keV गामा किरणों की गणना दर की विभिन्नता का विलयन के घनत्व की क्रिया के रूप में अध्ययन किया गया । इस रिपोर्ट में विलयन धारा में यूरेनियम के निर्धारण हेतु सतत मॉनीटर के विकास के विवरण प्रस्तुत किये गये हैं ।

Development of a technique for the on line determination of Uranium in solution by gamma ray spectrometry

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Abstract:

A technique based on gamma ray spectrometry has been developed for the continuous monitoring of uranium in the solution form. Simulated container and support system was designed and fabricated for the development of an efficiency calibration curve and to find the detection limit for the estimation of uranium using 185.7 keV (^{235}U) gamma ray. The system was calibrated for its counting efficiency using HPGe detector system, in a standard source mount to detector geometry. The sensitivity of the detection system and counting time for low-level estimation of uranium has also been established. The detection limit of the monitor is ~ 10 mg of Uranium per litre of the solution. In order to correct for the density variation of the solution, experiment was carried to study the variation of count rate of 185.7 keV gamma ray of ^{235}U as a function of the density of the solution. This report gives the details of the development of a continuous monitor for the determination of uranium in the solution streams.

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Introduction:

A continuous monitoring system is required to monitor uranium in the solution form at the input stage and at the output stage during any chemical separation of uranium. The amount of uranium in the solution form may vary from 10 mg per litre onwards. Uranium analysis in this concentration range is done by fluorimetry, spectrophotometry, I.C.P.[(1)] and by M.S. techniques. An alternative technique based on gamma ray spectrometry has been developed. Experiments were carried out to study the feasibility and to set up an on line monitoring system for the determination of uranium in solution form using gamma ray spectrometry. Though other techniques[(2)] like neutron activation, emission spectrometry, mass spectrometry, x-ray fluorescence can also be used, but the gamma ray spectrometry is a non-destructive, convenient and relatively less expensive technique for the estimation. The required simulated container and support system was designed and fabricated for the development of an efficiency calibration curve and to find the detection limit for the estimation of uranium using 185.7 keV (^{235}U) gamma ray. The representative sample of uranium was counted on a HPGe detector and also on a NaI(Tl) to have an idea of count rate. The very nature of interaction of gamma rays with matter results in the variation of detector efficiency as a function of energy of gamma ray. For quantitative estimation of radionuclides, it is therefore; necessary to calibrate the counting set up for its counting efficiency, in a standard source mount to detector geometry[(3)]. High resolution HPGe detector system has been used for the development of the monitoring system. The sensitivity of the detection system and counting time for low-level estimation of uranium has also been established. This report gives the details of the development of a continuous monitor for uranium in the solution form.

Fabrication of simulated container:

To estimate, the low concentration of uranium in the solution form, a simulated container was designed and fabricated to minimize the attenuation and to have maximum amount of counts in the photo peak. The 185.7 keV gamma ray of ^{235}U is absorbed up to 50% in the thickness of about 4 cm of water. A double walled cylindrical container was designed so as to surround the detector with maximum amount of solution within a depth of 2.0 cm. Plastic material was used for the fabrication of the double walled cylindrical container, so as to have minimum absorption of gamma rays. The details of the simulated container are shown in Figure 1. The simulated container has an inlet and an outlet for the solution to flow in and out respectively. This will maintain a continuous flow of liquid that can be monitored on line. A support system was also suitably designed so as to keep the simulated container on it. This is to avoid any undue pressure on the detector.

Preparation of standard solutions:

The efficiency calibration of the gamma ray detection system was done using standard solutions of known concentration. A standard calibrated stock solution of uranium was prepared by dissolving a known amount of uranium. A known amount of this stock solution was transferred to five different bottles. The volume of each solution was made up to 1500 ml by adding suitable

amount of distilled water in the bottle. These bottles were having standard uranium solutions with concentration varying from 10 mg/litre to 260 mg/litre. Before the counting of any standard solution, the simulated container was thoroughly washed and dried. The required standard solution from the bottle was transferred to the simulated container for the gamma counting.

Estimation using NaI(Tl) detector system:

The NaI(Tl) detector was calibrated for the energy of the gamma rays using standard sources of ^{241}Am, ^{137}Cs and ^{60}Co. A source of ^{235}U was also counted in the same settings to identify the 185.7 keV gamma ray. The simulated container was put on the support system so as to surround the NaI(Tl) detector. The simulated container was containing solution of uranium (66 mg/litre). The counting was continued for 24 hours. The clear peak of 185.7 keV gamma ray could not be seen in the gamma ray spectrum (Figure 2) for this low concentration of uranium. Though NaI(Tl) detector is having good efficiency, but the resolution is very poor (7%). Hence, the peak is broad and spreads in a large energy range. In case of NaI(Tl) detector the 185.7 keV peak is also lying on the rising part of backscatter peak. All these factors, makes it difficult to clearly see the 185.7 keV peak for low concentration of uranium with NaI(Tl) detector. Hence, it was decided to carry out the experiment using a high-resolution HPGe detector. Figure 3 shows the gamma ray spectrum of Uranium sample (10 mg/litre) using HPGe detector. Here the 185.7 keV peak is clearly resolved even for much lower amount of the sample.

Estimation using HPGe detector system:

The HPGe detector is having less efficiency as compared to NaI(Tl) detector. But because of high resolution, the gamma ray peak are very sharp. This is responsible for the lower detection limit of HPGe detector as compared to NaI(Tl) detector. Hence, it was decided to develop the monitoring system using a HPGe detector for low concentration of uranium. The HPGe detector was calibrated for its energy using ^{152}Eu source. The standard solution from the bottle was transferred to the simulated container for the gamma counting. The simulated container was surrounding the detector so as to have maximum efficiency. After attaining stability for the detection system, the counting was carried out for 20 to 25 hours. The counting was repeated for all the standard solution in the same geometrical settings. In order to have the idea of the Uranium impurity in the distilled water used for preparation of standard solutions and external background of 185.7 keV gamma ray due to the surrounding, the counting was also carried out for the simulated container by filling it with distilled water. This was done to have the idea of the net background. The gamma ray spectrum analysis was carried out using program DSPG[(4)]. The variation of the net count rate for the 185.7 keV gamma ray of ^{235}U was studied as a function of the uranium present in the solution. Table 1 gives the variation of count rate of the simulated container for different concentrations of uranium solution (mg/litre) using a HPGe detector. Figure 4 shows the plot for the variation of the count rate of 185.7 keV using a HPGe detector as a function of uranium concentration in the simulated container. The calibration plot was found to be nearly a straight line. This calibration plot can be used to determine the unknown amount of uranium in any solution form.

Determination of detection limit for the concentrations of uranium solution:

The concentration of uranium in the solution can vary from 10 mg/litre to 200 mg/litre. Hence, it was required to have an estimate for the lowest detection limit within a suitable counting time using simulated container (Cyl-1) and HPGe detector system. The solution containing lowest concentration of uranium (10 mg/litre) among the standard solutions was counted using the HPGe detector system. The counting time of the HPGe detector system was varied from a few seconds

to 70000 seconds, to study the variation of count rate as a function of the duration of the counting for the uranium solution present in the simulated container. Table 2 gives the results of the determination of minimum detection limit. The count rate varied maximum up to 30% for the duration of counting up to 2500 seconds. Where as for the longer duration of counting (10000 seconds), the count rate varied within 12%. Hence, we can detect the uranium in the solution form up to a concentration of 10 mg/litre within the counting time of 2500 seconds with an average precession of 30%. Similarly, experiment was also carried out for finding the detection limit of uranium solution containing higher concentration of uranium (~200 mg/litre) in the simulated container. The count rate varied maximum up to 30% for the shorter duration of counting (up to 200 seconds). Where as for the longer duration of counting (10000 seconds), the count rate varied less than 3%. Hence, we can detect the uranium solution having a concentration of 200 mg/litre within the counting time of 200 seconds up to an average accuracy of 30%.

In order to study the improvements in the results using higher volume of the solution and reducing the detector distance another simulated container (Cyl-2) was fabricated in which internal solution thickness was increased from 1.8 cm to 2.0 cm. Similar study was repeated using this container. The count rate (Table 2) varied maximum up to 20% for the shorter duration of counting (up to 1500 seconds). Where as for the longer duration of counting (10000 seconds), the count rate varied within 8%. Hence, we can detect the uranium in the solution up to a concentration of 10 mg/litre within the counting time of 1500 seconds with an average accuracy of 20%.

In order to study the improvements in the results using higher efficiency detectors. Another experiment was carried out using a 40% HPGe detector with the simulated container (Cyl-2). The counting time of this HPGe detector system was varied from a few seconds to 70000 seconds, to study the variation of count rate as a function of the duration of the counting for the Uranium solution (10 mg/litre) present in the simulated container. The count rate (Table 2) varied maximum up to 19% for the shorter duration of counting (up to 800 seconds). Where as for the longer duration of counting (10000 seconds), the count rate varied within 6%. Hence, we can detect the uranium in the solution up to a concentration of 10 mg/litre within the counting time of 800 seconds with an average accuracy of 19%.

Correction for the density variation of the solution:

Experiment was carried to study the variation of count rate of 185.7 keV gamma ray of ^{235}U as a function of density of solution, so as to correct for the density variation of the solution. Standard solutions of ^{235}U were prepared containing same amount of ^{235}U and having different types of aqueous solutions. These solutions were taken in the simulated container and counted on the HPGe detector in the required geometry. The gamma ray spectrum analysis was carried out to determine the variation of count rate as a function of density of the solution. The variation in density from 1 to 1.71 showed a variation up to 8.6% in the count rate (Figure 5). This data can be used for the correction of count rate for arriving at the actual amount of ^{235}U in the solution form.

Conclusions:

Detection limit for estimation of uranium in the solution form can be lowered by following modifications.

- Wall thickness of the simulated container facing the HPGe detector, can be reduced to reduce the absorption of 185.7 keV gamma rays of ^{235}U.
- The gap between the simulated container and HPGe detector can be reduced to increase the counting efficiency.

- The volume of the solution present in the simulated container can be increased by increasing the size of the simulated container. This will increase the amount of ^{235}U being counted.
- The detection limits of the system can be further improved using a large size HPGe detector. This will be increase the efficiency of detection as well as the detector will be able to see larger volume of the solution.

Acknowledgment:

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Table 1. Variation of count rate of the simulated container for different concentrations of uranium solution (mg/litre) using a HPGe detector.

Sample number	Concentration of Uranium mg/litre	C.P.S. for 185.7 keV peak	Net C.P.S. after background correction
U2Std-01	11.7	1.52E-01	1.32E-01
U2Std-02	23.3	1.94E-01	1.75E-01
U2Std-03	58.4	3.61E-01	3.42E-01
U2Std-04	116.7	6.37E-01	6.17E-01
U2Std-05	194.5	9.89E-01	9.70E-01
U2Std-06	256.7	1.27E+00	1.25E+00
Water	0 (Background)	1.92E-02	0.00E+00

Table 2. Determination of minimum detection limit of uranium solution (mg/litre) present in a simulated container using a HPGe detector system.

Solution	HPGe detector & Cylinder system	Concentration mg/litre	Minimum counting time required for detection (seconds)	Precision for minimum counting time	Precision for (10000 seconds)
U-10	10% HPGe & Cyl-1	10	2500	30%.	12%
U-200	10% HPGe & Cyl-1	200	200	30%	3%.
U-10	10% HPGe & Cyl-2	10	1500	20%.	8%.
U-200	10% HPGe & Cyl-2	200	200	20%	3%.
U-10	40% HPGe & Cyl-2	10	800	19%.	6%.

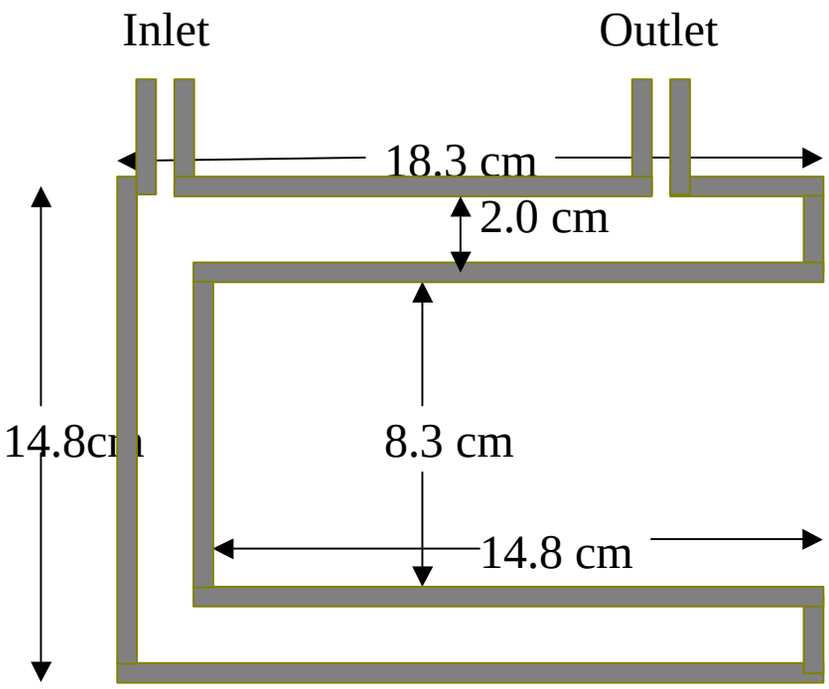

Figure 1. Simulated container for the estimation of low concentration of uranium (mg/litre) in the solution stream.

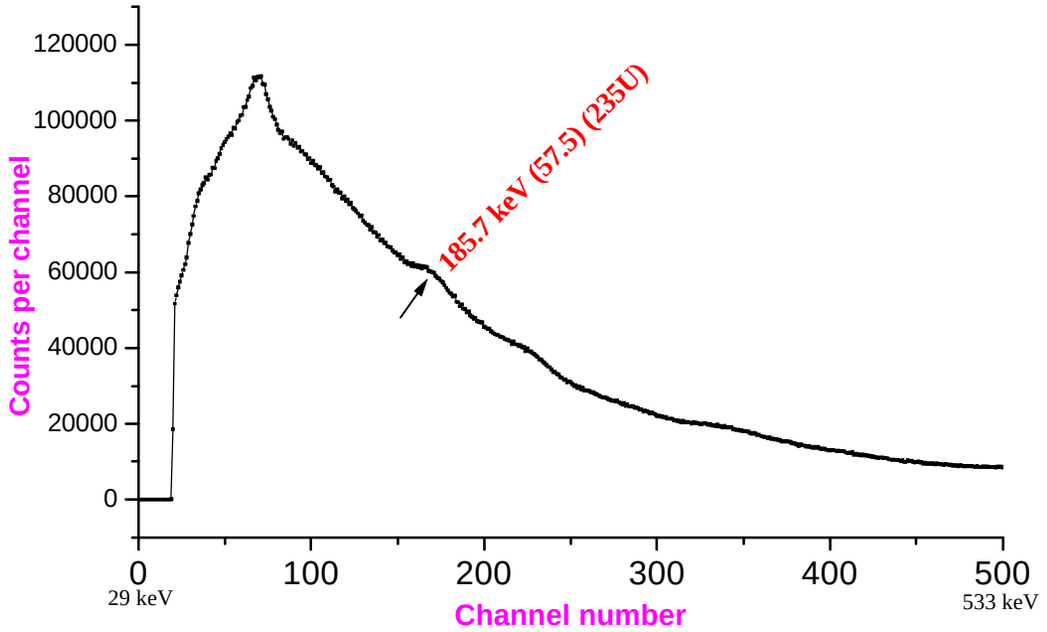

Figure 2. Gamma ray spectrum of Uranium (66 mg/l) on NaI(Tl) detector

Counting time = 68500 seconds

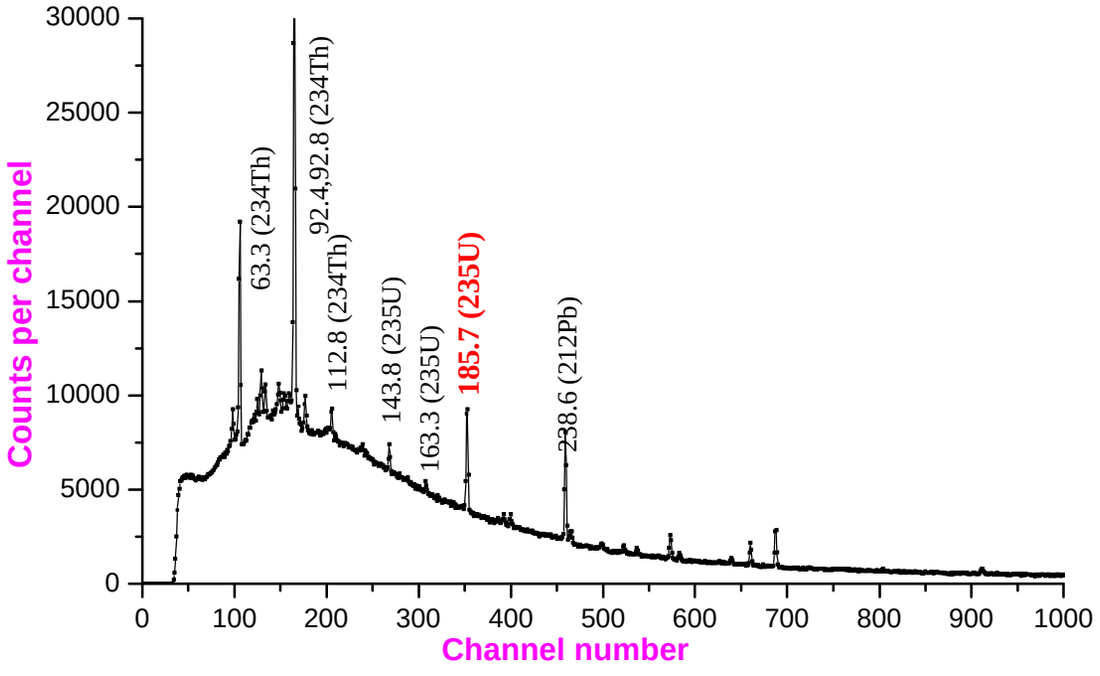

Figure 3. Gamma ray spectrum of Uranium (10mg/l) on 10% HPGe

Counting time = 99500 seconds

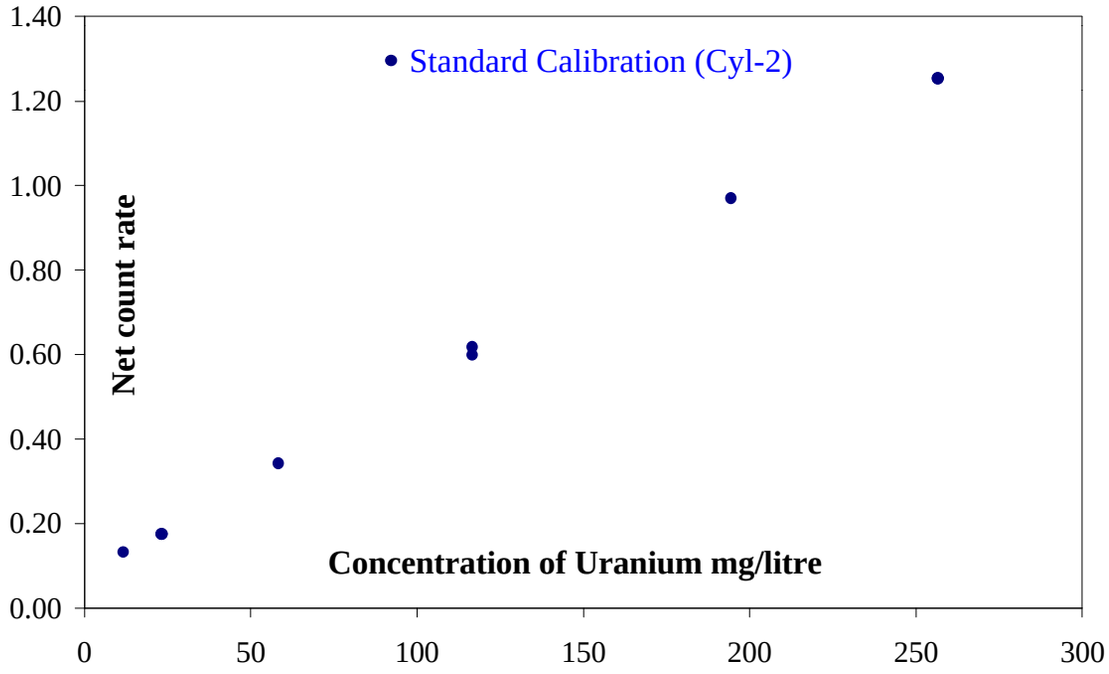

Figure 4. Plot count rate for 185.7 keV of 235-U as a function of amount of Uranium in the standard solution.

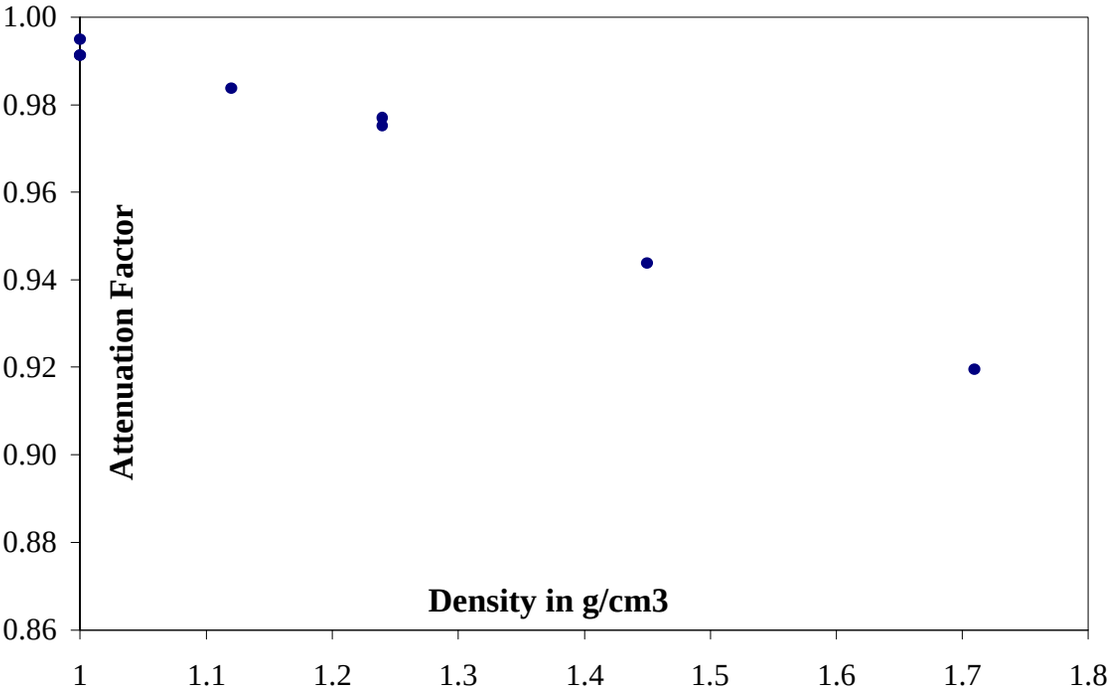

Figure 5. Plot for the variation of attenuation factor of 185.7 keV as a function of density of solution (Cyl-2)