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60 ***Abstract :*** Non destructive gamma ray techniques have been developed for analysis of isotopic abundances and concentrations of plutonium and uranium in the respective product solutions of a reprocessing plant. The method involves analysis of gamma rays emitted from the sample and uses a multichannel analyser system. Data reduction and interpretation of these techniques are tedious and time consuming. In order to make it possible to use them in routine analysis, computer programs have been developed in HP-BASIC language which can be used in HP-9845B desktop computer. A set of programs, for plutonium estimation by high resolution gamma spectrometry and for on-line measurement of uranium by gamma ray spectrometry are described in this report.

70 ***Keywords/Descriptors :*** GAMMA SPECTROSCOPY; ISOTOPE RATIO; PLUTONIUM; URANIUM; BASIC; COMPUTER CALCULATIONS; ON-LINE MEASUREMENT SYSTEMS; NONDESTRUCTIVE ANALYSIS; HIGH-PURITY GE DETECTORS; AQUEOUS SOLUTIONS; AMERICIUM 241; P CODES; U CODES; DATA PROCESSING; QUANTITY RATIO; URANIUM 235;URANIUM 238; PLUTONIUM 238; PLUTONIUM 240;PLUTONIUM 239; PLUTONIUM 242

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ABSTRACT

Non destructive gamma ray techniques have been developed for analysis of isotopic abundances and concentrations of plutonium and uranium in the respective product solutions of a reprocessing plant. The method involves analysis of gamma rays emitted from the sample and uses a multichannel analyser system. Data reduction and interpretation of these techniques are tedious and time consuming. In order to make it possible to use them in routine analysis, computer programs have been developed in HP-BASIC language which can be used in HP-9845B desktop computer. A set of programs, for plutonium estimation by high resolution gamma ray spectrometry and for on-line measurement of uranium by gamma ray spectrometry are described in this report.

1. INTRODUCTION

Determination of plutonium and uranium in the product solutions of a reprocessing plant are normally carried out by controlled potential coulometry(1) and Redox potentiometry(2) for the purpose of nuclear material accounting. Though these methods are accurate, they are time consuming. The nondestructive analysis (NDA) techniques are increasingly in use for the timely characterization, accounting and hence safeguarding of special nuclear materials. The advantages of NDA methods are; i) simplicity and speed of analysis(3), ii) comparable accuracy and precision, iii) no sampling is required, if used as on-line, and iv) additional information of isotopic composition and Am241 content (in the case of Pu analysis) is available. In PREFRE laboratory NDA methods are standardised for the measurement of plutonium and uranium using passive gamma ray spectrometry. These methods use a multichannel analyser with suitable detectors. Because of tedious and time consuming steps of data reduction and data interpretation of these methods, computers are commonly employed for carrying out this work. A computer program in FORTRAN language has already been developed by Gunnink(4) for the data reduction in plutonium analysis.

In our laboratory, two different programs in HP-BASIC language have been developed for the computation of data in plutonium and uranium analysis. These programs are written to use on a HP-9845B desktop computer interfaced

to a disk mass storage device.The present paper describes in detail the methods and the software involved in the data reduction of these techniques.

2. MEASUREMENT OF PLUTONIUM IN PRODUCT SOLUTION BY HIGH RESOLUTION GAMMA RAY SPECTROMETRY.

2.1 Method:

Plutonium isotopes and their daughter products emit more than 400 characteristic gamma rays. Isotope ratios of plutonium can be determined by measuring selected close lying gamma ray pairs in different energy regions. Seven of them in low energy region from 26 kev to 148 kev are used in the present work. The isotopes and gamma energies chosen are shown in Table 1. In general the isotope ratio of two plutonium isotopes i and j is determined by measuring intensities of selected gamma rays a and b using the following equation:

$$\frac{Ni}{Nj} = \frac{Ia}{Ib} * \frac{BIb}{BIa} * \frac{T(1/2)i}{T(1/2)j} * \frac{REb}{REa} \qquad \text{--------1.}$$

where, I = intensity of gamma ray(count rate).

BI = branching intensity.

T(1/2)= half life of the isotope.

RE = relative efficiency.

2.2 Instrumental

The experimental set up used is shown in fig.1. A high purity germanium (HPGe) detector having a sensitive volume one c.c. and resolution of 485 ev at 122 kev with vertical cryostat configuration and connected to a SILENA 8K multi-channel analyser (MCA) is used in this work.

The program for data reduction is run on a HP-9845B desk top computer. It has a 640K memory and mass storage devices of two cassette drives and two 8 inch flexible disc drives of 100K and 500K memory respectively. It is also equipped with an internal thermal printer and an external EPSON dot matrix printer.

2.3 Procedure

Plutonium product solutions from the intermediate product tanks(PUP & PUS) and plutonium oxide product dissolved solutions can be analysed by this technique. Solutions ranging in plutonium concentration from 1 g/l to 20 g/l are analysed. 2ml of plutonium nitrate solution is taken in a small sample bottle, sealed and pushed into another plastic bottle of nearly 5ml capacity. This sample is placed in a fixed position above the detector. Counting of the sample is carried out on the HPGe-MCA system for 1800 seconds. The analyser gives only gross and net counts under different regions of interest (Peaks). Further data reduction and data interpretations are carried out using the program titled PU/GS.

2.4 Software description.

Program is prepared in the operator interactive method. It is convenient method as the operator has only to answer the questions prompted by the program. Software involved is shown in appendix-1 .

2.4.1 Data inputs

Out put of the MCA system form the data inputs. These include sample identification, date of analysis, counting time and area counts of different regions of interest.

2.4.2 Calculations of relative efficiencies.

It is evident from equation 1 that to calculate the ratio of two isotopes from the count rate of two close lying energy peaks of these isotopes, it is necessary to know the efficiencies/ relative efficiencies of these energy peaks. Relative efficiency depends upon the detector efficiency, energy of the peak, sample matrix and sample to detector geometry. Hence first step of the program is to determine the relative efficiencies of different energy peaks. Option is provided in the program to use either an internal calibration method or a standard calibration method for obtaining the relative efficiencies.

Internal calibration method is normally employed for the analysis of plutonium solutions of higher concentration and for the plutonium isotopic assay in oxide samples. In this method the information of relative efficiencies is

generated from the measured sample spectrum itself. 26.34, 33.19 and 59.54 Kev lines of Am241 are chosen in lower energy region and 129.29, 144 and 203 Kev lines of Pu239 isotope are chosen in the higher energy region. Calculations carried out by the program are shown below.

$$Re1(26.34) = I1(26.34)/BI(26.34) \quad \text{----------2.}$$

$$Re2(33.19) = I2(33.19)/BI(33.19) \quad \text{----------3.}$$

$$Re3(59.54) = I3(59.54)/BI(59.54) \quad \text{----------4.}$$

where, Rei(E) is the relative efficiency of peak i of energy E and Ii(E) & Bi(E) are the area counts and branching intensities of respective energy peaks. Relative efficiencies thus determined are fitted into quadratic equation of the type shown, to extend the method of calculation of relative efficiencies for the other energy peaks.

$$Re1(26.34) = a0 + a1\ E1 + a2\ E1^2 \quad \text{-- (E1=26.34kev)----5}$$

$$Re2(33.19) = a0 + a1\ E2 + a2\ E2^2 \quad \text{-- (E2=33.19kev)----6}$$

$$Re3(59.54) = a0 + a1\ E3 + a2\ E3^2 \quad \text{-- (E3=59.54kev)----7}$$

where, a0, a1 and a2 are constants, found by following equations;

$$a2 = \frac{[(E2-E3)*(Re1-Re2)] - [(E1-E2)*(Re2-Re3)]}{[(E1-E2) * (E2-E3) * (E1-E3)]} \quad \text{----8.}$$

$$a1 = \frac{(Re1-Re2) - a2(E1^2 - E2^2)}{(E1-E2)} \quad \text{----------9.}$$

$$a0 = Re1 - a1E1 - a2E1^2 \quad \text{---------10.}$$

Having calculated the constants, relative efficiencies of other energy peaks in the low energy region (Pu isotopes) are calculated using the same quadratic equation. For

higher energy region (129 to 203 kev), Pu239 lines are used and relative efficiencies are calculated by the same procedure.

Standard calibration method for obtaining relative efficiencies is followed for plutonium nitrate solutions in a low range of concentration (1 to 15 g/l) where change in the relative efficiency due to sample attenuation is minimum In this method relative efficiences of different energy peaks are experimentaly determined using a set of standards and their values are available in the program for further calculations.

2.4.3 Isotope ratio calculation.

The isotope ratio of two plutonium isotopes is determined by measuring intensities of selected close lying gamma rays and using equation 1. Pu(240/239) ratio is calculated using the pairs of gamma lines 43.48 kev and 38.66 kev as well as 43.48 kev and 51.63 kev lines as follows.

$$N091 = \frac{I(43.48)}{I(51.63)} * \frac{Re(51.63)}{Re(43.48)} * 1.6134E\text{-}1 \qquad \text{-----11.}$$

$$N092 = \frac{I(43.48)}{I(38.66)} * \frac{Re(38.66)}{Re(43.48)} * \frac{6537*1.05E\text{-}4}{24150*4.5E\text{-}4} \qquad \text{---12.}$$

Average of these two isotope ratios is taken as the (240/239)ratio of the sample.

$$N09 = (N091 + N092)/2 \qquad \text{----------13.}$$

Same way (241/239) ratio is calculated by using 148.57 kev & 129.29 kev lines and is given by

$$N191 = \frac{I(148.57)}{I(129.29)} * \frac{Re(129.29)}{Re(148.57)} * 2.0099E\text{-}2 \quad \text{----------14.}$$

For Am241/Pu239 ratio 59.05 kev and 51.63 kev lines are used. Since Pu241 also contributes to 59.05 peak, a correction is incorporated in the calculation.

$$Na9 = \left[\frac{I(59.05)}{I(51.63)} * \frac{Re(51.63)}{Re(59.05)} - N191 * 5.2352E1 \right] * 1.3485E\text{-}5 \quad \text{------------15.}$$

In the case of (238/239) ratio 43.48 kev and 38.66 kev as well as 43.48 kev and 51.63 kev lines are chosen. Here again correction has been incorporated for the contribution of Am241 and Pu241 at 43.48 kev gamma line of Pu238.

$$N891 = \left[\frac{I(43.48)}{I(38.66)} * \frac{Re(38.66)}{Re(43.48)} - Na9 * 3.6917E2 - N191 * 9.3586E\text{-}2 \right] * 9.7023E\text{-}4 \quad \text{------------16.}$$

$$N892 = \left[\frac{I(43.48)}{I(51.66)} * \frac{Re(51.68)}{Re(43.48)} - Na9 * 1.4357E2 - N191 * 3.6395E\text{-}2 \right] * 2.4949E\text{-}3 \quad \text{------------17.}$$

(238/239) ratio is calculated from the average of two ratios obtained.

$$N89 = (N891 + N892)/2 \quad \text{------------18.}$$

Since Pu242 does not emit any characteristic gamma (242/239) ratio is computed by the relation,

$$N29 = K * (240/239) * (241/239) \quad \text{----------19.}$$

where K is a constant which depends upon the type of reactor. For power reactor fuels of CANDU type K is equal to 0.73 and this factor is obtained from mass spectrometric data of earlier processing campaign.

2.4.4 Isotopic composition/specific activity calculation.

Atom and weight percent of plutonium are calculated from the isotope ratios obtained and atomic mass of different plutonium isotopes. Using the nuclear data, the program also calculates the specific activity of plutonium which is required for use in radiometric analysis. Isotope ratios of 238/239, 239/239, 240/239, 241/239 and 242/239 are represented by R1 to R5 respectively.

$$T = \sum Ri \quad \text{----------20.}$$

$$\text{Atom percent of isotope i(Api)} = \frac{Ri}{T} * 100 \quad \text{--21.}$$

where, i = 1 to 5 for isotopes 238 to 242.

Weight percent is calculated using the atomic mass of different isotopes(Mi) and isotopic ratios(Ri)

$$\text{Average atomic weight} = \sum (Ri * Mi)/T \quad \text{---------22.}$$

$$\text{Weight percent of isotope i (Wpi)} = \frac{Ri * Mi}{\sum (Ri * Mi)} * 100 \quad \text{----23.}$$

Informations regarding the specific activity of different

isotopes in dpm/microgram (Spi) are available from the nuclear data i.e., atomic mass, half life of the respective isotopes and Avogadro number.

Specific activity of plutonium in sample (dpm/ug) is calculated from the weight percent of different isotopes and their specific activity and is given by

$$\text{Specific activity of plutonium (dpm/ug)} = \frac{\sum (Wpi * Spi)}{100} \quad \text{----24.}$$

2.5 Plutonium concentration determination.

Plutonium concentration is determined in solution from the intensities of 51 kev and 129 kev gamma lines of Pu239. Intensities(count rates) of these gamma lines with change in concentration are shown in figures 2 and 3. It can be seen that at 129 kev for a concentration range of 1 to 15g/l, the data fits with a linear curve of the type Y = mX + C, where as at 51 kev the curve is found to deviate slightly from the linear equation above 10 g/l due to sample attenuation. The program gives option to the user to find the concentration of plutonium using the exponential relation of the type Y=a*e^-bx or using linear equation in short intervals.

2.6 Data out.

Isotopic composition and plutonium concentration are printed out on a dot matrix printer or on an internal thermal printer. A typical out put is shown in Appendix-2.

3. MEASUREMENT OF URANIUM CONCENTRATION BY GAMMA RAY SPECTROMETRY.

3.1 Method

Uranium isotopes emit several gamma rays. The gamma rays associated with the daughter products of U235 and U238 185.7 kev, and 1001 kev respectively are used in this work. Isotope ratio is calculated from the ratio of measured intensities of these gamma rays.

3.2 Procedure.

A coaxial high purity germanium detector with a resolution of 1.8 kev at 1.33 Mev and volume of 50cc, coupled to a 8K multichannel analyser is used. A sketch of the set up is shown in figure 4. The sample cell of 50cc capacity is situated above the detector and uranium solution is pumped into the cell and circulated continuously througn the sample cell during the counting time of 1800 seconds. A Set of uranium standards in the concentration range of 50 g/l to 300 g/l are used for the calibration of the system.

3.3 Software description.

This program titled UONLINE is ''user friendly program''. Operator with minimum knowledge of computer can calculate the results using this program. Appendix-3 shows the listing of the program to perform this analysis.

3.3.1 Inputs

Data out put of multichannel analyser are entered along with other inputs namely, sample identification, date of analysis, and counting time.

3.3.2 Calculation of efficiency factor.

Efficiency factor of an energy peak includes both branching intensity and detection efficiency. They are predetermined using a set of standards. At 1001 kev (U238) there is little change in the sample attenuation over the entire concentration range and hence a constant efficiency factor (EF) is used. Sample attenuation of 185.7 kev energy peak (U235) increases with the concentration and hence the program includes a self calibration to compute the efficiency factor of 185.7 kev from the count rate obtained for the sample using a linear relation (fig.5)

3.3.3 Isotope ratio calculation.

Uranium isotope ratio of 235/238 is calculated from the intensities of gamma rays by,

$$R(235/238) = \frac{I(185.7)}{I(1001)} \times \frac{EF(1001)}{EF(185.7)} \times \frac{T1/2(235)}{T1/2(238)} \quad \text{----25}$$

Where T1/2 is the half life of the isotope. From this ratio weight fraction of U238 is calculated.

3.3.4 Uranium concentration measurement.

Concentration of U238 is calculated from the linear relationship of the type Y= MX + C, where Y is the intensity (I) of the 1001 kev and X is the U238 concentration (fig 6). M and C which are predetermined using a set of standards are available as data in the program.

Uranium concentration is further calculated by dividing U238 concentration with weight fraction of U238.

3.4 Data out

U235 content and uranium concentration are printed out. Options are provided for directing the data out to an internal thermal printer or an external dot matrix printer.

4. DISCUSSIONS

The programs have been successfully used for the routine analysis of plutonium product solutions and aged uranium product solutions. The programs make the otherwise tedious data reduction of these techniques simpler and faster for routine analysis of plant samples. Tables 2 and 3 give the results obtained using these techniques.

The programs can be adopted on a personal computer with changes in the print out statements. Microsoft Basic version is also prepared to use with IBM-PC.

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Appendix - 1
Listing of program for plutonium concentration and isotopic calculation by HRGS.

```
10    OPTION BASE 1
20    REM TITLE ---PU/GS
30    REM PROGRAME FOR NDA MEASUREMENT OF Pu ISOTOPICk CONC. BY HRGS
40    REM PREPARED BY A.D.MOORTHY ON 7/07/88
50    PRINTER IS 16
60    DIM E(12),Re(12),I(12),K(5),Sample$[30]
70    MAT READ E(12)
80    DATA 26.34,33.19,38.66,43.48,45.23,51.63,59.054,129.29,144.23,148.57,203.5
          2,208
90    DATA 238.0495,239.0522,240.0540,241.056,242.0587
100   MAT READ K
110   PRINTER IS 16
120   INPUT "DATE ?",Date$
130   INPUT "SAMPLE ?",Sample$
140   INPUT "COUNTING TIME seconds ?",Q
150   C=0
160   REM PROGRAME FOR CALCULATION BY STD. CALIBRATION METHOD
170   INPUT "Enter the area of 38, 43, 45, 51, 59, 129, 148 ",I(1),I(2),I(3),I(4
),I(5),I(6),I(7)
180   Re(7)=1
190   Re(8)=.78342*EXP(.02222*(I(2)/Q))
200   Re(9)=.96171*EXP(.0041657*(I(3)/Q))
210   Re(10)=1.02188*EXP(.007223*(I(4)/Q))
220   Re(3)=1.5104+1.8530E-4*(I(5)/Q)
230   Re(4)=.42945*EXP(.041439*(I(6)/Q))
240   Re(11)=.34454*EXP(.041439*(I(7)/Q))
250   N191=I(7)/I(6)*2.0099E-2*(Re(4)/Re(11))
260   Na9=(I(5)/I(4)*(Re(10)/Re(3))-N191*5.2352E1)*1.3485E-5
270   N891=(I(2)/I(1)*(Re(7)/Re(8))-Na9*3.6917E2-N191*9.3586E-2)*9.7023E-4
280   N892=(I(2)/I(4)*(Re(10)/Re(8))-Na9*1.4357E2-N191*3.6395E-2)*2.4949E-3
290   N89=(N891+N892)/2
300   N091=I(3)/I(4)*(Re(10)/Re(9))*1.6134E-1
310   N092=I(3)/I(1)*(Re(7)/Re(9))*6537*1.05E-4/(24150*4.5E-4)
320   N09=(N091+N092)/2
330   N29=.73*N09*N191
340   T=N89+N09+N191+N29+1
350   Ap(1)=100*N89/T
360   Ap(2)=100/T
370   Ap(3)=100*N09/T
380   Ap(4)=100*N191/T
390   Ap(5)=100*N29/T
400   Ap(6)=Na9*Ap(2)
410   W=0
420   FOR X=1 TO 5
430   O(X)=Ap(X)*K(X)
440   W=W+O(X)
450   NEXT X
460   Av_at_wt=W/100
470   FOR X=1 TO 5
480   Wp(X)=O(X)*100/W
490   NEXT X
500   Wp(6)=Ap(6)*241/Av_at_wt
```

```
510   PRINTER IS 16
520   PRINT "PUT ON THE EPSON PRINTER AND CONT. "
530   BEEP
540   PAUSE
550   PRINTER IS 6,2
560   PRINT TAB(10);"ISOTOPIC COMPOSITION BY HIGH RESOLUTION GAMMA SPECTROMETRY"
570   PRINT TAB(10);"************************************************************"
580   PRINT
590   PRINT TAB(10);"SAMPLE:-";Sample$;TAB(52);"DATE:-";Date$
600   PRINT TAB(10);"METHOD:- Online measurement"
610   PRINT
620   PRINT TAB(15);"ISOTOPE";TAB(36);"ATOM PERCENT";TAB(58);"WEIGHT PERCEN
T"
630   FIXED 5
640   PRINT TAB(15);"Pu-238";TAB(38);Ap(1);TAB(59);Wp(1)
650   PRINT TAB(15);"Pu-239";TAB(36);Ap(2);TAB(57);Wp(2)
660   PRINT TAB(15);"Pu-240";TAB(36);Ap(3);TAB(57);Wp(3)
670   PRINT TAB(15);"Pu-241";TAB(37);Ap(4);TAB(58);Wp(4)
680   PRINT TAB(15);"Pu-242";TAB(38);Ap(5);TAB(59);Wp(5)
690   PRINT TAB(15);"Am-241";TAB(38);Ap(6);TAB(59);Wp(6)
700   PRINT TAB(15);"AVERAGE AT.WT.";TAB(35);Av_at_wt
710   PRINT TAB(15);"-------------------------------------------------------
-------"
720   PRINTER IS 16
730   PRINT
740   PRINT TAB(15);"RATIO OF ISOTOPES";TAB(40);"RATIO OBTAINED"
750   PRINT
760   PRINT TAB(20);"238/239(using 43/38)";TAB(44);NB91
770   PRINT TAB(20);"238/239(using 43/51)";TAB(44);NB92
780   PRINT TAB(20);"238/239(  Average  )";TAB(44);NB9;"*"
790   PRINT TAB(20);"240/239(using 45/51)";TAB(44);N091
800   PRINT TAB(27);"(using 45/38)";TAB(44);N092
810   PRINT TAB(27);"(  average  )";TAB(44);N09;"*"
820   PRINT TAB(20);"241/239(using 148/129)";TAB(44);N191;"*"
830   PRINT TAB(20);"242/239";TAB(44);N29;"*"
840   PRINT
850   PRINT TAB(20);"*-Ratios used for isotopic coposition"
860   PRINT TAB(10);"************************************************************
********"
870   PRINTER IS 6,2
880   ! Programe for concentration measurement -ONLINE
890   Z=0
900   INPUT "CALIBRATION LINEAR orEXPONENTIAL or BOTH",W$
910   IF W$="L" THEN 1140
920   M=5.8677
930   C=7.67895
940   B1=13.14489
950   C1=.0807329
960   B2=1.494255
970   C2=.0883593
980   X=(I(4)/Q-C)/M
990   PRINT X
1000  Con51=I(4)/Q/B1*EXP(C1*X)
```

```
1010  PRINT Con51
1020  T_pu_con51=Con51*100/Wp(2)
1030  X=(I(4)/Q-C)/M
1040  Con129=I(6)/Q/B2*EXP(C2*X)
1050  PRINT Con129
1060  T_pu_con129=Con129*100/Wp(2)
1070  Av_con=(Con51+Con129)/2
1080  Av_t_pu_con=Av_con*100/Wp(2)
1090  Pu_con=Av_t_pu_con/W
1100  PRINTER IS 6,2
1110  PRINT "","EXPONENTIAL METHOD"
1120  PRINT "","------------------"
1130  GOTO 1630
1140  Z=Z+1
1150  IF Z=2 THEN 1830
1160  Cr4=I(4)/Q
1170  IF Cr4<7.85 THEN 1270
1180  IF Cr4<23.82 THEN 1330
1190  IF Cr4<34.51 THEN 1390
1200  IF Cr4<50.12 THEN 1450
1210  PRINT 5
1220  M51=3.85047
1230  C51=24.6452
1240  M129=.473886
1250  C129=2.23867
1260  GOTO 1500
1270  PRINT 1
1280  M51=10.0639
1290  C51=0
1300  M129=1.02986
1310  C129=0
1320  GOTO 1500
1330  PRINT 2
1340  M51=8.8054
1350  C51=.98214
1360  M129=.93861
1370  C129=.07121
1380  GOTO 1500
1390  PRINT 3
1400  M51=7.69674
1410  C51=3.8572
1420  M129=.75897
1430  C129=.53707
1440  GOTO 1500
1450  PRINT 4
1460  M51=5.9280
1470  C51=10.9001
1480  M129=.68893
1490  C129=.81597
1500  U=I(4)/Q
```

```
1510  V=I(6)/Q
1520  PRINT U,V
1530  Con51=(U-C51)/M51
1540  Con129=(V-C129)/M129
1550  T_pu_con51=Con51*100/Wp(2)
1560  T_pu_con129=Con129*100/Wp(2)
1570  Av_con=(Con51+Con129)/2
1580  Av_t_pu_con=Av_con*100/Wp(2)
1590  PRINTER IS 6,2
1600  PRINT " ","LINEAR METHOD"
1610  PRINT " ","-------------"
1620  Print=2
1630  PRINTER IS 6,2
1640  PRINT TAB(23);"ONLINE MEASUREMENT OF PLUTONIUM CONCENTRATION"
1650  PRINT TAB(23);"-----------------------------------------"
1660  STANDARD
1670  PRINT TAB(15);"SAMPLE:-";Sample$;TAB(59);"DATE:-";Date$
1680  PRINT TAB(15);"COUNT TIME:-";Q;TAB(59);"COUNT NO.:-";C
1690  PRINT
1700  FIXED 5
1710  PRINT TAB(32);"PLUTONIUM CONCENTRATION OF THE SAMPLE(G/L)"
1720  PRINT TAB(30);"(Using51)    (Using129)      ( Average )"
1730  PRINT
1740  PRINT TAB(15);"Pu-239";TAB(30);Con51;TAB(43);Con129;TAB(59);Av_con
1750  PRINT TAB(15);"TOTAL Pu";TAB(30);T_pu_con51;TAB(43);T_pu_con129;TAB(59);Av
_t_pu_con
1760  PRINT TAB(15);"-------------------------------------------------------
"
1770  IF Print=2 THEN 1790
1780  IF W$="B" THEN 1140
1790  C=C+1
1800  IF C=2 THEN 1830
1810  GOTO 160
1820  PRINTER IS 16
1830  END
```

Appendix-2

A typical out put for isotopic composition and plutonium concentration.

ISOTOPIC COMPOSITION BY HIGH RESOLUTION GAMMA SPECTROMETRY.
****

SAMPLE :- 293 Pu Lab Date:- 8/9/88
METHOD :- Online measurement

ISOTOPE	ATOM PERCENT	WEIGHT PERCENT
Pu-238	.09353	.09302
Pu-239	73.90958	73.81292
Pu-240	21.44993	21.51166
Pu-241	3.75205	3.77855
Pu-242	.79491	.80385
Am-241	.00303	.00305
AVERAGE AT. WT.	239.36523	

LINEAR METHOD

ONLINE MEASUREMENT OF PLUTONIUM CONCENTRATION

SAMPLE :- 293 Pu Lab Date:- 8/9/88
COUNT TIME:- 1800 COUNT NO.:-1

	PLUTONIUM CONCENTRATION OF THE SAMPLE(G/L)		
	(Using 51)	(Using 129)	(Average)
Pu-239	9.01588	9.26920	9.14254
TOTAL Pu	12.21451	12.55769	12.38610

Appendix - 3

Listing of program for uranium concentration by on-line HRGS.

```
10    REM TITLE UONLINE
20    REM PREPARED BY A.D.MOORTHY ON 22JANUARY 1987
30    INPUT "Enter the date of analysis & CONT.",Date$
40    INPUT "Enter the sample ident. & CONT. ",S$
50    INPUT "Enter the counting time in secods & CONT.",Q
60    INPUT "Enter the area of 63, 163, 185, 1001 Kev peaks & CONT. ",I(1),I(2
),I(3),I(4)
70    FOR X=1 TO 4
80    Cr(X)=I(X)*3600/Q
90    NEXT X
100   E(1)=2.08215E-3-2.5262E-8*Cr(1)
110   E(2)=4.46126E-3-1.66169E-7*Cr(2)
120   E(3)=4.58271E-2-1.1970E-7*Cr(3)
130   E(4)=1.0717E-4
140   U581=Cr(3)/Cr(1)*E(1)/E(3)*2.604/16.47
150   U48=.00005
160   T1=1+U581+U48
170   Af81=1/T1
180   U582=Cr(3)/Cr(4)*E(4)/E(3)*2.604/16.47
190   T2=1+U582+U48
200   Af82=1/T2
210   U583=Cr(2)/Cr(1)*E(1)/E(2)*2.604/16.47
220   T3=1+U583+U48
230   Af83=1/T3
240   U584=Cr(2)/Cr(4)*E(4)/E(2)*2.604/16.47
250   T4=1+U584+U48
260   Af84=1/T4
270   ! CONC. CALCULATION USING 63 KEV PEAK
280   A=19.73639
290   B=5.31138E-5
300   C863=A*EXP(B*Cr(1))
310   A2=5.05753E-7
320   A1=.0393837
330   A0=3.68828
340   C81001=A2*Cr(4)^2+A1*Cr(4)+A0
350   Cu1=C863/Af81
360   Cu2=C81001/Af82
370   Cu3=C863/Af83
380   Cu4=C81001/Af84
390   PRINTER IS 6,2
400   BEEP
410   DISP "PUT ON THE EPSON PRINTER & CONT. "
420   PAUSE
```

```
430   PRINT "              URANIUM BY GAMMA SPECTROMETRY"
440   PRINT "              ----------------------------"
450   PRINT "DETECTOR: Coaxial                                    DATE:";Date$
460   PRINT "SAMPLE  :";S$;"                                COUNT TIME(sec.):";Q
470   PRINT "..............................................................
*
480   PRINT
490   PRINT "63Kev (area)     163Kev (area)    185.72Kev (area)   1001Kev (area)
*
500   PRINT TAB(4);I(1);TAB(22);I(2);TAB(40);I(3);TAB(59);I(4)
510   PRINT "..............................................................
*
511   FIXED 4
520   PRINT "CONC. OF URANIUM 238(Mg/ml) USING 63 Kev   :";C863
530   PRINT "CONC. OF URANIUM 238(Mg/ml) USING 1001 Kev :";C81001
531   STANDARD
540   IMAGE "235/238 RATIO              :",Z.DDDDDD
550   IMAGE "ATOM FRACTION ON 238       :",Z.DDDDD
560   IMAGE "TOTAL URANIM CONC. (Mg/ml) :",DDD.DDD
570   PRINT
580   PRINT "PEAKS USED 63 Kev & 187.5 Kev"
590   PRINT "-----------------------------"
600   PRINT USING 540;U581
610   PRINT USING 550;Af81
620   PRINT USING 560;Cu1
630   PRINT
640   PRINT "PEAKS USED 1001 Kev & 187.5 Kev"
650   PRINT "------------------------------"
660   PRINT USING 540;U582
670   PRINT USING 550;Af82
680   PRINT USING 560;Cu2
690   PRINT
700   PRINT "PEAKS USED  63 Kev &  163 Kev"
710   PRINT "-----------------------------"
720   PRINT USING 540;U583
730   PRINT USING 550;Af83
740   PRINT USING 560;Cu3
750   PRINT
760   PRINT "PEAKS USED  1001 Kev &  163 Kev"
770   PRINT "------------------------------"
780   PRINT USING 540;U584
790   PRINT USING 550;Af84
800   PRINT USING 560;Cu4
810   PRINT
820   PRINT "..............................................................
*
830   DISP "CALCULATIONS DONE. IF YOU WANT TO REUSE JUST CONT "
840   END
```

Table 1.

Gamma ray used for high resolution gamma spectrometry

Isotope	Energy of the peak (Kev)	Branching intensity	Interference.
Pu 238	43	3.93E-04	Pu-241, Am-241
239	38	1.05E-04	
	51	2.7 E-04	
	129	6.26E-05	
240	45	4.53E-04	
241	145	1.87E-06	
Am-241	59	5.24E+01	

Table 2.

Plutonium concentration by HRGS-NDA method.

S. No.	Sample No.	Tank	Pu Concentration(mg/g)		B
			Coulometry (A)	HRGS (B)	A
1.	4	Audit	7.267	7.343 +/-0.068	1.0105
2.	606173	Pus	11.823	11.779 +/-0.037	0.9963
3.	606261	Pus	10.477	10.644 +/-0.259	1.0159
4.	606271	Pus	10.710	10.429 +/-0.036	0.9738
5.	606712	Pus	11.267	11.267 +/-0.211	1.0000
				Average	0.9993
				Std. dev. +/-	0.0163

Table 3.

Uranium concentration by Gamma ray spectrometry and Potentiometry.

S.No.	Sample No.	Concentration (mg/ml)		A / B	U235/U238 ratio by HRGS
		HRGS (A)	Potentiometry (B)		
1.	STD-5	281.53 +/- 0.43	283.53	0.9929	0.00769 +/-0.00003
2.	S-1	206.69 +/- 0.36	208.44	0.9916	0.00483 +/-0.00003
3.	S-2	257.61 +/- 3.77	266.13	0.9680	0.00504 +/-0.00012

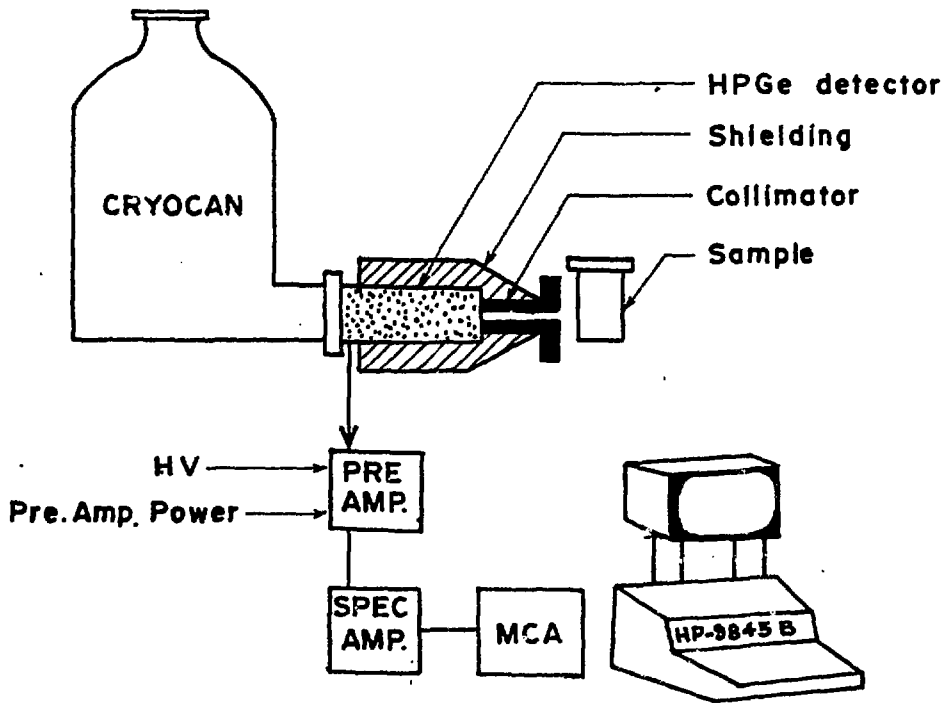


Fig.1. Set up used for Pu measurement.

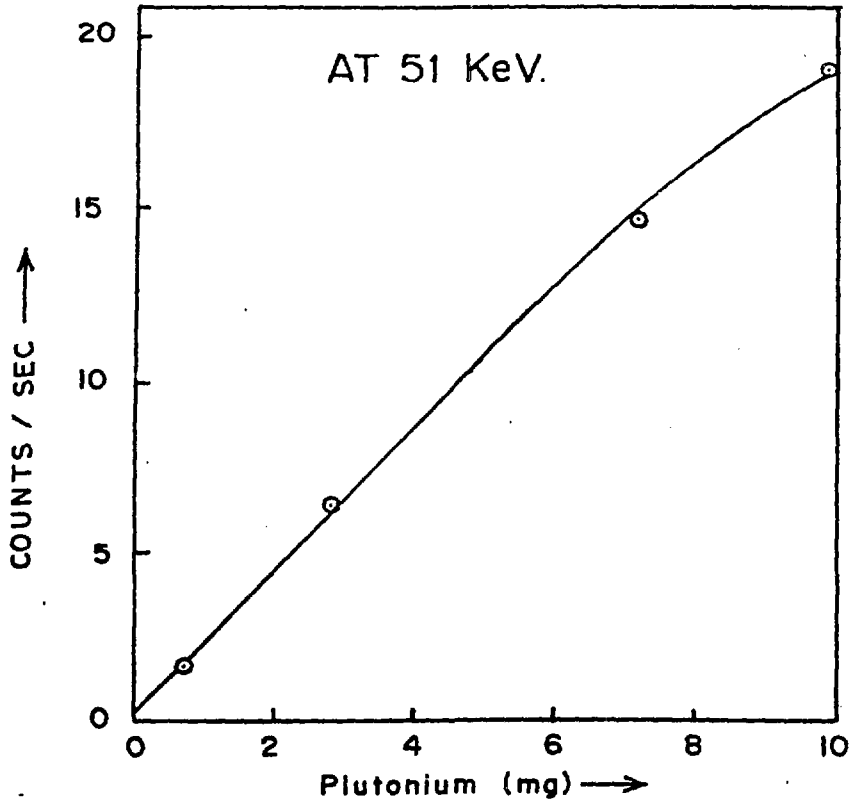
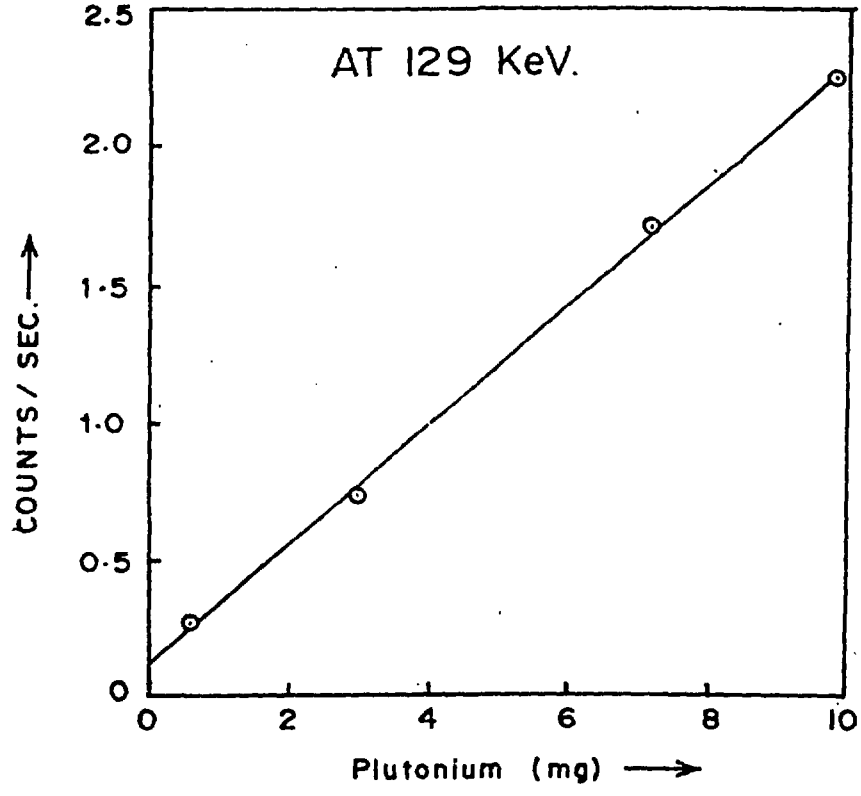


Fig. 2. Plutonium concentration vs. count rate.



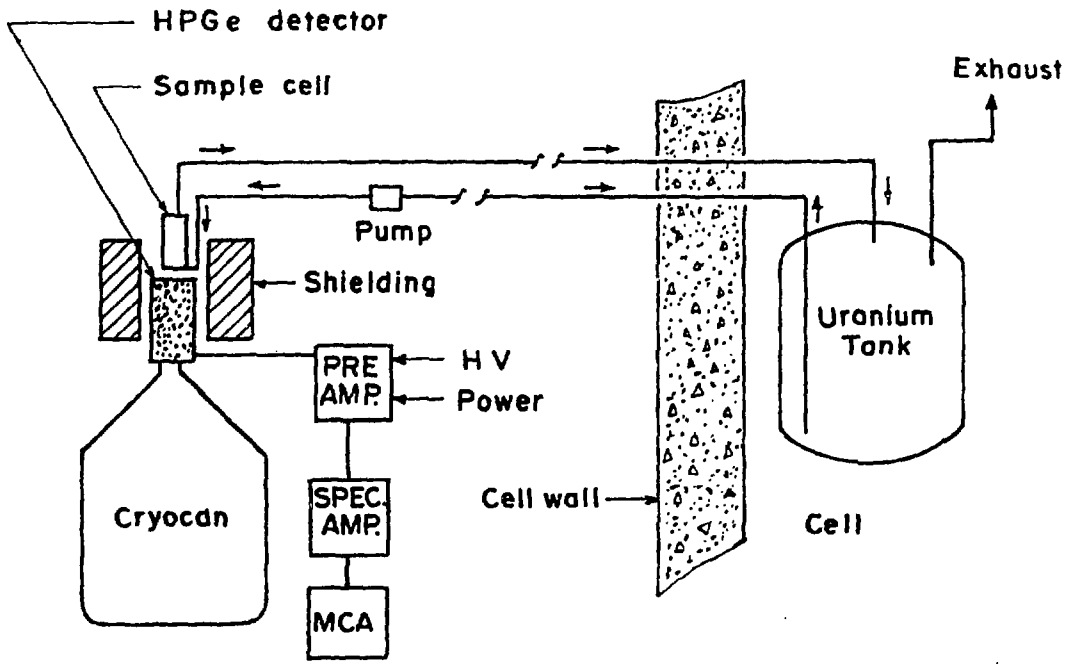


Fig. 4. Setup for Uranium online measurement.

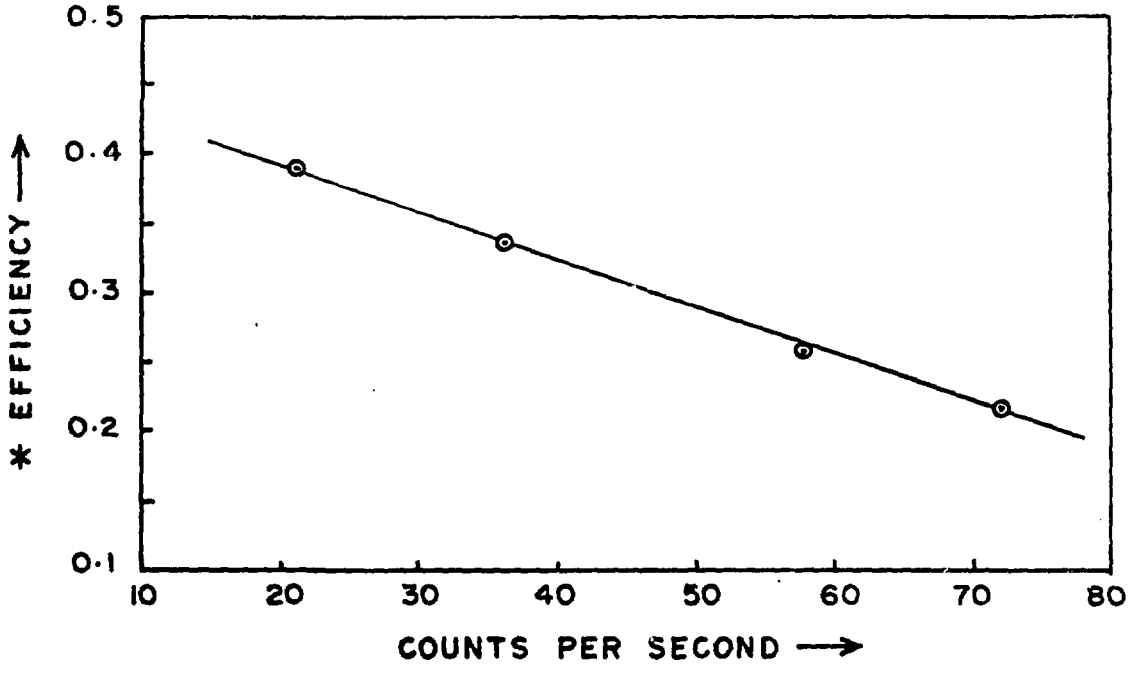


Fig. 5. Efficiency vs. count rate at 185.7 KeV.

$$* \text{ Efficiency} = \frac{\text{Observed count rate}}{\text{Theoretical count rate}} \quad \text{(at 185.7 KeV.)}$$

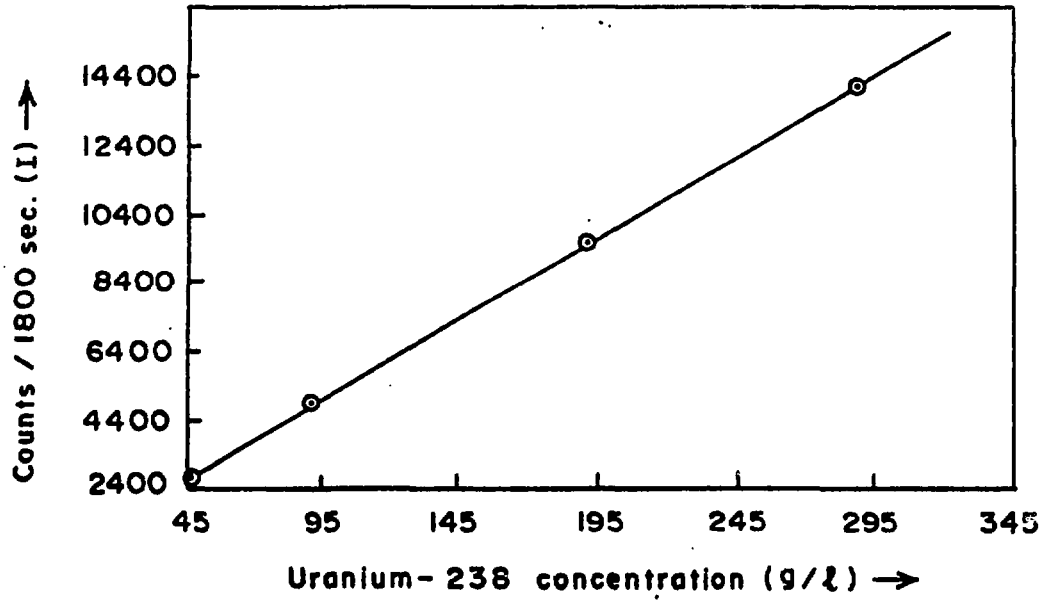


Fig. 6. Uranium concentration calibration curve using 1001 KeV peak.

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