RADIOMETRIC INSTRUMENTATION FOR SPENT FUEL MONITORING.

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ABSTRACT

The background to spent fuel measurement techniques and their application to burnup credit is presented. Options for the radiometric measurement of burnup are discussed as well as an alternative approach which could be used to directly measure the reactivity of spent fuel. Data is presented of burnup measurements on a campaign of 75 PWR fuel assemblies. Finally two monitoring system configurations are described which can incorporate a selection of radiometric techniques for the measurement of burnup or reactivity of spent fuel at a reactor site.

INTRODUCTION

Radiometric monitoring of spent nuclear fuel is an established accompaniment to many spent fuel handling processes. The monitoring is performed to determine fuel history parameters such as burnup, cooling time, initial %wt. ^{235}U enrichment and final %wt. ^{235}U enrichment equivalent. It is often very important that there is a high level of confidence in the accepted values of these parameters and measurement rather than the use of reactor operator records is the preferred method for their determination or confirmation. The parameters, which have a bearing on the fuel radionuclide content, heat output and fissile material content, are required because of their impact on the handling procedures and the costs during operations such as storage, transport, reprocessing and disposal. As a result considerable progress has been made by a variety of establishments in Europe, USA and Asia in developing spent fuel monitoring systems. These have been generally based upon radiometric analysis of passive gamma and neutron emission and active neutron measurement.

The applications for spent fuel monitoring include; (i) safeguards, principally for the determination of fissile material content, particularly fissile plutonium, (ii) process control, for example in the Thermal Oxide Reprocessing Plant at Sellafield, to provide fuel history parameters to ensure only "acceptable" fuel in accordance with flowsheet criteria are submitted into the plant for reprocessing and (iii) burnup measurement for burnup credit applications in areas of spent fuel storage, transportation and disposal.

As a result of the wide range of applications and requirements for spent fuel monitoring, BNFL Instruments Ltd is currently developing a series of modular spent fuel monitoring systems. This series is based on instrument systems which have so far monitored well in excess of one million fuel items in process control applications in the Sellafield facility.

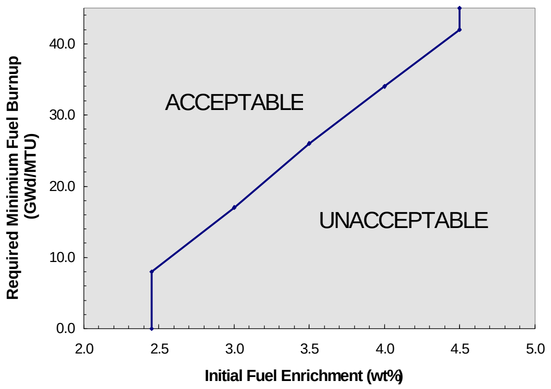
Internationally, burnup measurements on spent fuel are considered to have significant economic benefits derived from taking credit in nuclear safety assessments for the loss of fissile content and reactivity due to burnup. The emphasis in this paper is on those measurement techniques and monitoring systems that are relevant to burnup credit.

BURNUP CREDIT REVIEW

The justification for the use of radiometric monitoring systems for burnup credit appears well advanced in the USA and Japan where burnup credit has already featured in the design of spent fuel transport casks. In the USA, a Department of Energy topical report on a proposed methodology for the use of "actinide only burnup credit" is under review by the Regulators[1]. This methodology covers the measurement of spent fuel and the validation of procedures covering all forms of data that may be used to allow the application of burnup credit to the storage and transportation of spent fuel. It is clear from this document that the measurement of burnup to good accuracy and precision is crucial because of the conservative manner in which any measurement data must be treated. This treatment dictates that any unknowns in the form of measurement errors are cumulative and are subtracted from the measured burnup before it can be used for burnup credit. The requirement for high confidence in the measurement data may lead to a need to have diverse measurements of burnup, i.e. the use of two or more independent techniques.

The concept of “burnup credit loading curves” is described in the topical report. These provide a means of segregating fuel assemblies into “specified” assemblies, that meet the acceptance criteria for loading into a fuel storage rack or transport cask designed to take account of burnup credit. “Non-specified” assemblies are those which do not meet the criteria. The criteria are based on a combination of the fuel burnup and initial ^{235}U enrichment. An example of a loading curve is presented in Figure 1.

Figure 1 Typical fuel loading curve[1]



Built into the curves is a bias to account for any uncertainties in the data that relate burnup to the reactivity of the spent fuel as well as a contingency bias, to minimise the possibility of “overloading” a package, as required by safety case studies. In addition the measurement errors associated with monitoring spent fuel are taken into account in the accredited burnup value for comparison with the curves.

The case for taking credit for the reduced reactivity of spent fuel assemblies due to the burnup appears very strong. The contingencies required by the “fresh fuel assumption” that spent fuel is as reactive as when initially manufactured, leads to several tens of millions of dollars ‘added’ cost to the fuel handling programmes. The added cost comes from either having to maintain lower packing densities in transport flasks and storage pools or having to provide a means of neutron absorption using boron or gadolinium. An overall saving of 25% to 40% is anticipated for storage, transport and disposal costs with the application of burnup credit. With more than 100 000 fuel assemblies currently in storage pools in the USA alone, rising to over 250 000 over the operating lifetime of the US commercial reactors, the potential savings are very significant.

Monitoring of spent fuel assemblies for the purpose of burnup credit would be carried out, predominantly by the utilities, to allow greater packing densities within their storage pools, dry storage casks or transport casks. Monitoring system options to be operated by a utility include those where fuel is monitored by raising an assembly through a monitor collar positioned just above a fuel storage rack within a pool. This would enable measurement of an assembly without necessitating its complete withdrawal from the storage rack. This approach requires a mobile assay system which imposes additional constraints on its design. Alternatively monitoring can be carried out in a dedicated monitor station located, for example, between the fuel storage pool and a fuel loading facility immediately prior to loading into a different storage configuration or into a transport cask for transfer to a waste repository. Attention to the requirements of the operator with regard to space constraints, interface and operator control protocol, performance and throughput is essential if monitoring is to be satisfactorily integrated with a fuel loading procedure. All the above factors impact on the design of a monitoring system and will influence the selection of measurement techniques and processing algorithms.

Measurement Techniques. There are various techniques available for the measurement of burnup, although direct measurement of burnup can take place only when the fuel is resident in a nuclear reactor. This direct measurement is achieved by the use of in-core flux probes comprising ion chambers for gamma measurement and fission chambers or flux wires for neutron measurements. When used with interpolation codes to represent the power density distribution across the reactor core the burnup for individual fuel assemblies can be given to ±2%.

The measurement of burnup in spent fuel is generally by an indirect measurement made after discharge from a reactor. The time since discharge, or the cooling time, at which measurements are likely to be made is about 20 years as this is the average cooling time for current US fuels, with the majority falling into the range 5 to 40 years. Indirect determination of burnup is by the measurement of spent fuel parameters which can be correlated with burnup. These parameters, often referred to as “burnup indicators”, are individual or combinations of radiation emissions from radionuclides built up during irradiation. Appropriate gamma emitting radionuclides can include; ^{134}Cs, ^{137}Cs, ^{154}Eu, ^{144}Ce, and ^{106}Ru whilst the neutron emission is primarily from ^{244}Cm. Some of the relevant properties of these nuclides are listed in Table 1

Table 1 Some Properties Of Burnup Indicators

Nuclide *Mode Of Decay*	Half-Life *Specific Activity (Bq/g)*	Principal Gamma Rays (keV)	Branching Ratio %	Spontaneous Fission Rate (sf/g/s) *Fast neutrons per fission*
(Ce-144) *b-*	285d *1.2E+14*	134 2186(Pr-144)	11 7.7	
Cm-244 *a*	18.1y *3.05E+12*	100	0.0015	4E+6 *2.84*
Cs-134 *b-*	2.062y *4.79E+13*	604.7 795.9	97.6 85.4	
Cs-137 *b-*	30.0y *3.22E+12*	662(Ba-137m)	85.2	
Eu-154 *b-* ε (0.2%)	8.8y *9.77E+12*	723.3 873.2 1004.8 1274.5	19.7 11.45 17.9 35.5	
(Ru-106) *b-*	1.02y *1.2E+14*	512(Rh-106) 622(Rh-106) 1059(Rh-106)	20.7 9.8 1.7	

The measurement techniques employed include; total gamma counting, low resolution gamma spectrometry (LRGS), high resolution gamma spectrometry (HRGS) and passive neutron counting.

The selection of techniques for use in any particular burnup monitoring system should be made by reference to the required measurement performance. This may be expressed as; (i) the required level of confidence in the measurement results. Greater confidence may be achieved by incorporating more than one measurement technique in a system to give diversity. (ii) the measurement accuracy. This may influence the type or number of radiation detectors in a system and the sophistication of the mechanical arrangement, (iii) the acceptable measurement time. This will depend on the planned rate of fuel throughput in a plant or process and multiple systems operating in parallel may be necessary, (iv) the acceptable “degree of blindness”. The degree of blindness, described by N B McLeod[2], indicates the extent to which utility operator declared data can be used to assist the processing of the measurement data in the determination of burnup. Minimal assistance would be when only the fuel type, i.e. PWR or BWR would be used as input data into a calibration and measurement procedure. The other extreme is where there is a high level of assistance or reliance placed on operator data. In the case of passive neutron measurement the operator declared cooling time, initial ^{235}U enrichment and burnup are used to adjust the measured neutron count rate before correlation with the declared burnup. The “measurement” in this procedure acts as a confirmation of the reactor record data by checking for outliers in the fit between the count rate and the burnup.

Other factors to be considered in the selection of techniques are the cost of the system and the ease with which it can be installed into a plant. Early planning for the inclusion of a measurement system in the design stage of a facility can substantially reduce costs for the installation and operation of a system of optimum design to meet a particular measurement requirement.

Algorithms to correlate the measurable parameters with burnup make use of:

(i) *The absolute count rate of the 662 keV gamma ray from ^{137}Cs.* This technique is attractive because of the simple linear relationship between the activity of ^{137}Cs in spent fuel and burnup. This is because ^{137}Cs is a direct fission product and has an almost equal fission yield from uranium and plutonium. Also ^{137}Cs has a half life of 30 years, which renders its production insensitive to variations in reactor power rating and dwell time and to errors in its cooling time correction needed to account for its decay between the time of discharge from a reactor and the time of measurement. However as it is an absolute measurement technique there must be a well defined and reproducible geometry between the detectors and the fuel assembly. The linear relationship is as follows;

$$^{137}\mathrm{Cs} = a + b.\mathrm{BU}$$

where ^{137}Cs is the count rate of the 662 keV gamma ray corrected to zero cooling time,
a and b are constants in the linear correlation with burnup BU.

(ii) *The gamma measurement of the activity ratio ^{134}Cs / ^{137}Cs.* The ratio technique has an advantage over an absolute measurement because of its insensitivity to measurement geometry. Some correction for relative detection efficiency as a function of energy is required. Disadvantages are (a) the ratio has a 2.2 year half life and needs a significant cooling time correction, (b) its correlation with burnup is influenced by the initial ^{235}U enrichment and power rating and (c) its application is limited to fuel well below 20 years cooling due to the decay and disappearance of the shorter lived component. The relationship with burnup can be approximated to a linear function;

$$^{134}Cs/^{137}Cs = c(en, r) + d(en, r).BU$$

where $^{134}Cs/^{137}Cs$ is the zero cooled activity ratio,
c and d are coefficients which depend on enrichment "en" and power rating "r".

(iii) *The gamma measurement of the activity ratio* $^{106}Ru \times {}^{137}Cs/({}^{134}Cs)^2$. This has the advantage of being an activity ratio method, but unlike the $^{134}Cs/^{137}Cs$ ratio is virtually independent of enrichment and rating and is therefore subject to lower systematic errors. The half life of the ratio is 22 years, and therefore it has a lower sensitivity to cooling time correction errors. However due to decay of the short half life component ^{106}Ru this ratio can only be used on fuel up to a cooling time of 8 or 9 years. The relationship with burnup has the following form;

$$Ln(R_0) = e + f.Ln(BU)$$

where $Ln(R_0)$ is the natural log of the cooling corrected ratio $^{106}Ru.^{137}Cs/(^{134}Cs)^2$,
e and f are coefficients.

(iv) *The measurement of the passive neutron emission*. For fuel of greater than 15GWd/Te(U) burnup and more than 2 years cooling the primary spontaneous fission neutron emitter is ^{244}Cm. However, the measured neutron flux also depends on the fuel assembly's fission product neutron poison content and its neutron multiplication due residual fissile content. The advantages of this passive neutron measurement approach are; (a) it is a very strong indicator of burnup with the neutron emitting ^{244}Cm content proportional to the fourth power of burnup, (b) neutrons emitted from a fuel assembly in water cascade through the assembly by <u>induced</u> fission reactions to reach the external detectors. This is true for neutrons from central pins in the assembly as well as those from pins near the outside of the assembly. The measurement therefore represents the bulk of the fuel assembly in contrast to the outer two or three pins that are "visible" by a gamma measurement, (c) ^{244}Cm has a relatively long half life of 18.1 years with its associated reduction in sensitivity to cooling time correction errors. The disadvantages are (a) the quantity of ^{244}Cm produced during irradiation is strongly dependent on its ^{235}U initial enrichment, (b) the measurement of the neutrons is very sensitive to the geometry or water gap between the fuel and detectors and to the presence of any neutrons poisons in the pool water or within the fuel itself, (c) the measured neutron flux is influenced by neutron multiplication. The relationship between neutron count rate and burnup has the form;

$$Ln(n_0) = g(en) + h(en).Ln(BU)$$

or

$$n_0 = i . Bu^j$$

where n_0 is the cooling corrected passive neutron count rate,
g and h or i and j are the coefficients which depend on initial enrichment.

BURNUP MEASUREMENT PERFORMANCE IN THE THORP FEED POND FUEL MONITORS

The Thermal Oxide Reprocessing Plant (THORP) at Sellafield has two spent fuel monitors called the Feed Pond Fuel Monitors (FPFM)[3,4]. These operate in parallel in order to meet the throughput requirements and measure a number of fuel parameters to ensure that only those fuel assemblies within prescribed flowsheet limits enter the Head End of THORP for reprocessing. The flowsheet limits relate to the minimum cooling time and maximum burnup and initial ^{235}U enrichment for both light water reactor fuel (LWR) and advanced gas reactor fuel (AGR). The measurement techniques employed by the FPFM are high resolution gamma spectrometry (HRGS) and passive and active neutron counting by the use of an external ^{252}Cf interrogating source. The burnup measurements utilise the HRGS and passive neutron counting using algorithms similar to those described above. The required cooling time and initial enrichment corrections for burnup determination use measured values from the HRGS and active neutron measurements respectively. In this way the FPFM determines burnup with the minimal use of operator declared data. Only the fuel type is used to aid the measurement by the selection of an appropriate calibration.

Approximately 1000 AGR cans and 700 LWR assemblies have been measured since the start of commissioning measurements in 1993. A recent campaign of 75 PWR assembly measurements have been selected to illustrate the burnup measurement results. The range and average values of irradiation, cooling time and initial enrichment of this campaign are respectively; 15 to 34 GWd/Te(U) - average 25.4 GWd/Te(U), 2600 to 7000 days - average of 5565 days (15 years), and 2.5 to 3.4% - average 2.90%. Very few measurements, in this campaign made use of activity ratio techniques because the campaigning programme concentrated on the longer cooled fuel. Results from the absolute ^{137}Cs and passive neutron techniques only will therefore be presented.

Absolute ^{137}Cs Measurements

Measurement of LWR fuel assemblies are made at several positions along the assemblies as they are lifted past the monitoring position. These measurements can be correlated with local values of burnup calculated from the declared assembly average and axial profile factors. The factors are determined experimentally during system calibration. Figure 2 shows a plot of zero cooled 662 keV count rate from ^{137}Cs against the local declared burnup. Figure 3 shows assembly average ^{137}Cs gamma count rate against the declared assembly burnup

The reduced scatter on the plot in Figure 3 compared to that in Figure 2 is due to the use of the mean value from all the measurements on the assembly. The fit gives a standard error in the burnup of less than 2% for 25 000 MWd/Te(U). This error is close to the accuracy of the declared burnup which is also about 2%.

Figure 2 Zero cooled ^{137}Cs 662 keV gamma count rate vs. the local declared burnup (MWd/Te(U)).

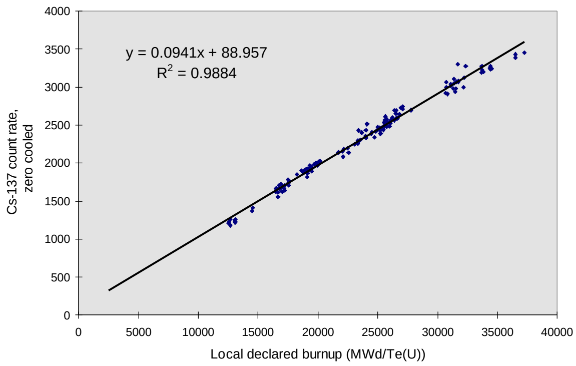
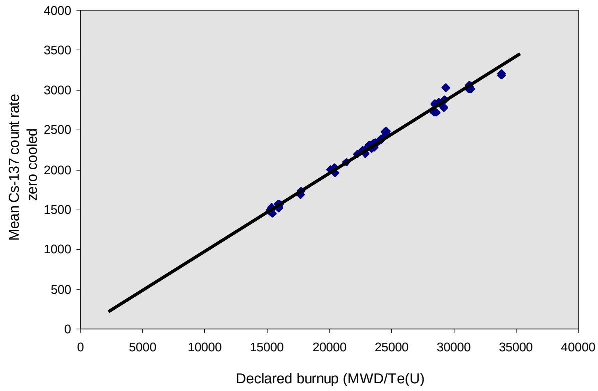


Figure 3 Zero cooled assembly average ^{137}Cs 662 keV count rate vs. declared burnup (MWd/Te(U)).



Passive Neutron Measurements

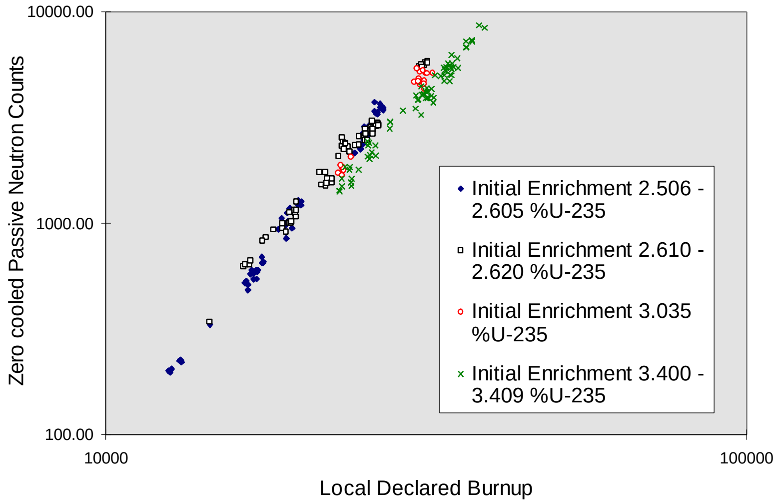
The passive neutron count rate from the fuel plotted against the local declared burnup is shown in Figure 4. As indicated there is some dependence on enrichment but this is not as strong as suggested by fuel burnup codes such as ORIGEN2 or FISPIN[5]. This dependence on enrichment, with lower initial enrichment giving more ^{244}Cm per unit irradiation, is likely to be offset by an increase in the production of neutron poisons in the fuel per unit irradiation as well as a decrease in multiplication. The variation of neutron count rate with burnup gives an average exponent "j" of 3.5 in the expression $n_0 = i \, . \, Bu^j$; this is consistent with other reported values[6].

The standard error of the fit between passive neutron count rate and burnup gives an error of less than 5%, with no enrichment correction, for a burnup of 25 000 MWd/Te(U) and less than 4% with an enrichment correction.

BURNUP CREDIT MEASUREMENTS - AN ALTERNATIVE APPROACH

The standard justification for taking credit for the reduction in fissile content and reactivity of spent fuel is by the indirect process of measuring burnup. However the key factor in burnup credit is the loss of reactivity of the spent fuel not the value of burnup. It would be useful, therefore, to measure directly the reactivity of the spent fuel. This could then be used with a new form of loading curve to translate the reactivity in the measurement configuration to that in any particular storage or transport configuration.

Figure 4 Zero cooled passive neutron count rate vs. local declared burnup(MWd/Te(U)).



There are a number of measurement techniques that could be used to determine reactivity of spent fuel. These include:

(i) Measurement of the induced fission neutron flux from the application of an external neutron interrogation source such as ^{252}Cf. This is the approach used on the FPFM to give a parameter related to reactivity, the neutron multiplication, for determination of the residual ^{235}U equivalent enrichment.

(ii) Measurement of the neutron flux decay rate following interrogation of the residual fissile material with a pulsed D-T tube neutron generator.

(iii) Measurement of the change in reactivity by perturbing a spent fuel assembly in a neutron multiplying system. This can be correlated with the intrinsic multiplication and the burnup. A variant of this technique called "self interrogation" does not require an external source of neutrons. Instead the source of neutrons are those emitted by the ^{244}Cm within the fuel assembly. This may be particularly important for high burnup or mixed oxide (MOX) fuels which have a high ^{244}Cm content.

Any movement away from burnup measurements towards the more direct reactivity determination of spent fuel would require regulatory support and approval.

DISCUSSION AND CONCLUSIONS

As discussed above there is a wide range of applications and requirements for fuel monitoring associated with spent fuel handling. In particular strong economic arguments exist for measurements in support of burnup credit applications.

Both gamma and neutron measurement techniques used on the THORP plant have provided reliable values of burnup for PWR fuel. The results from the ^{137}Cs gamma ray approach consistently give a lower error than those from the neutron measurements. This is due to the inherently lower sensitivity of the ^{137}Cs determination to systematic effects.

In general the technique selection for burnup credit measurements is likely to be made on the basis of a number of criteria relevant to the specific application. The criteria will be related to fuel history, performance, cost, and mechanical installation constraints.

The installation constraints will influence the way in which a system is deployed and operated and therefore the choice of system configuration for a particular facility. A key factor is whether the fuel measurements are to be made above a storage rack by lifting the fuel into the monitor without its complete removal from the rack or whether the measurements are to be made at a location remote from the fuel storage rack in a pool wall mounted arrangement. Examples of monitoring system configurations are presented in Figures 5, and 6. Figure 5 is a pool wall configuration whereas Figure 6 shows a configuration for monitoring fuel above a rack.

Both the above rack type and wall mount configurations incorporate facilities to accommodate any of the techniques listed above. The common features of the systems are (i) the re-entrant tubes which allow the insertion of the detectors into their respective monitoring positions deep into the pool at the monitoring height, (ii) the horizontal collimator between the fuel position and the gamma detector in its shielded housing, (iii) the monitoring collar around which neutron detectors and if required a neutron interrogation source can be placed close to the fuel position.

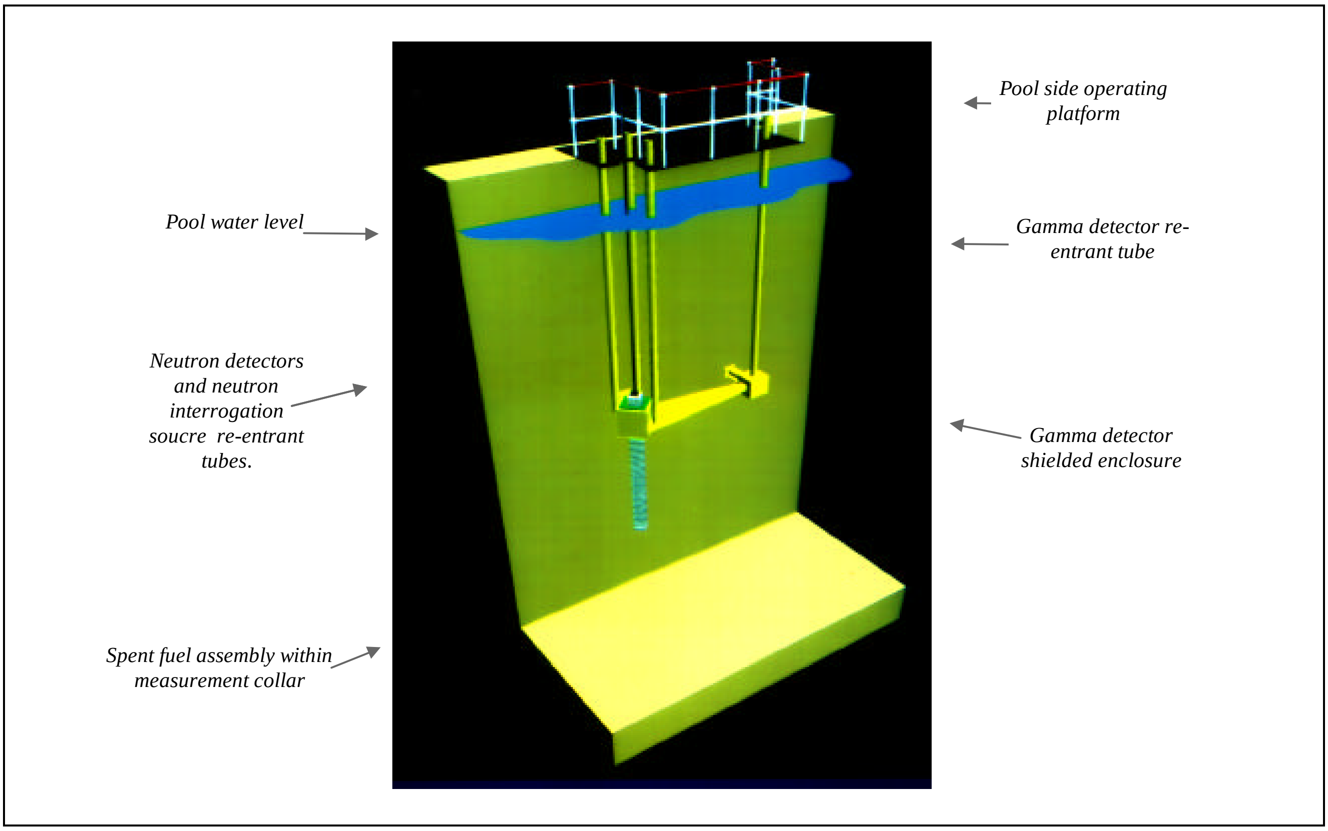
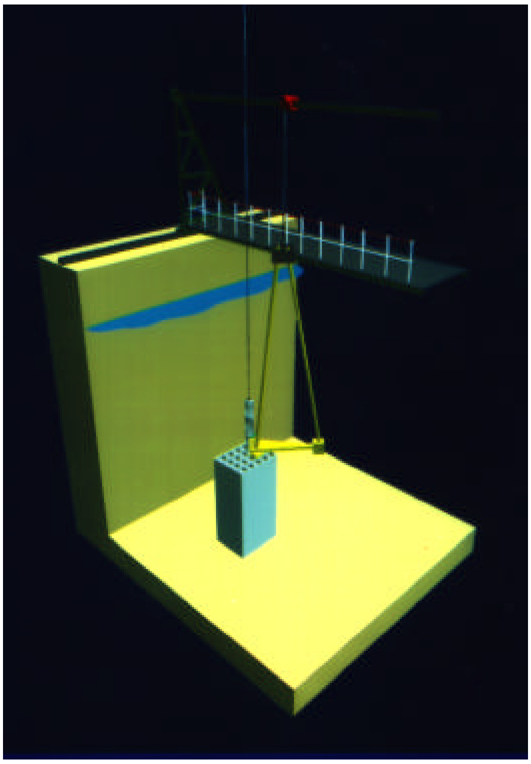


Figure 5 Pool wall location monitoring system configuration

Figure 6 Rack location montoring system configuration



A prototype of the pool wall configuration, using a HRGS system, is currently undergoing commissioning trials in the pool of a US utility with the intention that several hundred assemblies are to be measured over the next few months. This will provide an effective demonstration of burnup credit measurements as part of the operations at a reactor site.

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