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Quality Assurance in Nuclear Fuel Research at the  
Laboratory of High- and Medium-level Activity at  
SCK•CEN

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**TABLE OF CONTENTS**

<b>1</b>	<b>Introduction.....</b>	<b>3</b>
<b>2</b>	<b>Discussion.....</b>	<b>3</b>
	2.1 <i>Non-destructive examinations.</i> .....	3
	2.2 <i>Fysico-chemical examinations.</i> .....	11
	2.3 <i>Microstructure examinations.</i> .....	15
<b>3</b>	<b>Conclusion.....</b>	<b>16</b>
<b>4</b>	<b>Recommendations. ....</b>	<b>16</b>
<b>5</b>	<b>References. ....</b>	<b>17</b>
<b>6</b>	<b>List of abbreviations .....</b>	<b>17</b>

**ABSTRACT**

Quality assurance in nuclear fuel research demands specific calibration and validation methodologies. Indeed the analytical experiments in hot-cells on highly radioactive objects are non-standard and many times unique. The standards and validation methods developed for and applied to the main nuclear fuel research experiments in the SCK•CEN hot laboratory are outlined.

**KEYWORDS**

Hot Laboratory  
Nuclear Fuel Research  
Quality Assurance

## 1 Introduction.

Quality awareness began in many laboratories in the 1980s and has developed rapidly during the last decades. Many standardised and validated methods have become available and have been implied, also by hot laboratories, in order to strive at a maximum traceability and reliability of results.

In the 1990s, increasing demands from industrial end-users and general competition are forcing Post-Irradiation Examination (PIE) laboratories to seek third-party recognition of their quality. There is a tendency to move, via certification of the laboratory quality system (ISO 9000 family and/or the European Standard series EN 45 000), to accreditation needing proof of professional and technical competence in laboratory tasks. The costs of achieving accreditation are considerable and mainly involve the production of quality handbooks and written work procedures by personnel. The main reward is an open quality system provided with transnational recognition through participation of independent authorised accrediting bodies.

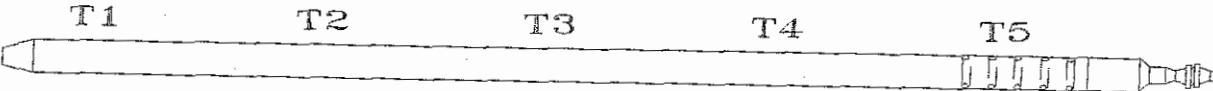
The process of setting up a quality system according to international standards is at present going on at the LHMA at SCK•CEN. The validation of the methods applied is a key item in this process. Validation of a test method aims at the demonstration that the method is fit for its intended purpose. It consists of two interrelated steps: the demonstration of its representativeness and the demonstration of its technical capability. Representativeness means the suitability to respond to the customers need, or in other words to measure the right properties - requiring accurate knowledge of the practical use of the results and of the real service conditions of the object of the test. In the second step the appropriate technical performance of the method relative to the acceptable uncertainty has to be proofed. This paper concentrates on this second step and more specifically on the calibrations. It gives an overview of the calibration methods applied at present in the different nuclear fuel related examinations at the LHMA at SCK•CEN to proof their accuracy.

## 2 Discussion.

### 2.1 *Non-destructive examinations.*

The *fuel rod length* measurement, performed by a contact sensing device determining the position of the free end of the fuel rod that is mounted accurately with its other end at a 0-reference position, is calibrated with certified length standards. Different length standards (individually from 300 mm up to 1000 mm long, and with an accuracy of  $\pm 0.01$  mm) allow assembling a standard length approximating as close as possible the nominal length of the fuel rod to be measured. Standard measurements are performed both before and after the test rod. At least three length measurements are performed at two azimuthal directions ( $0^\circ$  and  $180^\circ$ ) both on the standards and the rod. Furthermore, temperature measurements with a thermocouple on the standards as well as on different axial positions on the rod are performed

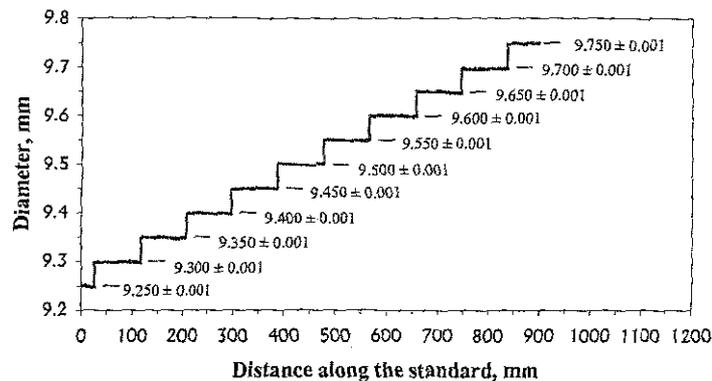
to allow correcting for thermal dilatation. A typical example of a fuel rod length determination data sheet is given in table 1. The accuracy amounts to  $\leq \pm 0.05$  mm.

SCK•CEN		FUEL ROD LENGTH MEASUREMENT				RMO/LHMA
IDENTIFICATION :	F6547	DATE (of measurement) :	10/5/99			
PROGRAM :	BACCHANAL 3	OPERATOR(s) :	A.Gys/G.Cools			
CUSTOMER :	SCK•CEN					
STANDARD BEFORE ROD MEASUREMENT						
Type :	1					
CERTIFIED LENGTH ( $L_{S,C,20}$ ) :	1136.08 $\pm$ 0.03 mm			$L_{S,20} = L_{S,T_S} / (1 + \alpha_{ST} (T_S - 20))$ $\alpha_{ST} = 1.36 \pm 0.01 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$		
Temperature ( $T_S$ , $^\circ\text{C}$ )	Length ( $L_{S,T_S}$ , mm)			Length 20 $^\circ\text{C}$ ( $L_{S,20}$ , mm)		
	0 $^\circ$	180 $^\circ$				
25.5 $\pm$ 0.5	1136.10	1136.13		1136.02 $\pm$ 0.03		
	1136.08	1136.13				
	1136.08	1136.10		Length 25,5 $^\circ\text{C}$ ( $L_{S,T_S}$ , mm)		
Mean Value	1136.09 $\pm$ 0.03	1136.12 $\pm$ 0.03		1136.10 $\pm$ 0.03		
ROD MEASUREMENT		F6547				
NOMINAL LENGTH ( $L_{R,N}$ ) :	1136.0 $\pm$ 0.5 mm					
Hot Cell Temp. ( $T_C$ , $^\circ\text{C}$ )	Length ( $L_{R,T_{mean}}$ , mm)					
	0 $^\circ$	180 $^\circ$				
25.5 $\pm$ 0.5	1138.90	1138.90		Length 38.1 $^\circ\text{C}$ ( $L_{R,T_{mean}}$ , mm)		
	1138.93	1138.90				
	1138.95	1138.88		1138.91 $\pm$ 0.03		
Mean Value	1138.93 $\pm$ 0.03	1138.89 $\pm$ 0.03				
STANDARD AFTER ROD MEASUREMENT						
Temperature ( $T_S$ , $^\circ\text{C}$ )	Length ( $L_{S,T_S}$ , mm)			Length 20 $^\circ\text{C}$ ( $L_{S,20}$ , mm)		
	0 $^\circ$	180 $^\circ$				
25.5 $\pm$ 0.5	1136.06	1136.06		1135.98 $\pm$ 0.03		
	1136.06	1136.07				
	1136.06	1136.05		Length 25,5 $^\circ\text{C}$ ( $L_{S,T_S}$ , mm)		
Mean Value	1136.06 $\pm$ 0.03	1136.06 $\pm$ 0.03		1136.06 $\pm$ 0.03		
TEMPERATURE MEASUREMENT ROD ( $^\circ\text{C}$ )						
	$T_{Mean}$	$T_1$	$T_2$	$T_3$	$T_4$	$T_5$
value	38.1	40.9	41.9	40.9	34.7	28.8
weight	90	7	32	12	32	7
precision	5.0	0.5	0.5	0.5	0.5	0.5
						
Rod Length 20 $^\circ\text{C}$ :		$L_{R,20} = 1138.79 \pm 0.03$ mm			$L_{R,20} = L_{R,T_{mean}} / (1 + \alpha_{Zry} (T_{mean} - 20))$ $\alpha_{Zry} = 5.84 \pm 0.01 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$	
COMMENT						
$\Delta$ Length = + 2.8 $\pm$ 0.5 mm or + 0.25 $\pm$ 0.04 %						

**Table 1:** Fuel rod length measurement.

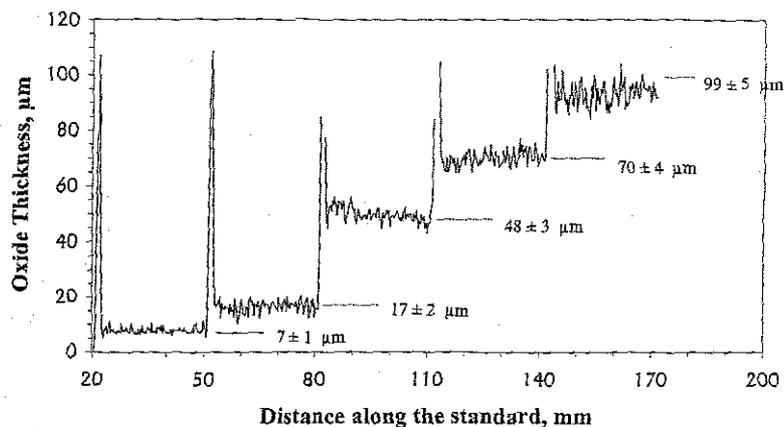
The *fuel rod diameter* is measured by two opposed axial movement gauge heads (linear displacement transducers) properly positioned on the fuel rod surface by a sensing head carriage.

The linear displacement transducers have a measuring range of 4 mm but they show only exact linear behaviour in a limited range of 0.5 mm, where their accuracy is  $\leq \pm 2 \mu\text{m}$ . This limited range is however appropriate to cover creep and PCMI induced diametrical changes, even in the event of a severe transient. In view of the foregoing, step standards containing an appropriate range of certified diameters are necessary and available, such as to enable calibration to be performed on a standard of diameter approximating the nominal rod diameter as close as possible – e.g. Fig. 1. Under these circumstances, an accuracy within  $\pm 5 \mu\text{m}$  ( $2\sigma$ ) is obtained.



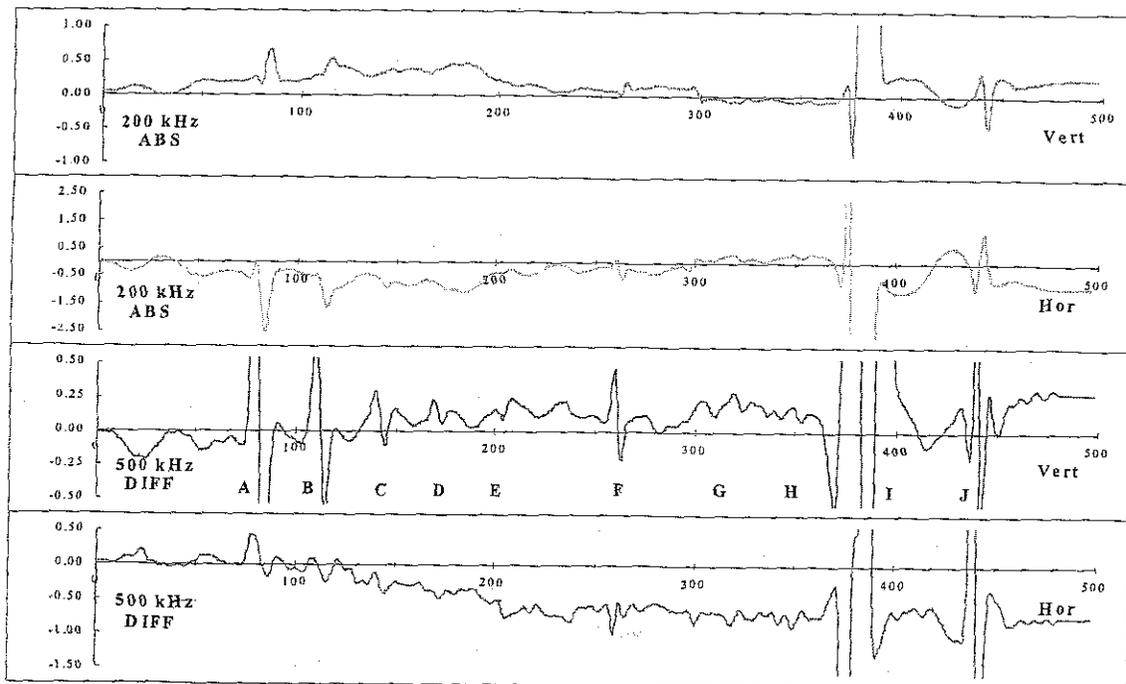
**Fig. 1:** Profilometry – step standard calibration.

*Fuel rod waterside corrosion* is measured by an Eddy Current probe coil excited at high frequency ( $\sim 3 \text{ MHz}$ , thus giving rise to superficial eddy currents in the clad). The net coil impedance is affected by the proximity between coil and metal and thus dependent on the thickness of the insulating oxide (and crud) layer on the outer clad surface. The method is calibrated with oxidised zircaloy tube samples with known oxide layer thickness (as determined by optical microscopy) – Fig. 2. The accuracy amounts to  $\pm 5 \mu\text{m}$ , and is regularly confirmed by the analysis by optical microscopic examinations on fuel rod cross sections sampled from fuel rods previously been analysed by the Eddy Current probe.



**Fig. 2:** Eddy Current oxide measurement calibration on oxidised Zry standards.

Fuel rod *clad integrity testing* is performed by an *Eddy Current technique using encircling coils*. By using two closely spaced differentially wound coils and by measuring simultaneously in absolute and differential mode as well as in multifrequency mode, a high sensitivity with appropriate discrimination between defect and noise signals is obtained. In addition to the detection and axial location of cladding deformations and defects, qualitative and quantitative indication on the type (inner clad defect, outer clad defect, through wall hole) and size of the cladding perturbation can be obtained through comparison with standards. These standards are of design as close as possible with the to be examined fuel rod cladding (i.e. similar or the same diameter, wall thickness and material type) and accurately known artificial defects have been machined on it – an example is given in fig. 3. At the examination of a fuel rod with the eddy current equipment, the artificial defects provide a correlation with similar defect signals observed while controlling the fuel rod cladding.



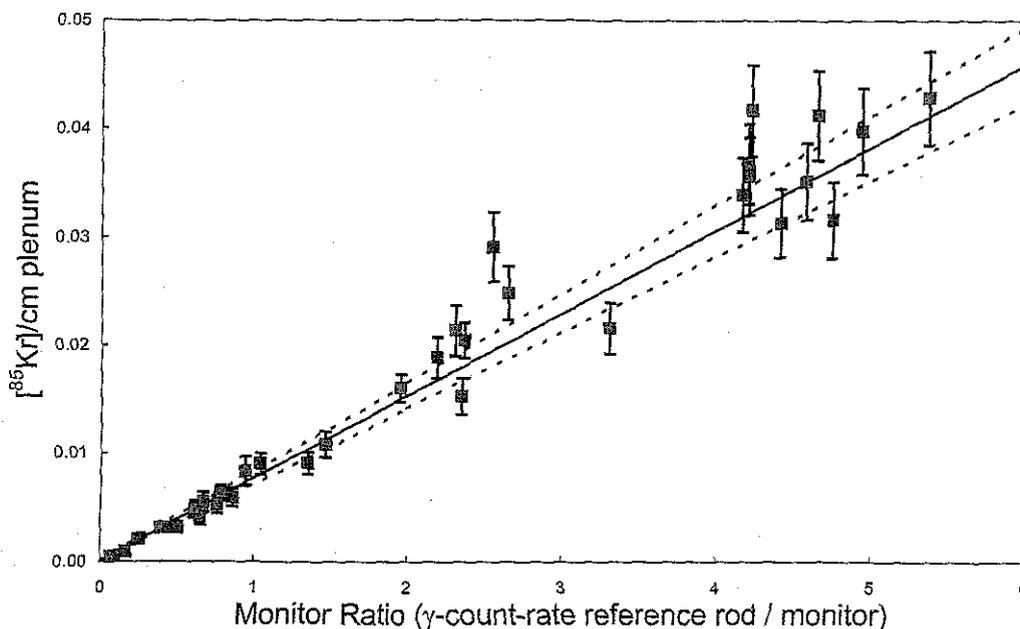
**Fig. 3:** Eddy Current scan of standard

Standard defects :	A-E	Internal notches $4.5 \times 0.2 \times 0.3-0.04$ mm (length $\times$ width $\times$ depth)
	F	Through wall hole $\varnothing 0.4$ mm
	G-H	External notches $4.7 \times 0.2 \times 0.04$ mm
	I	Circumferential notch $10.5 \times 0.06$ mm (external, i.e. simulating a diameter decrease)
	J	Ridge 0.03 mm

In the  $\gamma$ -spectrometric analyses, the fuel rod moves in front of a collimator directing the  $\gamma$ -radiation beam to an intrinsic Ge detector. Collecting  $\gamma$ -spectra along the axis of the fuel rod and analysis of these spectra allows determining the  $\gamma$ -activity of discrete fission products and its axial distribution. Combined with appropriate standard measurements, the isotope specific  $\gamma$ -activities can be converted into absolute fission product concentrations.

This is applied to non-destructively evaluate *fission gas release* by measuring the  $^{85}\text{Kr}$  activity at the plenum of the rod. Comparative measurements are performed, under the same experimental conditions, on both the rod to be analysed and on a so-called standard monitor rod at disposal at LHMA.

The monitor rod is a fuel rod of a special design, i.e. that is appropriate (sleeve plenum) and accurately characterised (materials & geometry). It has been irradiated up to an appropriate fission gas release; i.e. up to an easily measurable  $^{85}\text{Kr}$  content was present in its plenum. It is calibrated for its plenum  $^{85}\text{Kr}$  content by comparative  $^{85}\text{Kr}$  measurements with reference rods, i.e. fuel rods to be punctured afterwards. As such the measured  $^{85}\text{Kr}$  content of the punctured rods serve to calibrate the  $^{85}\text{Kr}$  content of the monitor rod – Fig. 4. The accuracy of one single calibration is estimated at 10 – 30 % (depending on the accuracy of the reference rod design characteristics). At present about 37 calibrations of the monitor rod have been performed resulting in a calibration factor accuracy of 4 %.



**Fig. 4:**  $^{85}\text{Kr}$  monitor rod calibration.

The calibration and analysis methodology is compiled in Table 2, together with the uncertainty of all the individual steps as well as the therefrom resulting final accuracy on the determination of the fission gas release, i.e. 10 – 25 % relative.

$\gamma$ -Spectrometric Analysis of Fission Gas Release ( $^{85}\text{Kr}$ method)	$2\sigma$ uncertainty
<b>Monitor Rod Calibration</b>	
$^{85}\text{Kr}$ $\gamma$ -activity evaluation of reference & monitor rod	
➤ $\gamma$ -peak analysis	1 – 15 %
➤ decay correction	0.2 %
➤ shielding correction	3 – 5 %
➤ corrected $^{85}\text{Kr}$ activity monitor & reference rod	3 – 15 %
➤ monitor ratio ( $\gamma$ -count-rate reference / monitor)	5 – 20 %
<b>Puncture of the reference rod</b>	
➤ total gas content fuel rod	3 %
➤ MS analysis of $^{85}\text{Kr}$ isotopic content	3 – 10 %
➤ decay correction (reference = end of irradiation date)	0.2 %
➤ rod geometry	
→ total free rod volume	1 – 5 %
→ free volume per cm plenum	2 – 5 %
➤ [ $^{85}\text{Kr}$ ] / cm plenum	5 – 15 %
Calibration factor ( [ $^{85}\text{Kr}$ ] / cm plenum / monitor ratio)	10 – 30 %
<b>FGR measurement</b>	
$^{85}\text{Kr}$ $\gamma$ -activity evaluation of analysed & monitor rod	
➤ $\gamma$ -peak analysis	1 – 15 %
➤ decay correction (reference = end of irradiation date)	0.2 %
➤ shielding correction	3 – 5 %
➤ corrected $^{85}\text{Kr}$ activity monitor & analysed rod	3 – 15 %
➤ monitor ratio ( $\gamma$ -count-rate analysed rod / monitor)	5 – 20 %
Calibration factor ( [ $^{85}\text{Kr}$ ] / cm plenum / monitor ratio)	3 %
➤ [ $^{85}\text{Kr}$ ] / cm plenum	7 – 20 %
<b>Fission Gas Release (FGR)</b>	
➤ total FG	
→ free volume per cm plenum	2 %
→ total free rod volume	5 %
→ [ $^{85}\text{Kr}$ ] / [ $^{81}\text{Kr}$ ] isotopic content	3 – 5 %
→ [ $^{136}\text{Xe}$ ] / [ $^{85}\text{Kr}$ ] ratio	3 – 10 %
→ [ $^{136}\text{Xe} + ^{85}\text{Kr}$ ] / total plenum	10 – 25 %
➤ fission gas generation ( $30\text{ cm}^3$ (STP) / $\text{kg}_{\text{HM}}$ / $\text{GWd}/t_{\text{M}}$ )	5 %
➤ FGR	10 – 25 %

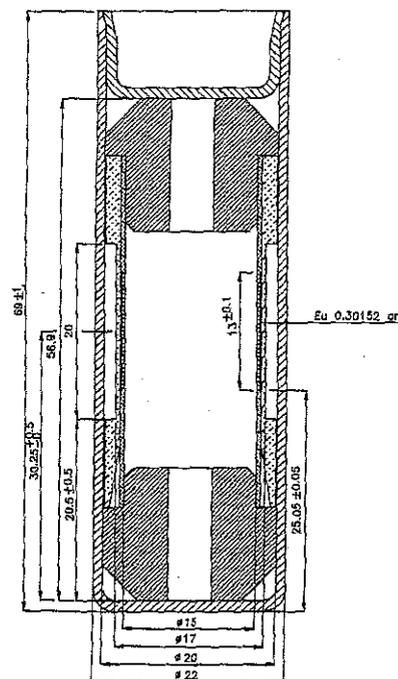
**Table 2:** Calibration and analysis methodology of the  $^{85}\text{Kr}$  evaluation of fuel rod fission gas release.

A second application of quantitative  $\gamma$ -spectrometry concerns the evaluation of the *rod burnup*. The method consists in performing comparative measurements of the  $^{137}\text{Cs}$  content of the fuel to be investigated and will-known reference fuel samples, i.e. fuel rod sections that have been accurately characterised by wet chemical analysis of an adjacent section. The results of the comparative measurements have to be corrected for the different history and geometry conditions of the reference and the examined rod. These corrections include:

- self absorption of  $\gamma$ 's in the fuel ;
- absorption of  $\gamma$ 's by materials between fuel and detector ;
- decay of  $^{137}\text{Cs}$  during the irradiation ;
- decay of  $^{137}\text{Cs}$  during the cooling ;
- fission yield ;
- energy released per fission.

The last two corrections are important if the measurements concern different fissile nuclides. The uncertainty of the thus obtained burn-up value is approximately 8 – 10 % ( $2\sigma$ ), mainly due to the uncertainties of the burn-up of the reference samples and the determination of the peak area of the  $^{137}\text{Cs}$   $\gamma$  peaks.

A third application of quantitative  $\gamma$ -spectrometry concerns the evaluation of the *rod power rating*. The linear fission power of a fuel rod during irradiation is determined by measuring short-life fission products such as  $^{140}\text{La}$ ,  $^{131}\text{I}$ ,  $^{95}\text{Zr}$ , etc. The half lives range from a few days to two months. Due to the short half-lives it is impossible to construct reference samples of these nuclides. A more direct method has been developed to compare the measured activity in an irradiated fuel rod with a calibrated reference source – Fig. 4. This reference source is a 45.7 GBq (01-07-92)  $^{152}\text{Eu}$  source, calibrated very precisely at IRMM at Geel, Belgium.



**Fig. 5:**  $^{152}\text{Eu}$  reference source.

The reference source and the irradiated fuel rod are measured under the same geometrical conditions. The following corrections are used to determine finally the linear power:

- $\gamma$  yield ;
- self absorption of  $\gamma$ 's in the fuel ;
- absorption of  $\gamma$ 's by materials between fuel and detector ;
- decay during the irradiation ;
- decay during the cooling ;
- fission yield ;
- energy released per fission.

Taking into account the uncertainty on the activity measurements and the estimated uncertainties on all of the above individual processing steps, the final overall accuracy of the quantitative  $\gamma$ -spectrometric linear power determination is determined as  $\pm 5 - 10 \%$ .

The *validity of the  $\gamma$ -spectrometric test methods of the rod burnup and power rating* is assessed by comparing its results to those obtained by means of other independent test methods, which have been developed at the SCK•CEN for the same purpose:

- the thermal balance method – an on-line method applied within the PWC-CCD irradiation device which, on base of differential temperature measurements and cooling water flow rate measurements, evaluates the energy dissipated by the fuel rod during irradiation and hence provides the fuel rod power – this method has been qualified itself by calibration with electrically heated rods in steady state conditions and at nominal reactor power conditions and by proper assessment of nuclear ( $\gamma$ ) heating and heat losses in transient and high reactor power conditions – resulting in an estimated uncertainty on the fuel rod power  $\leq 5 \%$  ;
- the fluence-dosimetry method – based on the measured responses of neutron dosimeters (located near to the fuel rod under irradiation) combined with neutron calculations of the fuel rod environment, enabling to obtain the n-flux at the fuel rod and therefrom (taking into account the actinide concentration in the fuel and their resonance-self-shielded cross-section) the fuel rod power – this relatively new method is still under development and is as such not yet qualified ;
- the destructive radiochemical method – relying on the radiochemical and TIMS analysis of selected fission products (Nd & Cs isotopes) as well as the U, Pu and transplutonium isotopes (Am & Cm) – it is a highly reliable and accurate method with several cross-checking points within the procedure and recognised as reference (qualified as ASTM E321-69) – its accuracy amounts to 2 – 4 %.<sup>[1]</sup>

Examples of results obtained with these different methods in comparison with the results obtained with the  $\gamma$ -spectrometric measurements are given in table 3 for the fuel rod burnup and in table 4 for the fuel power rating. The results indicate a good agreement between the results obtained with three of the four methods; i.e. they all fall within the expected uncertainty limits. For the fourth - fluence dosimetry - method an agreement within 20 % is observed with the  $\gamma$ -spectrometry or the thermal balance method, whatever the type of dosimeter considered. There seems to be a systematic underestimation of the fission power as

obtained from the fluence dosimetry. The causes of these deviations beyond acceptable limits should be clarified and the fluence dosimetry method is to be redefined accordingly.

Fuel rod burn-up (GWd/t <sub>HM</sub> )			
Fuel Rod	Thermal balance	$\gamma$ -Spectrometry	Radiochemistry
UN1	23.6 [-8.4]	25.8	25.9 [+0.4]
MN1	25.3 [+5.3]	24.0	23.5 [-2.1]
MN2	45.0 [-1.8]	45.8	45.2 [-1.3]
MN3	57.0 [+4.4]	54.6	54.3 [-0.5]

**Table 3:** Comparison of various methods for the determination of fuel rod burn-up.  
([ ] = percent difference with respect to  $\gamma$ -spectrometry)

Fuel rod linear power (W/cm)			
Fuel Rod	Thermal balance	$\gamma$ -Spectrometry	Fluence Dosimetry (Fe / Co Dosimetry)
UR1	477.5 [+1.1]	472.4	427.5 [-9.5] 479.2 [+1.4]
UR1	476.1 [+2.9]	462.9	382.2 [-17.4] 386.5 [-16.5]
MR1	439.6 [-4.2]	458.8	385.2 [-16.0] 421.7 [-8.1]

**Table 4:** Comparison of various methods for the determination of fuel rod linear power.  
([ ] = percent difference with respect to  $\gamma$ -spectrometry)

## 2.2 Fysico-chemical examinations.

**Rod puncture** is the most accurate and complete fission gas release determination method. The rod is partially enclosed at the plenum level in an in-cell vacuum puncture chamber that is connected by a capillary tube to an out-of-cell collection system. This collection system is equipped with vacuum and transfer pumps, calibrated volumes and high performance pressure transducers (accuracy  $\pm 0.3$  %), enabling to measure the amount of gas collected on puncturing the rod. The accuracy relies essentially on the calibrated volumes, which have been calibrated prior to their assembling. The volume-valve assemblies were calibrated with a mercury-(and water-)filling weighing technique, resulting in an accuracy of  $\leq 2$  %. The rest of the collection system is calibrated by expanding gas from these calibrated volumes and measurement of the corresponding pressure drop. As such the accuracy of the total gas volume determination is estimated at  $\pm 3$  %.

Both the elemental composition of the gas and the isotopic composition of the fission gases Kr and Xe are determined by mass spectrometric analysis. The quadrupole mass spectrometer is calibrated by standard practices:

- the inlet system is equipped with an accurate diaphragm Baratron pressure gauge (range 0.0001 – 1 mbar / resolution 0.001 mbar / precision 0.1% / independent on the nature of the gas);
- measurement of the absolute sensitivity (A / mbar) for the different m/e peaks for each gas over the complete pressure range using ultrapure gases ;
- measurement of the to be analysed gas over the whole appropriate mass range (1 – 150 amu) and identification and determination of the partial pressure of each component on base of foregoing absolute sensitivities, using all relevant peaks ;
- independent control of the performance by checking the correspondence between the calculated (sum of individual partial pressures) and measured total inlet pressure.

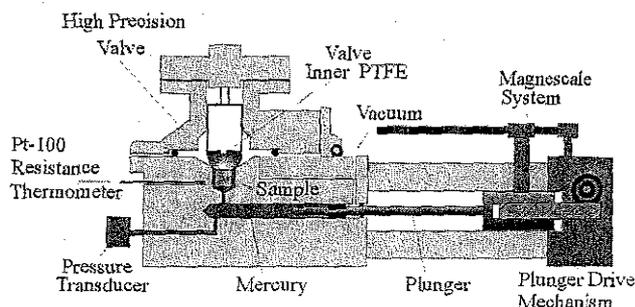
The therefrom-derived accuracy is as follow ( $2\sigma$ ):

- for the gas components (if the amount is  $> 1$  ml STP) :
  - $\pm 2\%$  at component level  $> 10\%$  ;
  - $\pm 5\%$  at component level 1 - 10 % ;
  - $\pm 10\%$  at component level 0.1 - 1 % ;
  - $\pm 20 - 50\%$  at component level 0.01 – 0.1 % ;
- for the isotopic composition of Xe and Kr (if level  $> 1\%$ )
  - $\pm 1\%$  at component level  $> 10\%$  ;
  - $\pm 2\%$  at component level 1 - 10 % ;
  - $\pm 5\%$  at component level 0.1 - 1 % ;
  - $\pm 10 - 20\%$  at component level 0.01 – 0.1 %

The rod free volume is determined by backfilling it with a known quantity of inert gas while monitoring the corresponding pressure drop, i.e. using the same method as applied on the collection system. The accuracy of the void volume determination amounts to  $\pm 3\%$ .

The overall system of total gas measurement and void volume determination is controlled for its accuracy by puncturing a special fabricated, fuel rod simulating and known volume filled with air or inert atmosphere (Ar or He) at a known pressure.

The *density of fuel* is determined by a *mechanical vacuum mercury pycnometer* of own SCK•CEN design – fig. 6.



**Fig. 6:** Mechanical vacuum mercury pycnometer.

The remote friendly apparatus measures the volume of fuel samples. It essentially consists of an accurately reproducible sealed geometrical volume that contains a constant volume of mercury. After evacuation the geometrical volume is filled up (as indicated by a pressure transducer) by the penetration of a plunger, i.e. a metallic bar of exactly known and uniform cross-section (within  $\pm 1\mu\text{m}$ ). The plunger displacement is accurately measured (within  $\pm 1\mu\text{m}$ ) so that the specimen volume can be accurately determined by the difference of plunger travel when filling the "empty" and the sample containing specimen chamber. The uncertainty on the volume determination is estimated by a direct scientific approach, i.e. by the assessment of the different constitutive elements and features known to be the relevant main influencing factors:

- influence of the uncertainty of the sample chamber seal (on base of the design) =  $\pm 0.4 \text{ mm}^3$ ;
- influence of the uncertainty of plunger cross-section + plunger displacement =  $\pm 0.04 \text{ mm}^3$ ;
- uncertainty on the mercury volume and its thermal dilatation =  $\pm 0.08 \text{ mm}^3$ ;

leading to an overall estimated uncertainty of  $\pm 0.5 \text{ mm}^3$  ( $1 \sigma$ ), the reproducibility of the sample chamber seal being the dominant influencing factor. Thus for the capacity of the pycnometer, i.e.  $500 - 1000 \text{ mm}^3$ , the estimated uncertainty on the volume determination amounts to  $\pm 0.2 - 0.1 \%$  on the  $2 \sigma$  level. This performance is routinely checked experimentally by the measurement of control samples. These control samples are accurately machined cylindrical metal samples of uniform diameter ( $\pm 1 \mu\text{m}$ ) and accurately known length ( $\pm 10 - 30 \mu\text{m}$ , inclusive the parallelism of the end surfaces). The results of these experimental control measurements confirm the uncertainty as obtained from the above-described scientific approach - table 5. As such it is demonstrated that the relevant influencing factors have been analysed correctly and that they are under control.

Mechanical vacuum mercury pycnometer - control measurements								
Control samples								
ST 301 298.2 mm <sup>3</sup> $\pm 0.2 \text{ mm}^3$ (0.07 %)			ST 501 499.0 mm <sup>3</sup> $\pm 0.2 \text{ mm}^3$ (0.04 %)			ST 301 1000.7 mm <sup>3</sup> $\pm 0.3 \text{ mm}^3$ (0.03 %)		
Measurements								
Volume mm <sup>3</sup>	Accuracy		Volume mm <sup>3</sup>	Accuracy		Volume mm <sup>3</sup>	Accuracy	
	mm <sup>3</sup>	%		mm <sup>3</sup>	%		mm <sup>3</sup>	%
298.5	+0.3	+0.10	499.7	+0.7	+0.14	1000.6	-0.1	-0.01
297.9	-0.3	-0.10	498.6	-0.4	-0.08	1001.0	+0.3	+0.03
298.2	0.0	0.00	499.4	+0.4	+0.08	999.2	-1.5	-0.15
297.7	-0.5	-0.17	499.6	+0.6	+0.12	1001.0	+0.3	+0.03

**Table 5:** Quality control of the volumetric measurements as performed by the mechanical vacuum pycnometer.

The  $H_2$  content of Zry clad is determined at LHMA by a *hot vacuum extraction method* (RGA = residual gas analysis equipment). Small weighted pieces of clad (0.05 – 1 g) are raised to high temperature (~ 1200 °C) in vacuum. Under these conditions, the diffusivity of  $H_2$  becomes high enough to escape within 5 to 10 min. The released gas is transferred to an analytical volume equipped with an accurate pressure measurement system. It is analysed for its composition by a quadrupole mass spectrometer.

The quality of the hot vacuum extraction system is assured as follows:

- the background is carefully controlled and assessed (hydrogen being the main gas constituting the background in a high vacuum system) :
  - all parts are backable at 150 – 300 °C ;
  - an isolated (multi-)sample loading system is provided such that the analytical part don't has to be vented on sample loading ;
  - the vacuum is monitored by ionisation gauges ;
  - prior to the sample measurements the background is checked to be at a constant, reproducible low value (< 1  $\mu$ l / min)
- the sample outgassing is monitored in time in order to ensure complete release ;
- the detection system is calibrated
  - by the admittance of a known amount of gas through a build-in calibrated volume ( $44.2 \pm 0.1$  ml) ;
  - and the measurement of the corresponding pressure rise by the diaphragm Baratron gauge (independent of nature of gas – precision  $\pm 0.1$  % – resolution 0.001 mbar) ;
  - resulting in a calibration factor giving the amount of gas per unit of pressure rise ( $\mu$ l at STP / mbar) with an accuracy  $\leq 2$  % ;
  - the quadrupole mass spectrometer is directly coupled to the analytical volume and its calibration proceeds as described above in the puncture method.

The overall quality is controlled by the analysis of Ti standards with certified  $H_2$  content – table 6. The overall accuracy amounts to  $\pm 3$  %.

Hot vacuum extraction method – certified standard measurements			
Standard		RGA Measurement	
Type	Certified $H_2$ content ppm	ppm $H_2$	Accuracy
NBS 352	$32 \pm 2$ (6.3 %)	31.1	- 2.8 %
		32.5	+ 1.6 %
NBS 353	$98 \pm 5$ (5.1 %)	96.9	- 1.1 %
		97.3	+ 0.7 %
NBS 354	$215 \pm 6$ (2.8 %)	217.7	+ 1.3 %
		220.4	+ 2.5 %

**Table 6:** Validation of the hot vacuum extraction method.

### 2.3 *Microstructure examinations.*

Post-irradiation examination of nuclear fuel includes optical and electron optical investigation and analysis. These examinations are often related to an "open" questioning of nuclear fuel microchemistry and morphology. We are thus confronted with a situation where the analysis methodology and the instrument are to be assured rather than a specific method. This situation asks for a different approach which is much more open in nature than in the normal situation where a test method (oxide thickness probing, zircaloy hydrogen content, etc.) is to be validated. At present, we are preparing ourselves to work in accordance with the relevant ASTM guidelines regarding optical microscopic, electron microscopic and X-ray microanalysis guidelines.<sup>[2, 3, 4]</sup> These guidelines refer to rules for calibration and good practice of operation, but adaptations are necessary to make them applicable to the specific laboratory application.

The guidelines relevant for electron microscopic investigations refer to existing standards for calibration methods, while for optical microscopy, the magnification calibration standard will only be available by the end of this year. At the time being, we have prepared working instructions for sample preparation, calibration and data recording. Since we have recently moved from analogous data recording (photographic plate) to digital data acquisition, the procedures are currently being adapted before being finalised.

Digital data acquisition, processing and storage do modify the data stream significantly and on several levels. The process of data logging and ensuring that data storage is done systematically and safely (back-up!) becomes very important. In our case, the digital recording does involve the use of several software tools. As an example, for the scanning electron microscope, it involves the transformation of the initial image, as recorded on the microscope in a rectangular pixel format to one with a square pixel format, followed by a calibration of the image itself to allow for digital measurements.

The calibration of a scanning electron microscope is a two-step procedure, one regarding the resolution of the microscope, which varies with the instrumental settings such as working distance, beam current and accelerating voltage. The other is the calibration of the true magnification of an image, which again is a function of instrumental settings: the selected magnification (of course), but also the working distance and the accelerating voltage. For both calibrations, the use of an entirely digital system is a great advantage as compared to the analogous methods, insofar as reproducibility of the method is concerned. Standards for calibration of the concerned properties are available from NIST for the range of magnifications and the resolutions that are obtained by conventional electron microscopes. The problems related to quality assurance of electron microprobe analysis (EPMA) are at this moment being studied. Since at present there is no standard practice available from ASTM, one has to rely upon related analysis methods and procedures such as X-ray Fluorescence (XRF) and Energy Dispersive X-ray Spectroscopy (EDS). Working instructions for EPMA are at this moment under study at our institute.

### 3 Conclusion.

The validation of the test methods applied in the fuel research does not rely on one single procedure. A palette of different quality assurance techniques are implemented:

- the analysis of (certified) reference materials, which is the preferred method whenever possible
  - as applied on the fuel rod length measurement, the fuel rod diameter measurement, the eddy current outer clad corrosion measurement and the hot vacuum extraction hydrogen analysis of the clad ;
- the comparative approach, i.e. the assessment of the test method by comparing its results to those obtained by means of other independent methods ;
  - as applied on the  $\gamma$ -spectrometric analyses ;
- the direct scientific approach, i.e. the assessment of the relevant influencing factors, their uncertainties and the therefrom derived global uncertainty – combined with the analysis of control samples ;
  - as applied in the puncture test and in the density analysis by the mechanical vacuum mercury pycnometer ;

In addition the validation process is supported by the utilisation of direct and comparative calibrations and the exploitation of internal consistency checks.

As compared to the validation of the "exact" physical or chemical tests, the validation of the more "subjective" microstructure examinations focuses on the most critical factors affecting the results: sample preparation, resolution calibration, true image magnification calibration, digital data acquisition, processing and storage.

Method validation is often based on the combined use of several techniques such as to obtain optimal quality assurance justified on a cost-benefit basis while maintaining the fitness-for-purpose objective. In this way adequate laboratory services can be provided in accordance with scientific and industrial demands.

### 4 Recommendations.

The formal procedure-oriented quality systems (ISO 9000 family and/or the European Standard Series EN 45 000) do not automatically guarantee good results, or in other words accreditation does not necessarily lead to higher quality.<sup>[5]</sup> Indeed, the trueness of a laboratory result is significantly effected by differences in details of the (even same standardised) method applied at each individual laboratory. The only way to tackle this problem is validation of the method on base of certified reference materials. However, in many cases in the hot laboratory environment, certified reference materials are not available.

Many reference materials, produced worldwide, are certified by laboratory intercomparison, involving a large number of independent laboratories. Each of the participating laboratories measures the requisite property on different samples, with each sample measurement consisting of a number of independent repeated observations. It is then assumed that the differences between individual results, both within and between laboratories, are all of a statistical nature regardless of their causes. Hence, the certified value and its

uncertainty are estimated on the basis of an analysis of variance, after verification that all data belong to the same normally distributed population. This procedure finds its justification in the fact that one presumes that, if a large variety of independent laboratories and methods is used, possible systematic effects in the individual laboratory results will be "randomised" and that, eventually, both the residual systematic error and its uncertainty are reduced to zero.

Therefore, co-operation and interlaboratory comparisons are recommended in order to strive after a maximum reliability of the hot-laboratories analyses. The most obvious way is to organise proficiency tests, with the participation of a large number of independent laboratories, and including the use of reference materials and reference methods. The European Working Group on "Hot Laboratories and Remote Handling", bringing together a large number of independent laboratories, might be the appropriate forum for such co-operation.

## 5 References.

- [1] S. Van Winckel, M. Gysemans, L. Vandeveldel, "How to apply 'Quality' in Nuclear Analytical Chemistry: an Illustration", this plenary meeting of the European Working Group on "Hot Laboratories and Remote Handling".
- [2] ASTM E 883 Standard guide for reflected-light photomicrography.
- [3] ASTM E 986 Standard Practice for Scanning Electron Microscope Performance Characterisation.
- [4] ASTM E766 Standard Practice for Calibrating the Magnification of a Scanning Electron Microscope.
- [5] R.G. Visser, "Is accreditation useful for quality improvement ?", *Accred. Qual. Assur.* (1999) 4: 108-110, Springer-Verlag 1999.

## 6 List of abbreviations

amu	atomic mass unit
BWR	Boiling Water Reactor
FG(R)	Fission Gas (Release)
IRMM	Institute for Reference Materials and Measurements
LHMA	Laboratory of High- and Medium-level Activity
PCMI	Pellet-Clad Mechanical Interaction
PIE	Post-Irradiation Examination
PWC-CCD	Pressurised Water Capsule – Cycling and Calibration Device
PWR	Pressurised Water Reactor
RGA	Residual Gas Analyser
RMO	Reactor Materials Research department
SCK•CEN	Nuclear Research Centre
STP	Standard conditions of Temperature (0 °C) and Pressure (1 atm)
TIMS	Thermal Ionisation Mass Spectrometry