

# RECENT DEVELOPMENTS IN ACTIVE NEUTRON INTERROGATION OF $^{235}\text{U}$ BY COMBINED TRANSMISSION AND EMISSION MEASUREMENTS.

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## ABSTRACT

This paper describes an active neutron interrogation device for the assay of fissile material based on the combination of neutron emission and transmission signals. The latter enables characterisation of samples with respect to moderation and absorption. This technique allows the determination of fissile mass without prior knowledge of the sample's characteristics, e.g. enrichment, density and moisture content. The technique has been demonstrated on LEU and HEU standards from the PERFORMANCE testing LABORATORY (PERLA) at the JRC Ispra and was tested on scrap material originating from a HEU reprocessing facility.

## 1. INTRODUCTION

The 'Phonid' measurement technology was developed<sup>1</sup> by the JRC Ispra in 1975 to support EURATOM measurements of low and high enrichment uranium (LEU and HEU). EURATOM have successfully implemented the technique for safeguards purposes throughout the European Union<sup>2,3</sup>. The JRC Ispra continue to support the use of the original Phonid technology. However, like other currently available active neutron interrogation techniques, Phonid requires difficult and costly calibration and is only accurate for well characterised materials. No development of the basic technique was carried out until the JRC Ispra entered into a joint development project with BNFL Instruments Ltd.. This collaboration has investigated the possibility of improving the performance of Phonid, by implementing an additional transmission measurement.

The main requirements for the device are:

- High penetrating power,
- Simplified routine calibration,
- No need for a prior knowledge of the sample composition,
- Low detection limit in reasonable counting times.

These are fulfilled by using an external neutron source and a neutron detection systems with specific characteristics. The energy of the interrogating neutron flux must be; high enough to ensure sufficient penetration, below the fission threshold of fertile material, low enough to discriminate the source neutrons from the induced fission neutrons and low enough to obtain a good efficiency for the transmission signal. A source which fulfils the above requirements is the  $^{124}\text{Sb}$ - $^9\text{Be}$  photoneutron source currently used in Phonid. Detection of the emission signal must provide a means of separating the epi-thermal source neutrons from the fast induced fission neutrons. This is achieved by using  $^4\text{He}$  proportional counters which enable an energy threshold to be set above the energy of the interrogating flux.

The performance of this interrogating source in combination with a bank of  $^4\text{He}$  detectors has been compared with the performance of an Active Well Coincidence Counter (AWCC)<sup>4</sup>. This comparison illustrates the advantages of Phonid in terms of higher penetrating power and shorter counting times.

## 2. MEASUREMENT PRINCIPLE

Active neutron interrogation relates the amount of fissile material present in a sample to the number of induced fission neutrons measured. For a homogeneous slab sample, irradiated with a broad parallel beam, the total number of induced neutrons,  $F_{nf}$  created in the sample can be obtained by:

$$F_{nf} = \frac{N_A}{A} \int \mathbf{r} \cdot \mathbf{n}_p \cdot \mathbf{s}_{nf} \cdot \mathbf{j}_0 \cdot e^{-\Sigma_a \cdot d} \cdot dV \quad (1)$$

which results in:

$$F_{nf} = \frac{N_A}{A} \mathbf{r} \cdot \mathbf{n}_p \cdot \mathbf{s}_{nf} \cdot \mathbf{j}_0 \cdot S \cdot \frac{1 - e^{-\Sigma_a \cdot d}}{\Sigma_a} \quad (2)$$

where:

|               |  |
|---------------|--|
| $N_A$         | Avogadro's number.   |
| $A$           | Atomic mass number of the fissile material.                          |
| $\rho$        | Density of the fissile material ( $\text{g}/\text{cm}^3$ ).          |
| $\nu_p$       | Number of prompt fission neutrons per induced fission.               |
| $\sigma_{nf}$ | Neutron induced fission cross section ( $\text{cm}^2$ ).             |
| $\phi_0$      | Neutron fluence of the external neutron source ( $\text{cm}^{-2}$ ). |
| $\Sigma_a$    | Total macroscopic interaction cross section ( $\text{cm}^{-1}$ ).    |
| $d$           | Slab thickness (cm).   |
| $S$           | Slab area ( $\text{cm}^2$ ).   |

In the approximation of equation 1 the energy degradation of the source neutrons due to moderating material in the sample is not taken into account. The number of induced fission neutrons is, therefore, only directly proportional to the mass of fissile material,  $m$  when  $\Sigma_a \cdot d \ll 1$  and equation 2 can be approximated by:

$$F_{nf} = \frac{N_A}{A} \cdot \mathbf{n}_p \cdot \mathbf{s}_{nf} \cdot \mathbf{j}_0 \cdot m \quad (3)$$

For most samples assayed by active neutron interrogation this condition is not fulfilled. It follows from equation 2 that the result obtained from an active neutron interrogation device, based on the detection of induced fission neutrons only, is not only dependent on the mass of the fissile material. The result also depends on the neutronic properties of the sample. Samples with different enrichments but the same total amount of fissile material will result in different values for  $F_{nf}$ . The same considerations have to be made for samples containing neutron poisons (i.e. Gd, B). In addition the external neutron source can undergo elastic and inelastic scattering due to the presence of, for example, hydrogenous material and carbon. This will result in energy degradation of the source spectrum and consequently lead to a change in the effective neutron induced fission cross section.

To overcome the shortcomings of existing active neutron interrogation devices a new instrument is proposed. Advanced Phonid is based on the combination of a neutron transmission measurement, sensitive to matrix characteristics, and a neutron emission measurement, sensitive to the sample's fissile content. For the emission measurement, induced fission neutrons are

counted by  $^4\text{He}$  fast neutron detectors. The transmission measurements use a  $^3\text{He}$  proportional counter. The response  $C(s)$  of a  $^3\text{He}$  detector, with length,  $L$ , to a parallel beam passing through a slab sample with height,  $H$  can be approximated by:

$$C(s) = \epsilon \cdot j_0 \cdot (H \cdot e^{-\Sigma_a \cdot d} + (L - H)) \quad (4)$$

which can be transformed to a parameter,  $P_a$

$$P_a = \frac{C(s)}{\epsilon \cdot j_0 \cdot L} = \frac{H}{L} \cdot (1 - e^{-\Sigma_a \cdot d}) \quad (5)$$

where  $\epsilon \cdot j_0 \cdot L$  is a measure of the  $^3\text{He}$  response without the sample present in the measurement cavity. The absorption parameter,  $P_a$  can be used to correct for the absorption characteristics of the sample. The efficiency of the  $^3\text{He}$  detector to the source neutrons is indicated by  $\epsilon$ . Since this efficiency is energy dependent, the  $^3\text{He}$  response also contains a measure of the source neutrons energy spectrum after passing through the sample. A parameter that reflects the mean energy of these source neutrons is the moderation parameter  $P_m$ , defined as the ratio of the responses from bare and Boron wrapped  $^3\text{He}$  counters.

The basic array of the advanced Phonid device is shown in fig. 1. A  $^{124}\text{Sb}$ - $^9\text{Be}$  photoneutron source provides epi-thermal neutrons for active interrogation of the sample. Without moderating material in the measurement cavity the source spectrum has an average neutron energy of 12 keV at the centre of the cavity. Source neutrons induce fissions in any fissile material contained within the sample and the induced fission neutrons are counted in a bank of 27  $^4\text{He}$  detectors which surround the sample. The source geometry is not well defined and, therefore, the interrogating flux cannot be considered as a parallel beam or an isotropic source. This restricts the interpretation of the transmission measurements to a simple ratio of responses with no positional information. The short  $^4\text{He}$  detectors lead to the emission signal having a strong dependence on the position of the sample in the measurement cavity<sup>3</sup>. For these reasons equations 2 and 5 cannot be directly applied to the true measurement geometry. They do, however, indicate the general principles of induced neutron production and absorption by the sample. To minimise spatial distribution effects the samples are placed in the flat response region<sup>3</sup> of the cavity.

Prompt fission neutrons are counted using  $^4\text{He}$  fast neutron detectors of 5 cm diameter, 50 cm length and 15 atm pressure. The absolute detection efficiency of this detection system, consisting of 27  $^4\text{He}$  detectors is 0.3% for the spontaneous fission spectrum of a  $^{252}\text{Cf}$  point source at the centre of the cavity. Fission neutrons have an energy distribution with a modal energy of  $\sim 1\text{MeV}$ . These are separated from the epi-thermal source neutrons and the gamma background by low energy discrimination of the  $^4\text{He}$  signal at the amplifier stage of the counting chains. The source neutrons are measured using 3  $^3\text{He}$  detectors of 2.5 cm diameter, 50 cm length and 4 atm pressure. The transmission measurement employs of two bare (1 and 2) and one boron wrapped (3)  $^3\text{He}$  detectors, with reference numbers as given in fig. 1.

### 3. DATA TREATMENT

The total count rate  $C_t(t)$  in the  $^4\text{He}$  detector chain consists of the passive ( $C_p$ ), active ( $C_a(t)$ ) and background  $C_b(t)$  neutron count rates.

$$C_t(t) = C_a(t) + C_p + C_b(t) \quad (6)$$

where, the active count rate and the background vary with time due to the decay of  $^{124}\text{Sb}$  ( $T_{1/2} = 60.2$  days). For pure  $\text{UO}_2$  material the passive count rate has been measured and is negligible. For the

HEU scrap material discussed in section 5.2 no passive measurements have been performed due to time limitations. This could lead to a small systematic error in the fissile mass determined during that study. The background  $C_b(t)$  for each measurement campaign was measured at regular intervals, the background for each measurement was then obtained by linear interpolation. The long term stability of the emission signal has been reported<sup>3</sup> and was rechecked during the measurement campaign in PERLA, where it was found to be stable, after decay correction, to within 0.3%.

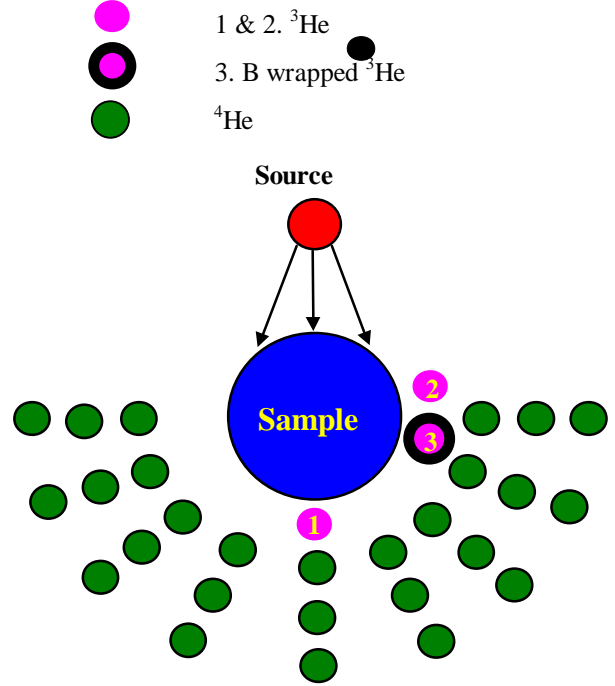


Figure 1. Schematic view of the advanced Phonid physical arrangement.

For the transmission signals only a decay correction is performed resulting in time independent count rates. The background on the transmission signal has been checked by active measurements both with and without a fixed PERLA standard in the measurement cavity. The count rate of the  $^3\text{He}$  counter (1), as a function of time can be described with a precision of 0.1% by applying a decay correction with  $T_{1/2} = 60.2$  days. Other possible sources of background were investigated by using a fit procedure including a time independent parameter. This constant term was negligible within the statistical precision of the data. One concludes, therefore, that the  $^3\text{He}$  result can be used without additional background correction.

Characterisation of the sample using results from the transmission measurement defines two parameters:

The absorption parameter  $P_a$  is calculated by;

$$P_a = 1 - \frac{C_t^{(1)}(s)}{C_t^{(1)}(c)} \quad (7)$$

where,  $C_t^{(1)}(s)$  and  $C_t^{(1)}(c)$  are, respectively, the responses of the bare  $^3\text{He}$  detector (labelled 1 in figure 1) with and without a sample in the sample container at the centre of the measurement cavity.

The response ratio of the bare  $^3\text{He}$  detector (labelled 2) to the Boron wrapped  $^3\text{He}$  detector (labelled 3), defines the moderation parameter  $P_m$ :

$$P_m = \frac{C_t^{(2)}(s)}{C_t^{(3)}(s)} \quad (8)$$

#### 4. CALIBRATION.

##### 4.1 Calibration for the absorption correction.

The uranium samples used for calibration of the absorption correction with a range of enrichments, U-content and densities are summarised in table 1. The samples cover high enrichment  $\text{UO}_2$  powder and low enrichment  $\text{U}_3\text{O}_8$  powder. The broad  $^{235}\text{U}$  mass range which was available for each sample type made this an ideal calibration set.

| Sample type i | Enrichment & U-content (%) | $^{235}\text{U}$ mass (g) | a(i) (g.s) | b(i) (g.s) | d(i) $\cdot 10^{-3}$ (s) | m from eq 11 (g) |
|---------------|----------------------------|---------------------------|------------|------------|--------------------------|------------------|
| 1             | 3.1                        | 14.0                      | 2.14       | 1.057      | 2.530                    | 13.6 (0.3)       |
|               | 84.8                       | 29.3                      | (0.14)     | (0.018)    | (0.061)                  | 29.2 (0.5)       |
|               |                            | 44.3                      |            |            |                          | 45.0 (0.7)       |
|               |                            | 73.8                      |            |            |                          | 72.6 (1.1)       |
| 2             | 5.0                        | 13.5                      | 2.22       | 1.052      | 1.743                    | 13.0 (0.4)       |
|               | 84.8                       | 49.3                      | (0.14)     | (0.016)    | (0.033)                  | 48.2 (0.8)       |
|               |                            | 68.8                      |            |            |                          | 68.3 (1.2)       |
|               |                            | 118.2                     |            |            |                          | 115.6 (1.4)      |
| 3             | 19.9                       | 24.9                      | 2.25       | 1.066      | 0.660                    | 25.1 (0.7)       |
|               | 87.3                       | 49.7                      | (0.07)     | (0.006)    | (0.008)                  | 48.9 (0.8)       |
|               |                            | 79.6                      |            |            |                          | 78.3 (1.0)       |
|               |                            | 99.5                      |            |            |                          | 96.6 (1.3)       |
|               |                            | 237.3                     |            |            |                          | 235.3 (2.0)      |
|               |                            | 248.6                     |            |            |                          | 250.0 (1.9)      |
|               |                            | 298.4                     |            |            |                          | 297.1 (1.4)      |
|               | 348.2                      |                           |            |            | 345.9 (2.3)              |                  |
| 4             | 35.0                       | 50.2                      | 1.94       | 1.108      | 0.515                    | 51.1 (1.0)       |
|               | 87.6                       | 100.3                     | (0.06)     | (0.006)    | (0.008)                  | 99.6 (1.3)       |
|               |                            | 200.7                     |            |            |                          | 198.0 (1.7)      |
|               |                            | 401.4                     |            |            |                          | 408.5 (3.0)      |
|               |                            | 598.8                     |            |            |                          | 594.4 (3.9)      |
| 5             | 60.1                       | 50.0                      | 1.85       | 1.142      | 0.370                    | 52.4 (0.9)       |
|               | 87.3                       | 99.9                      | (0.04)     | (0.004)    | (0.006)                  | 96.6 (1.2)       |
|               |                            | 100.0                     |            |            |                          | 102.7 (1.3)      |
|               |                            | 249.7                     |            |            |                          | 244.8 (2.0)      |
|               |                            | 499.3                     |            |            |                          | 489.6 (3.1)      |
|               | 998.5                      |                           |            |            | 979.7 (5.0)              |                  |
| 6             | 92.4                       | 51.4                      | 1.80       | 1.161      | 0.298                    | 54.4 (1.0)       |
|               | 87.4                       | 102.7                     | (0.03)     | (0.003)    | (0.003)                  | 99.6 (1.2)       |
|               |                            | 205.4                     |            |            |                          | 206.6 (2.1)      |
|               |                            | 308.1                     |            |            |                          | 312.3 (2.8)      |
|               |                            | 513.5                     |            |            |                          | 488.9 (3.4)      |
|               |                            | 1027.1                    |            |            |                          | 1056.3 (6.0)     |
|               |                            | 1540.7                    |            |            |                          | 1560.5 (8.0)     |

Table 1 Sample description and measurement data for the calibration procedure to correct for the different absorption characteristics of the samples.

For each sample family  $i$  ( $i = 1 \dots 6$ ), with fixed enrichment and U-content, the  $^{235}\text{U}$  mass ( $m$ ) can be linked with the emission signal  $C_e$  with a precision of 1% by:

$$m = a(i).C_e^{b(i)} \quad (9)$$

where  $a(i)$  and  $b(i)$  are material dependent calibration factors. This data confirms the reported performance of the Phonid device<sup>2-4</sup>.

The absorption parameter  $P_a$  for the same set of samples is given as a function of the emission signal in fig. 2. The solid lines represent direct proportionality between the absorption parameter  $P_a$  and the emission signal  $C_e$ :

$$P_a = d(i).C_e \quad (10)$$

The material dependent parameter  $d(i)$  are given in table 1.

From fig. 2 one concludes that each sample type is characterised by a well defined correlation between the emission signal and the absorption parameter.

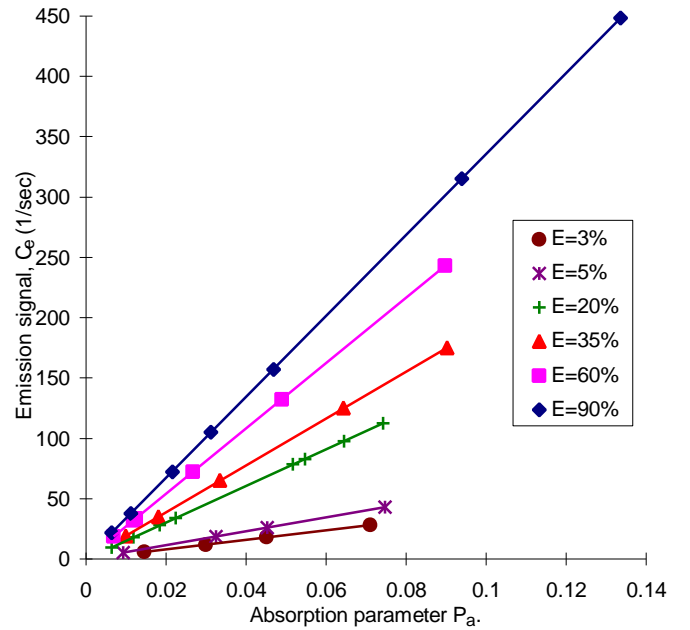


Figure 2. The absorption parameter as a function of emission signal for a range of enrichments.

Using the calibration parameters of equation 9 and 10 a data base has been established from which for a fixed absorption parameter  $P_a$  the  $^{235}\text{U}$  mass as a function of the emission signal can be studied. Examples of the relationship between  $^{235}\text{U}$  mass and emission signal  $C_e$  for two fixed absorption parameters  $P_a$  are given in fig. 3. The behaviour can be approximated by:

$$C_e = \frac{f(P_a).m}{1 + g(P_a).m} \quad f(P_a) = a_0 P_a^{a_1} \quad g(P_a) = b_0 + b_1.P_a^{b_2} \quad (11)$$

Equation 11 defines an intrinsic calibration based on two experimentally determined values and 5 material independent parameters.

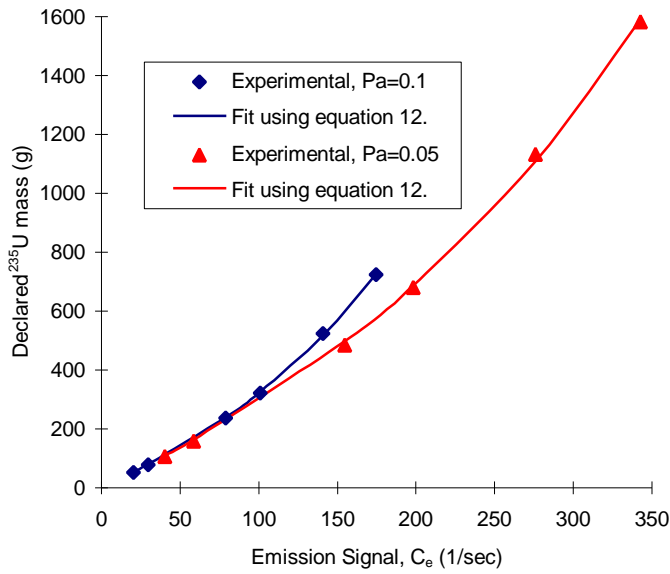


Figure 3. The relationship between the emission signal and the  $^{235}\text{U}$  mass for two fixed values of the absorption parameter.

#### 4.2 Calibration for the moderation correction.

A study of the emission signal  $C_e$  as a function of the moderation parameter  $P_m$  was carried out using the reference samples listed in table 2. Apart from two containing HEU all of these contained natural uranium. As a first exercise the dependency of the moderation parameter  $P_m$  on the sample's fissile mass was checked by measuring two sets of natural uranium samples in different containers over a range of  $^{235}\text{U}$  content. From table 2 one concludes that for both families the moderation parameter is almost independent of the  $^{235}\text{U}$  mass and the emission signal is directly proportional to the  $^{235}\text{U}$  mass for fixed moderation powers.

| Sample description           | $m_d$<br>(g) | $P_m$         | $C_e$      | $m_d/C_e$<br>(g.s) |
|------------------------------|--------------|---------------|------------|--------------------|
| Natural $\text{UO}_2$ powder | 12.66        | 1.010 (0.003) | 3.1 (0.2)  | 4.00 (0.25)        |
| in an Aluminium              | 19.10        | 1.014 (0.003) | 4.9 (0.2)  | 3.86 (0.16)        |
| container                    | 26.02        | 1.011 (0.003) | 6.6 (0.2)  | 3.91 (0.12)        |
| Natural $\text{UO}_2$ powder | 8.65         | 1.053 (0.003) | 3.0 (0.2)  | 2.83 (0.19)        |
| in a plastic container       | 25.06        | 1.053 (0.003) | 8.6 (0.2)  | 2.89 (0.13)        |
| Natural $\text{UO}_2$ powder |              |               |            |                    |
| in a plastic container       |              |               |            |                    |
| +100g polyethylene           | 8.65         | 1.142 (0.003) | 7.6 (0.2)  | 1.13 (0.03)        |
| +125g polyethylene           | 8.65         | 1.178(0.003)  | 8.3 (0.4)  | 1.04 (0.05)        |
| +175g polyethylene           | 8.65         | 1.248 (0.004) | 11.6 (0.4) | 0.75 (0.03)        |
| +225g polyethylene           | 8.65         | 1.314 (0.004) | 14.7 (0.4) | 0.59 (0.02)        |
| +275g polyethylene           | 8.65         | 1.407 (0.005) | 18.2 (0.6) | 0.48 (0.02)        |
| +325g polyethylene           | 8.65         | 1.483 (0.005) | 21.5 (0.6) | 0.40 (0.01)        |
| +375g polyethylene           | 8.65         | 1.567 (0.005) | 23.6 (0.6) | 0.37 (0.01)        |
| HEU Uranium plate            |              |               |            |                    |
| in a plastic container       | 11.30        | 1.142 (0.003) | 4.5 (0.2)  | 2.48 (0.11)        |
| HEU Uranium plate            |              |               |            |                    |
| in a plastic container       |              |               |            |                    |
| +500g of polyethylene        | 11.30        | 1.142 (0.003) | 47.8 (0.6) | 0.24 (0.003)       |

Table 2 Sample description and measurement data for the calibration procedure to correct for the different moderation characteristics of the samples.

The behaviour of the  $^{235}\text{U}$  mass divided by the emission signal as a function of the moderation power is shown in fig. 4. This behaviour is approximated by:

$$C_e = a.m. \frac{P_m^c}{1+b.P_m^c} \quad (12)$$

with  $a$ ,  $b$  and  $c$  as calibration parameters. Before discussing the validation of this technique it must be made clear that this calibration is very limited. Direct proportionality between the emission signal and the  $^{235}\text{U}$  mass is assumed for a fixed moderation parameter with no reference to the absorption. Equation 12, therefore, supposes that absorption and moderation within the matrix can be accounted for by the moderation parameter alone. The direct proportionality and mass independence of the moderation parameter is only checked by experiment for natural  $\text{UO}_2$  samples with low moderation. Increasing the moderation power causes a departure from this direct proportionality due to an increase in the effective fission cross section and a corresponding increase in source neutron absorption.

A more complete calibration procedure is required, in which the emission signal as a function of the  $^{235}\text{U}$  mass is studied for fixed moderation parameters and absorption parameters whilst accounting for the influence of fast neutron moderation on the  $^4\text{He}$  detection efficiency. The calibration samples available during this exercise did not allow such an extensive study.

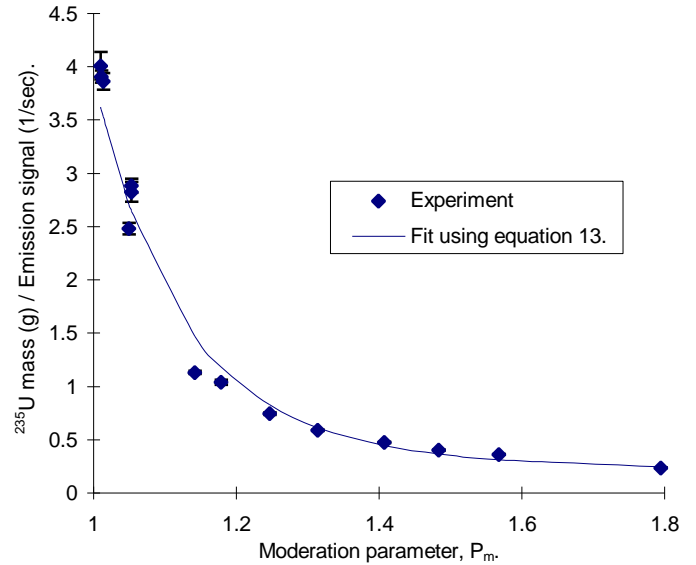


Figure 4. The variation in  $^{235}\text{U}$  mass divided by the emission signal as a function of the moderation parameter.

## 5. VALIDATION

### 5.1 Validation of the absorption correction

Confirmation of the intrinsic calibration was achieved by comparing the measured and declared masses of the samples in table 3. These results shows that even for samples with totally different densities, i.e. pellets and pure metal; the discrepancy between the two mass values are within the statistical precision of the measurement, without any indication of a systematic error. The deviation of the masses determined by this procedure from the declared masses are all within the statistical precision of the

measurement. On average an accuracy of 2% is obtained for the measurements made on these samples. This accuracy is better than the 5% obtained in ref. 2 where an attempt was made to create a calibration curve including an enrichment correction.

| Material type                        | Enrichment (%) | U-factor (%) | Declared mass (g) | Measured mass (g) | Conformance ratio |             |
|--------------------------------------|----------------|--------------|-------------------|-------------------|-------------------|-------------|
| U <sub>3</sub> O <sub>8</sub> powder | 1.0            | 84.8         | 16.6              | 17.1 (0.4)        | 0.97 (0.02)       |             |
|                                      | 1.0            | 84.8         | 23.3              | 23.6 (0.5)        | 0.99 (0.02)       |             |
| UO <sub>2</sub> powder               | 45.6           | 87.3         | 41.3              | 43.5 (0.9)        | 0.95 (0.02)       |             |
|                                      | 45.6           | 87.3         | 82.5              | 82.4 (1.1)        | 1.00 (0.01)       |             |
| UO <sub>2</sub> pellets              | 19.9           | 88.1         | 234.0             | 228.0 (5.9)       | 1.03 (0.03)       |             |
|                                      | 19.9           | 88.1         | 287.0             | 279.5 (5.1)       | 1.03 (0.02)       |             |
|                                      | 19.9           | 88.1         | 292.0             | 290.0 (5.3)       | 1.01 (0.02)       |             |
|                                      | 19.9           | 88.1         | 294.0             | 293.0 (6.3)       | 1.00 (0.02)       |             |
|                                      | 35.0           | 88.1         | 70.8              | 70.3 (1.7)        | 1.01 (0.02)       |             |
|                                      | 45.6           | 88.1         | 91.1              | 90.7 (1.5)        | 1.00 (0.02)       |             |
|                                      | 60.1           | 88.1         | 120.2             | 118.3 (2.6)       | 1.02 (0.02)       |             |
|                                      | 92.4           | 88.1         | 186.9             | 187.5 (3.8)       | 1.00 (0.02)       |             |
|                                      | U-metal        | 93.1         | 100.0             | 1723.2            | 1698.0 (20.0)     | 1.02 (0.01) |

Table 3. Results for the validation of the absorption correction.

## 5.2 Validation of moderation correction

The samples measured during the campaign at BNFL Springfields are given in table 4. These samples represent a wide variety of ‘scrap’ material originating from the production stream of the HEU reprocessing facility at Dounreay. The first column in the table (DA) gives the fissile masses as determined from destructive analysis. This analysis involved dissolution of the sample followed by mass spectroscopy. These results have a quoted precision of ~3% which is largely due to the corrections made to account for any undissolved material. Also given are; the <sup>235</sup>U masses as determined by Advanced Phomid and AWCC measurements<sup>5,6</sup>, the mass per unit response values for both techniques and the moderation parameter as determined by Advanced Phomid measurements.

| DA m <sub>d</sub> (g) | AWCC m <sub>d</sub> /R (g.s) | AWCC m <sub>a</sub> (g) | Moderation parameter P <sub>m</sub> | m <sub>d</sub> /C <sub>e</sub> (g.s) | Phomid m <sub>p</sub> (g) |
|-----------------------|------------------------------|-------------------------|-------------------------------------|--------------------------------------|---------------------------|
| 5.3                   | 0.047                        | 2.4(0.3)                | 1.017 (0.004)                       | 3.29 (0.04)                          | 5.5 (0.4)                 |
| 5.0                   | 0.039                        | 2.9(0.3)                | 1.015 (0.003)                       | 3.05 (0.19)                          | 5.7 (0.3)                 |
| 4.9                   | 0.034                        | 3.4(0.3)                | 1.058 (0.003)                       | 2.75 (0.16)                          | 4.5 (0.2)                 |
| 3.4                   | 0.037                        | 1.8(0.3)                | 1.058 (0.003)                       | 2.957 (0.26)                         | 3.0 (0.2)                 |
| 6.1                   | 0.032                        | 4.9(0.5)                | 1.423 (0.004)                       | 0.41 (0.01)                          | 6.3 (0.2)                 |
| 29.7                  | 0.044                        | 26.2(0.9)               | 1.068 (0.003)                       | 2.44 (0.06)                          | 28.9 (0.8)                |
| 16.3                  | 0.033                        | 17.5(0.7)               | 1.057 (0.003)                       | 2.05 (0.05)                          | 20.4 (0.5)                |
| 11.4                  | 0.036                        | 9.4(0.2)                | 1.535 (0.004)                       | 0.40 (0.01)                          | 9.3 (0.3)                 |
| 1.8                   | 0.034                        | 0.9(0.2)                | 1.060 (0.003)                       | 5.29 (1.56)                          | 0.9 (0.2)                 |
| 4.7                   | 0.053                        | 1.8(0.2)                | 1.050 (0.003)                       | 4.16 (0.74)                          | 3.1 (0.2)                 |
| 26.7                  | 0.038                        | 27.5(1.0)               | 1.431 (0.004)                       | 0.53 (0.01)                          | 21.0 (0.5)                |
| 23.4                  | 0.040                        | 21.4(0.9)               | 1.313 (0.003)                       | 0.55 (0.01)                          | 26.1 (0.6)                |
|                       |                              | 22.3(0.5)               | 1.020 (0.003)                       |                                      | 26.0 (0.8)                |
|                       |                              | 56.5(0.8)               | 1.472 (0.004)                       |                                      | 49.0 (1.2)                |

Table 4. Results of the validation of the moderation correction. These data apply to measurements which were taken on HEU material originating from the Dounreay reprocessing facility.

From table 4 one concludes that the emission signal of the AWCC is more proportional to the <sup>235</sup>U mass as compared to the signal obtained from the Phomid measurements. The AWCC emission signal is less sensitive than that of Phomid due to the fact that the AWCC interrogating flux is almost thermal. This means that the effective neutron energy and consequently the effective neutron induced fission cross section does not change as a function of the sample’s moderation power. Although this is advantageous for very small samples with low fissile material density it results in an under estimate of the <sup>235</sup>U mass for large or dense samples. In contrast, the emission signal of Advanced Phomid is very sensitive to the moderation power of the sample due to the high energy of the interrogating source neutrons. Therefore, the change in emission signal with moderation power is corrected for by using the moderation parameter obtained from the transmission measurement. This ‘self categorisation’ of samples is a major advantage for the Advanced Phomid device since there is no requirement to have detailed knowledge of the sample composition prior to calibrating the instrument with a set of similar samples.

In fig. 5 the emission signal of the AWCC, the background corrected reals (R), are plotted against the emission signal obtained from Phomid (C<sub>e</sub>). Here the samples can be classified into two distinct groups as indicated by the solid lines. The fact that the samples are different with respect to the behaviour of the interrogating neutrons is even more obvious from the moderation parameter values given in table 4. The two groups are, therefore, be classified by their moderation parameters.

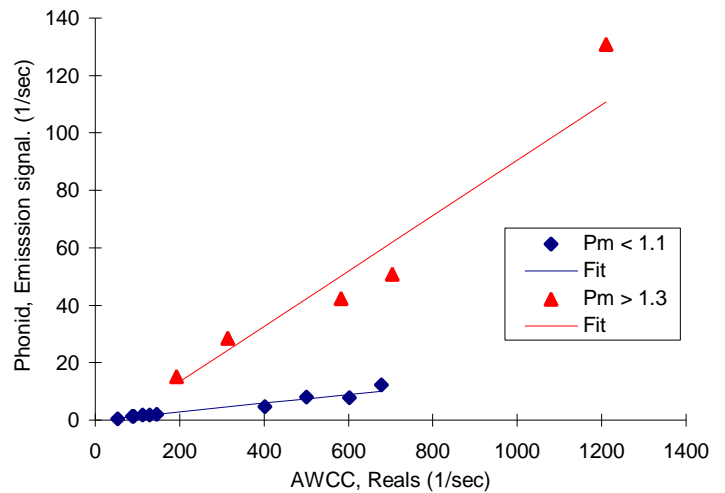


Figure 5 A comparison of the emission signals from Phomid and AWCC.

A comparison between the masses determined with the AWCC and Advanced Phomid is shown in fig. 6. This data shows that with the exception of the sample with m<sub>d</sub> = 1.8g the Advanced Phomid does not introduce a systematic error. The overall accuracy, which for this set of miscellaneous scrap is of the order of 15%, will be improved by a more extensive systematic study as mentioned in section 4.2. The values obtained with the AWCC suffer from systematic error which is not constant but will depend on the density of the fissile material in the sample. It should also be made clear that the error bars shown are one sigma statistical errors, which in the case of the Advanced Phomid result from measurement times of ~5 minutes and in the case of AWCC result from measurement times of ~30 minutes.

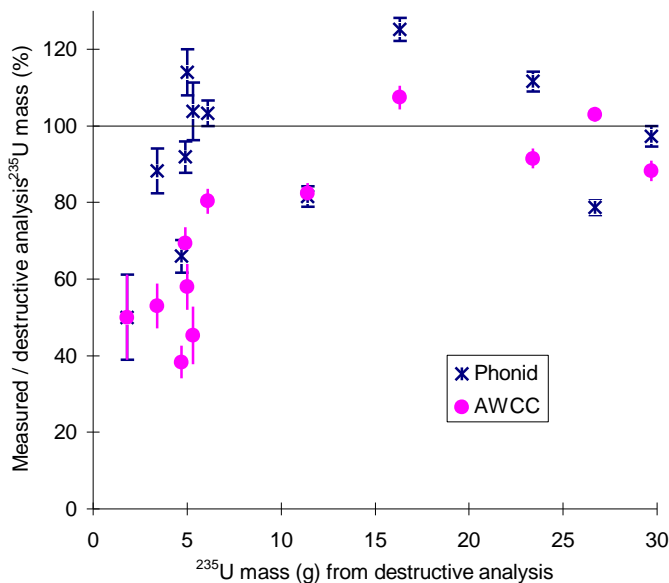


Figure 6. This graph shows the level of agreement between masses determined by destructive chemical analysis and those determined by Advanced Phonid and AWCC measurements.

## 6. CONCLUSIONS

This paper presents an advanced development of the existing 'Phonid' device, currently used by EURATOM for Safeguards measurements of enriched uranium within the EU. The technique is based on the combination of the emission and transmission signals which allows 'self categorisation' of samples and enables an intrinsic correction for their absorption and moderation to be made. The correction for the absorption power of the sample has been demonstrated and results in an overall accuracy of 2% for the sample set shown in table 3.

Correction for the moderation power, especially when used with the absorption correction, still needs confirmation. This will require a more detailed study involving measurements on a set of well calibrated samples which can be separated into several fixed moderation parameter groups each with a wide range of parameters, e.g.  $^{235}\text{U}$  mass and enrichment. The present data does, however, show a reduction in the systematic errors that are usually associated with active neutron interrogation techniques.

This technique can be summarised by highlighting the following advantages which it offers:

- **Improved accuracy.** Small differences in the sample composition are accounted for by the transmission measurement. This is not possible in either the original Phonid device or AWCC.
- **Reduced operating costs.** This device eliminates the requirement to perform long and costly routine calibrations for each material type to be measured. Instead, a full intrinsic calibration is shipped with the instrument. This calibration is periodically verified during measurement campaigns by measuring known samples of natural uranium. This also removes the necessity to calibrate the instrument using a set of standards with similar neutronic characteristics to those of the unknown samples. Even when the measurement campaign includes highly enriched uranium there is no requirement to calibrate using HEU samples. This means that the cost and

complication of fabricating, transporting and using such material can be avoided.

- **A wider range of materials can be measured.** The sample characterisation which is provided by the transmission measurements allows samples of unknown composition to be measured accurately. Our experience has shown that reasonable accuracy (~15%) can be obtained by using a calibration based on natural uranium standards to measure samples with uranium enrichments of up to 90%. The results listed in table 4 were obtained in this way.
- **Greater measurement independence.** No prior knowledge of the material type is required. This complies with the continued pressure that regulatory bodies are placing on instrument manufacturers to reduce their dependence on all types of declared data.

All the statistical errors quoted in this paper are at the 68% confidence level (one sigma).

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