Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

LA-UR--85-2736

DE85 015730

TITLE: MEASUREMENT OF THE ENRICHMENT OF URANIUM IN THE PIPEWORK OF A GAS CENTRIFUGE ENRICHMENT PLANT

AUTHOR(S): T. W. Packer and E. W. Lees
Nuclear Physics Division, AERE, Harwell, Oxon. OX11 ORA, U.K.

D. Close, K. V. Nixon, J. C. Pratt, and K. Strittmatter
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, U.S.A.

SUBMITTED TO: 26th Annual Meeting, Institute of Nuclear Materials Management
The Regent Hotel, Albuquerque, New Mexico
July 22-25, 1985

DISCLAIMER

MEASUREMENT OF THE ENRICHMENT OF URANIUM IN THE PIPEWORK OF A GAS CENTRIFUGE ENRICHMENT PLANT

T. W. Packer and E. W. Lees

Nuclear Physics Division, AERE, Harwell, Oxon. OX11 ORA, U.K.

D. Close, K. V. Nixon, J. C. Pratt and R. Strittmatter

Los Alamos National Laboratory, Los Alamos, New Mexico 87545, U.S.A.

ABSTRACT

The US and UK have been separately working on the development of a NDA instrument to determine the enrichment of gaseous UF_6 at low pressures in cascade header pipework in line with the conclusions of the Hexapartite Safeguards Project viz. the instrument is capable of making a "go/no go" decision of whether the enrichment is less than/greater than 20%. Recently, there has been a series of very useful technical exchanges of ideas and information between the two countries. This has led to a technical formulation for such an instrumentation based on γ-ray spectrometry which, although plant-specific in certain features, nevertheless is based on the same physical principles. Experimental results from commercially operating enrichment plants are very encouraging and indicate that a complete measurement including set up time on the pipe should be attainable in about 30 minutes when measuring pipes of diameter around 110 mm.

1. INTRODUCTION

In November 1980 the Hexapartite Safeguards Project comprising the operators, the IAEA and Euratom was launched in an attempt to find an acceptable solution to the problem of safeguarding centrifuge enrichment plants. It soon became apparent that a NDA instrument which could detect the presence, or confirm the absence, of HEU (over 20% enriched) UF_6 gas on a rapid GO, NO-GO basis in cascade header pipework would be of great benefit in attaining the safeguard goals. Any NDA instrument would have to meet both the constraints of the operators (viz. the operating gas pressure is proprietal information and the measurement should not interfere with normal operation of the plant) and also those of the Inspectorates (viz. the measurements should not be over complicated or time consuming since they have a large number of pipes to sample in a relatively short time). The US and UK have been separately working on such an instrument using γ-ray spectrometric techniques over the past few years but recently, technical progress has been much improved by a frank and full exchange of experimental results.

In principle, the total mass of ^{235}U in the gas can be determined by measuring the intensity of the 186 keV γ-rays resulting from the decay of ^{238}U nuclei in the gas. The total mass of U in the gas is then determined by energy-dispersive X-ray fluorescence (XRF) analysis. The enrichment (after suitable calibration) can simply be given by the ratio of these two measurements. In practice, the development of such an instrument has been complicated by the complex behaviour of the U deposits (mainly from reactions of UF_6 gas with water vapour) and direct Th deposits (resulting from the α-decay of the UF_6 gas) which occur on the walls of the pipes in these centrifuge plants. The intensity of γ-rays emitted by the deposits relative to the corresponding γ-rays emitted by the gas, varies from plant to plant and even within the same plant at equivalent locations; in the UK, the ratio is typically in the range 2 to 12 whereas in the US this ratio is likely to be ~1.

Thus the most recent work by both countries has concentrated on two aspects:

(i) correcting the measured 186 keV intensity for the contribution arising from the U deposit on the wall,

(ii) developing an XRF technique which will maximise the gas signal without detecting any X-rays from the U deposit on the walls.

The results of the XRF studies have been reported elsewhere[1,2] and so this paper will concentrate on the deposit correction methods and the measured enrichments obtained after combining these with the XRF measurements.

2. TECHNIQUE

All methods of determining the enrichment of the UF_6 gas in the pipe are based on the number of the 185.72 keV γ-rays emitted from the gas, which is proportional to the enrichment of the gas x its pressure. (Note that this dependence on the gas pressure rules out the use of a purely passive γ-ray count since the signals from 5% enriched gas at normal pressure and from 20% HEU gas at quarter pressure are identical.) As 185.72 keV γ-rays are also emitted from the deposited uranium, it is necessary to establish the number emitted by the UF_6 gas from the total value.

The γ-rays of interest to the present work which are emitted from the decay chains of ^{235}U and ^{238}U are shown in Table 1. Since the decay of ^{238}U is not accompanied by any significant γ-ray emission, then the 63.29 keV γ-rays emitted by ^{234}Th are used as an indication of either the mass of deposited uranium on the pipe or from directly deposited thorium[3,4]. The former occurs due to chemical reaction of UF_6 gas with the aluminium pipe and with any water vapour and volatile impurities that may be present in the pipe. The latter results from deposition of thorium due to the decay of uranium nuclei as the UF_6 gas passes through the plant. The thorium is formed as a non-volatile fluoride which will be either deposited locally or carried along as a suspended aerosol.

Isotope	Energy of γ-ray (keV)	Abundance (%)	Emitted from: Gas	Deposited uranium	Directly deposited thorium
^{235}U	89.95	1.50	Yes	Yes	No
	93.35	2.50			
	185.72	54.00			
^{234}Th	63.29	3.90	No	Yes	Yes
	92.38	2.57			
	92.80	3.00			
^{231}Th	84.21	8.00	No	Yes	Yes
	89.95	1.25			
	92.29	0.43			
	95.87	0.76			

Table 1 Energies and abundances of the principal γ-rays emitted from UF_6 gas, deposited uranium and directly deposited thorium

Two techniques have been developed for separating the 186 keV γ-rays originating from the gas and from the deposit. The first method is a deposit correction method based on the 63 keV γ-ray intensity and the second is a two geometry technique based on varying the geometrical detection efficiencies for γ-rays emitted from the deposit and the gas.

2.1 DEPOSIT CORRECTION TECHNIQUE

The technical exchanges resulted in refinements of the deposit correction technique and the jointly favoured approach is to use the intensities of the 186 keV γ-ray (^{235}U) and the ^{238}U decay chain γ-ray at 63 keV (^{234}Th). From the discussion above, it follows that:

$$186_{TOT} = 186_{DEP} + 186_{GAS}$$

and

$$63_{TOT} = 63_{DEP} + 63_{TH}$$

where 186_{TOT} and 63_{TOT} refers to the total 185.72 and 63.29 keV γ-ray countrates respectively and DEP, GAS and TH refer to the deposited uranium, UF_6 gas and directly deposited thorium contributions. If one assumes radioactive decay equilibrium, each decay of a uranium gas atom produces a thorium atom which is very likely to be locally deposited on the pipework, where its deposit will be matched by the decay of a directly deposited thorium atom. This process is referred to as 100% direct thorium deposition[3]. Thus neglecting any difference in geometrical detection efficiency between γ-rays emitted from the gas and deposit, which have been shown to be small for pipe diameters between 30 and 168 mm, it can be assumed that under normal operation

$$186_{GAS} = KE_G\ 63_{TH}$$

where K is a constant depending on the relative detection efficiencies for 185.72 keV and 63.29 keV γ-rays and E_G is the enrichment of the gas. It also follows that in secular equilibrium,

$$186_{DEP} = KE_D\ 63_{DEP}$$

where E_D is the enrichment of the deposited uranium. Under normal operating conditions it has been found that $E_D \sim E_G$.

Hence a measure of abnormal operation of the plant, i.e. enrichment of UF_6 gas higher than the declared value, can be investigated by measuring the excess 186 counts, X_{186}, where

$$X_{186} = 186_{TOT} - KE63_{TOT}$$

where E is now the enrichment of the gas as declared by the operator to the inspector.

In practice even under normal operational conditions X_{186} will not always be zero, in fact it can be positive or negative, due to differences between E_D and E and the fact that the direct thorium deposit is not always 100%. Measured values of ~60% to 90% have been obtained at Capenhurst.

The UF_6 gas pressure can also be indirectly measured by energy-dispersive X-ray fluorescence analysis by comparing the number of detected UK X-rays excited in the UF_6 gas, U_K say, with those obtained from a secondary standard gas sample.

The pressure of the UF_6 gas is given by CUK where C is a calibration constant which need only be made available on a need to know basis in order to conserve the restricted nature of the absolute gas pressure.

Hence the excess enrichment E_X of the UF_6 gas in the pipe at the time of measurement can be calculated from

$$E_X = \frac{X_{186}}{CU_K} = \frac{186_{TOT} - KE63_{TOT}}{CU_K}.$$

In practice the biggest statistical error on the measurement is in the measurement of 63_{TOT}. The overall error in the measurement can be reduced by approximately $\sqrt{2}$ by simultaneously measuring the number of 92.38 and 92.80 keV γ-rays emitted by ^{234}Th and using a different value of K. However a correction for unresolved 93.35 keV γ-rays emitted by ^{238}U and 92.29 keV γ-rays emitted by ^{231}Th is necessary. Although this complication is not justified for the present work, it may be useful in reducing the measurement time required for smaller diameter pipes in areas where total measurement time is restricted.

2.2 TWO GEOMETRY TECHNIQUE

During the X-ray fluorescence measurement, the spectrum shows that the 185.72 keV γ-ray from ^{238}U remains visible above the energy of the ^{57}Co 122 keV excitation source. The countrate from the 185.72 keV γ-ray obtained by this measurement is lower than that obtained from a simple passive measurement using a wider collimator. Both measurements of the 185.72 γ-ray have contributions from both deposit and gas. However, the proportions counted from these two sources are different for the narrow/XRF collimation than from those proportions in the "wide-open"/passive measurement. This suggests that from two measurements with two geometries the deposit and gas contributions could be isolated after the different geometrical detection efficiencies had been established. The geometrical acceptances for the narrow collimator (Phase 2), for both gas and deposit ^{238}U, are smaller than those of the open-collimator measurements (Phase 1). A series of laboratory measurements (acquisition of spectra with each collimator geometry on deposit-bearing pipes, for these pipes with and without gas) completely determines the four geometrical acceptance factors. Thus,

$$M1 = A(1,1) * G + A(1,2) * D \qquad (1a)$$

$$M2 = A(2,1) * G + A(2,2) * D \qquad (1b)$$

Where:

M1	= Measured 185.72 countrate of Phase 1
M2	= Measured 185.72 countrate of Phase 2
G	= Quantity of ^{235}U in gas phase
D	= Quantity of ^{235}U in wall deposit
A(i,j)	= 2X2 matrix of geometrical acceptance factors

The above equations are solved for the deposit and gas concentrations.

The ^{235}U signal from the gas, obtained by the above two-collimator technique, forms the numerator of the enrichment calculation. This method does not need any information about the γ-rays of the ^{238}U daughters. The geometry for both the Phase 1 and Phase 2 measurements must be carefully controlled to allow this interpretation method, but no other assumptions are required. The ratio nature of this calculation means that an automated measurement process can calculate the enrichment without explicit knowledge of the pressure.

3. EQUIPMENT

Either a high resolution gamma-X semi-conductor detector or a planar Ge detector can be used for both the γ-ray and X-ray fluorescence measurements as they both have high efficiencies for detecting 63.29 keV γ-rays.

The Harwell shielding consists of 3 mm lead to attenuate the 185.72 keV γ-rays, with an inner shielding of 6 mm iron to absorb excited Pb K X-rays which interfere with the 84.21 keV reading (see Ref. [2]). The US shielding is similar with 3 mm tungsten, lined with 2 mm copper.

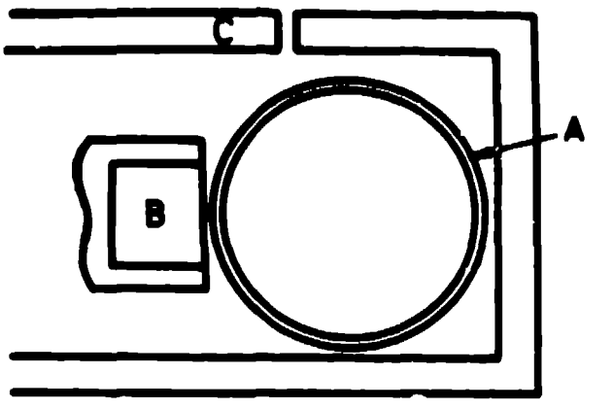
The geometrical arrangement which gives the highest detection efficiency for γ-ray spectrometry measurements is shown schematically in Fig. 1(a). This geometry can be easily modified, as shown in Fig. 1(b) to enable UK X-rays to be excited in the UF_6 gas without exciting UK X-rays in the deposited uranium. A 10 mCi ^{57}Co source is used to excite the characteristic X-ray of U.

Calibration standards have been produced to enable rapid verification of the NDA instruments. For the passive γ-ray measurements, these consist of U samples ($\sim$ 1 mg/cm^2) painted on the inside of aluminium pipes of the same type as used in the centrifuge plants; for the XRF measurements a thin foil of U was mounted at the centre of an aluminium pipe.

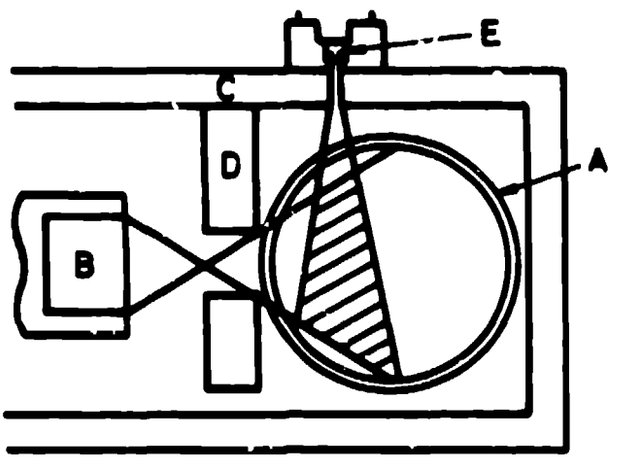
4. EXPERIMENTAL RESULTS

4.1 U.S. RESULTS, LABORATORY ENRICHMENT MEASUREMENTS

A laboratory proof-of-principle was established that the ratio of the ^{238}U gas-only signal to the total uranium gas as measured by XRF is directly proportional to the enrichment of the gas and independent of the pressure. The experimentally measured ^{238}U gas-only countrate was determined by measuring the cylinder with gas in it and then evacuating the cylinder to measure only the deposit. The ^{238}U gas-only countrate is the difference between these two measurements. As is shown in Table 2 the experimentally determined ^{238}U enrichment and the actual ^{238}U enrichment are in very good agreement. Thus a ratio of the ^{238}U gas-only to the total uranium gas-only signal does



a) γ-ray spectrometer



(b) X-ray fluorescence

A Sample B Detector C Shielding
D Collimator E Source

Fig. 1 Geometries used for the two measurements

Pressure (torr)	Enrichment (%)	Calculated Enrichment (%)
28	20.79	20.89 +/- 0.41
16	20.79	23.93 +/- 1.05
9	20.79	22.77 +/- 1.08
1	20.79	19.16 +/- 4.28
15	5.36	5.34 +/- 0.13
10	5.36	5.25 +/- 0.17
5	5.36	5.19 +/- 0.30
13	0.71	0.75 +/- 0.04
8	0.71	0.71 +/- 0.05
3	0.71	0.73 +/- 0.10
10	3.00	2.82 +/- 0.10
7	3.00	2.98 +/- 0.18
4	3.00	2.65 +/- 0.43
10	7.92	8.21 +/- 0.43
10	1.00	0.98 +/- 0.06

Table 2 Comparison of actual and calculated ^{235}U enrichment for proof-of-principle measurements on a 4" nominal ID pipe

indeed yield a quantity which is proportional to the enrichment of the gas and furthermore, such a measurement is independent of the gas pressure.

The next logical step was to use the formalism of equations 1a and 1b above to correct for the deposit contribution to the total ^{235}U signal without evacuating the cylinder. Six data points (labelled (a) in Table 3) of the calculated gas-only ^{235}U signal were used to obtain an enrichment calibration curve; the calculated enrichment from the fitted calibration curve is statistically the same as the stated enrichment for these 6 points. The lower part of Table 3 lists the calculated ^{235}U enrichment for the rest of the data. There is generally good agreement between actual enrichment and calculated enrichment except at low pressures. It is at low pressures and low enrichment that the deposit-to-gas ratio is rather large (>5), and necessitates large deposit corrections to the data.

Pressure (torr)	Enrichment (%)	Calculated Enrichment (%)
28	20.79 (a)	21.03 +/- 0.84
10	7.92 (a)	7.47 +/- 0.75
15	5.36 (a)	5.69 +/- 0.36
10	3.00 (a)	2.06 +/- 0.33
10	1.00 (a)	0.59 +/- 0.43
13	0.71 (a)	1.18 +/- 0.27
16	20.79	24.32 +/- 1.44
9	20.79	26.48 +/- 1.64
1	20.79	76.40 +/- 17.67
10	5.36	5.26 +/- 0.41
5	5.36	7.11 +/- 0.72
7	3.00	0.50 +/- 0.57
4	3.00	1.38 +/- 0.64
8	0.71	1.61 +/- 0.33
3	0.71	6.73 +/- 1.07
20	20.79	19.52 +/- 2.53
10	20.79	24.78 +/- 4.56
5	20.79	19.50 +/- 7.92
20	2.82	2.10 +/- 1.81
10	2.82	6.77 +/- 3.08

(a) Self-consistency check, used to obtain enrichment calibration parameters

Table 3 Comparison of actual and calculated ^{235}U enrichment, using calculated gas-only ^{238}U countrates. The measurement times ranged from 20 minutes to 4 hours

The studies of the direct Th deposition have shown that the daughters from gas-phase uranium decay may be deposited on the pipe walls in a non-uniform fashion dependent on pipe geometry. An example of this nonuniform deposit is shown in figure 2 for the Oak Ridge Centrifuge Production Demonstration Facility (CPDF) where there was a 30-meter straight pipe which was followed by a vertical bend. This shows the γ-ray countrates (after evacuation of the pipe) from ^{238}U and the daughters of ^{238}U in the vicinity of this bend and indicates increased direct Th deposition close to the bend. Thus, the measuring locations must be carefully selected for any verification technique

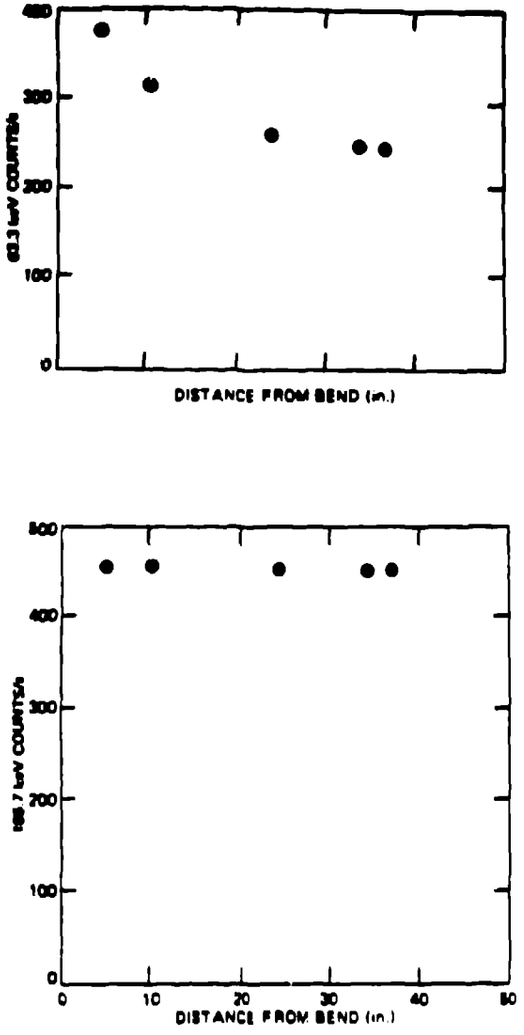


Fig. 2 CPDF product pipe has a 30m straight section, followed by a bend. The figure shows the 63.3 keV (^{234}Th) and 185.72 keV (^{235}U) photopeak countrates as a function of position after the bend. The units shown are arbitrary.

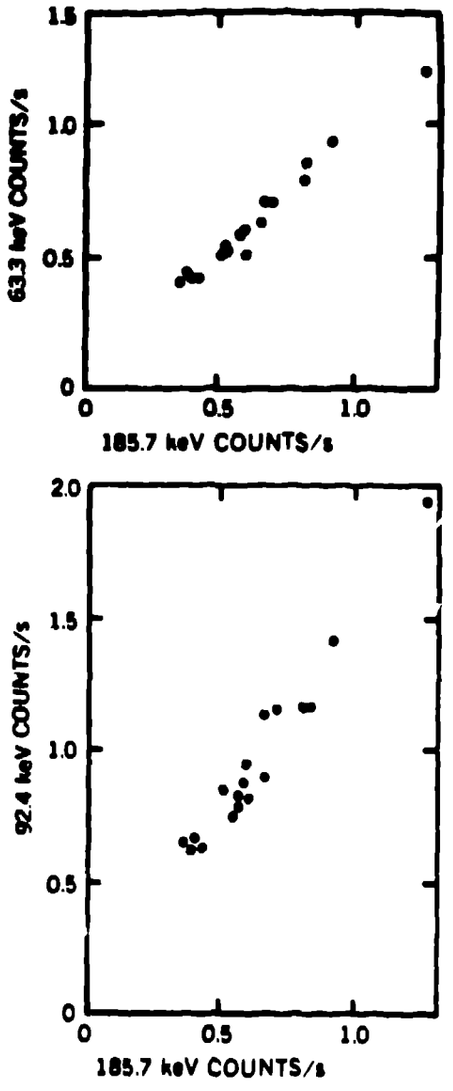


Fig. 3 Centrifuge Test Facility (CTF) product pipe deposit photopeak countrates for 185.72 keV (^{235}U), 63.3 keV (^{234}Th), and 92.4 keV (^{234}Th). Units shown are counts per second. Error bars of approximately 5% are omitted for clarity

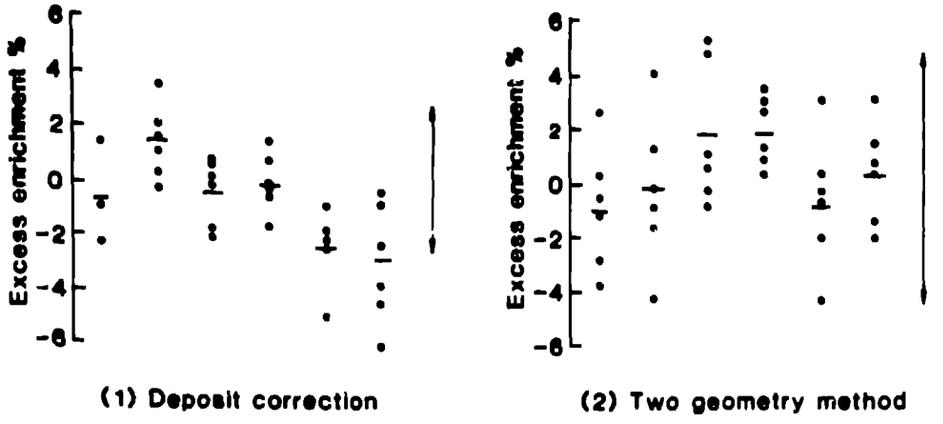
which uses these daughter γ-ray countrates as an indication of ^{238}U amount.

However, figure 3 shows the relationship between the ^{235}U signal and ^{238}U daughter signals from many measurements along a pipe in the Oak Ridge Centrifuge Test Facility (CTF) after the facility was shut down. These observations indicate that the ratio of ^{235}U signals to those from ^{238}U daughters can be stable. It should be noted that this stable relationship could be upset by radioactive disequilibrium introduced by recent U deposition and variable direct Th deposition and practical experiences on these points is discussed in the next section.

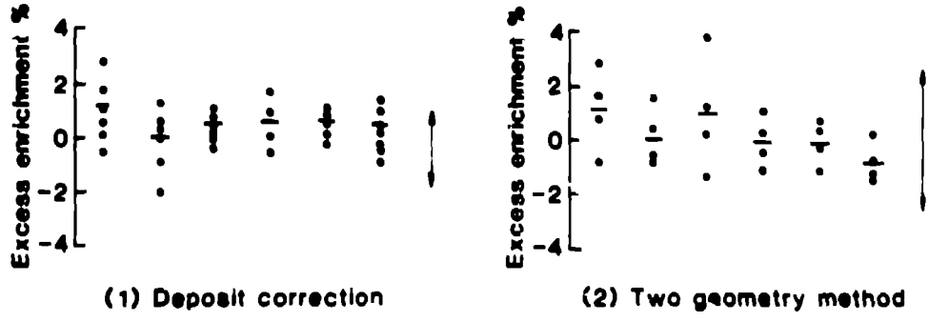
4.2 CAPENHURST PLANT RESULTS

Repeat measurements of 600 seconds for both the γ-ray spectrometry and X-ray fluorescence analysis were made on twelve pipes in two plants at Capenhurst over the period October 1984 to March 1985. The X-ray fluorescence measurements were corrected to allow for the decay of the ^{57}Co source. The detailed results of measurements made on pipe 2, which were made in the presence of members of the IAEA Development Division, are given in reference 5. The estimated excess enrichments calculated using the deposit correction technique as described in Section 2.1 are shown in figure 4. Also shown in this figure are the results for the enrichment deduced from the two geometry technique for 12 pipes in the same two plants which required an additional γ-spectroscopy measurement of 3600 seconds for the narrow/XRF collimation after the ^{57}Co source had been removed.

For the deposit correction results from plant A, there appears to be a systematic underestimate



(a) Plant A



(b) Plant B

Fig. 4 Excess enrichment for 12 cascades in 2 plants at Capenhurst as deduced by the deposit correction and two geometry techniques. The dots represent the individual measurements on each cascade and the horizontal bars represent the mean value for each cascade. The arrows represent the typical statistical uncertainty in the excess enrichment at the 95% confidence limit

of the excess enrichment of ~3% for the last 2 pipes studied; this is due to the enrichment of the deposited uranium being much lower (~ 2%) than the current gas enrichment of 3.2%. For plant B, the statistical precision is improved (1.5%) relative to plant A (2.7%) mainly because of the lower U deposits and there is evidence of a slight positive bias in the results of ~0.5% for the deduced excess enrichment; this is due to the direct thorium deposition in this plant only being approximately 60 to 70%. All 69 measurements of the excess enrichment lie in the range -6.2 to +3.5% and would enable an inspector to confidently confirm that none of the pipes contained HEU at the time of measurement. If 20% enriched U had been present, values in the range of 11 to 23% would have been expected, and a more accurate assessment of the UF_6 gas enrichment would normally be obtained by making longer measurements or by using the 2-geometry technique. It is equally important to note that although the measurement is subject to differences between the declared enrichment of the UF_6 gas and the deposited uranium, the errors introduced into the estimation of the enrichment of the gas are considered very unlikely to give false HEU readings or enable an operator to conceal the presence of HEU in the pipework.

As expected, the results for the 2-geometry technique show no statistically significant bias for either plant. The statistical precision (95% confidence limits) is typically 4.7% for plant A and 2.7% for plant B; again the difference reflects the larger deposits in plant A. The 61 results lie in the range -5.1 to +6.3% and would enable an inspector to confirm that none of the pipes contained HEU at the time of measurement. The sole disadvantage of the 2 geometry technique is that it requires a much longer measurement time (over a factor of 10) to obtain the equivalent statistical precision of the deposit correction technique.

5. CONCLUSIONS

The US and UK have been separately working on the development of an NDA instrument to determine whether the enrichment of UF_6 in the pipework of a centrifuge enrichment plant is less than or greater than 20%. The primary objective of this instrument is to measure the enrichment of gaseous UF_6 in the pipework at the time of the measurement; and in line with the conclusions of the Hexapartite Safeguards Project provide a "go/no go" decision whether the enrichment is in the LEU range while not divulging information which is of particular sensitivity to the facility operator. To facilitate the development of the measurement techniques there have been a series of very useful technical exchanges of ideas and information between the two countries. This has led to a technical formulation for such an instrument based on an integration of passive γ-ray spectrometry and X-ray fluorescence which, although plant-specific in certain features, is based on the same physical principles. The measurement techniques have been demonstrated at commercially operating enrichment plants and laboratory test loops to be applicable over a wide range of plant conditions. The experience obtained indicate that a complete measurement including set up time on the pipe should be attainable in about 30 minutes when measuring pipes of diameter approximately 110 mm.

6. ACKNOWLEDGEMENTS

The authors wish to thank Mr. D. Lester, Mr. D. Pook and the operations staff at Capenhurst for their help in making the measurements.

7. REFERENCES

1. D. A. Close, J. C. Pratt, J. J. Malanify and H. F. Atwater, "X-ray Fluorescent Determination of Uranium in the Gaseous Phase", Nuclear Instruments and Methods in Physics Research A234, 1985, pp556-561.

2. T. W Packer and E. W. Lees, Measurement of the Enrichment of UF_6 gas in the pipework of a gas centrifuge plant. 7th ESARDA Symposium, Liege, May 1985, pp299-303.

3. T. W. Packer, R. Howsley and E. W. Lees, Measurement of the Enrichment of Uranium in Pipework of a Gas Centrifuge Plant, 6th ESARDA Symposium, Venice, May 1984, pp243-248.

4. J. N. Cooley, L. W. Fields and D. W. Swindle, Results from uranium deposition studies for development of a LFUA inspection strategy for gas centrifuge enrichment plants. 7th ESARDA Symposium, Liege, May 1985, pp209-213.

5. T. W. Packer, E. W. Lees, A. R. Talbot and J. L. Hirst, Monitoring the Enrichment of UF_6 gas in Product Header Pipes in a Centrifuge Enrichment Plant Operating at Low Pressure, SRDP-R121, February 1985.