Development of Gamma Scanning Applications in Post Irradiation Examination

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1. Abstract / Introduction

Gamma scanning as a Post Irradiation Examination (PIE) technique has existed for many decades. Modern developments, including increase in efficiencies of detector and more compact design with convenient electric cooling, has allowed gamma scanning to remain at the forefront of PIE techniques.

Whilst the majority of non-destructive PIE techniques provide only visual details on the surface of a nuclear item, gamma scanning is a powerful non-destructive PIE tool. It offers the capability to 'see' the location of nuclear material within an item as well as understanding the detail of the isotopic distribution in the X, Y and Z planes. This paper discusses how the capability can be optimised through different collimator designs, how it can be utilised for fission gas and burn up predictions in fuel and looks at detailed isotopic mapping of nuclear items. Further applications for this area may in future include enhancing the capabilities of non-destructive characterisation, and applications in safeguarding and examination of fuel after extended periods of underwater storage.

2. Gamma Spectrometry as a Post Irradiation Examination Technique

The key aspects of gamma spectrometry for PIE are:

- Passive technique with penetrating radiation: requires no source other than the subject material and is forgiving of environment (e.g. underwater).
- Emitted radiation is specific to isotopic composition, permitting a range of quantities to be observed.
- Equipment is relatively compact with efficient cooling systems now available.

Two main applications of gamma spectrometry to spent nuclear fuel are:

1. On site spectrometry (at station or spent nuclear fuel storage facility).
2. Condition monitoring of fuel during PIE, at dedicated hot cell facilities.

With on-site spectrometry, examination is frequently limited to complete assemblies. This has given rise to the use of tomography in order to provide more rod-specific information. Holcombe et al1 describes a system for Halden Boiling Water Reactor (HBWR) for performing tomography of whole assemblies. In this case, the assembly is scanned in-air. The measurements were sufficient to allow conclusions on the rod-wise power distribution and fission gas product distribution.

In pond scanning of assemblies (without use of tomography) has been more widely used, as the simpler assembly handling arrangements required make this more practical than tomography. Of particular note is the use in Swedish reactors and at the Swedish CLAB facility, where scanning of assemblies in pond has been applied for (amongst other uses) safeguarding purpose2. In these cases the pin is viewed through a collimator arrangement in the pond wall.

CLAB measurements show the ability to assess cooling time and burn-up of assemblies based on developing fits to a “control group”. This is most useful for identifying deviations to normal irradiation conditions, or assemblies which are not as described (e.g. for safeguards purposes). As described, it does not allow determination of the burn-up/cooling time of an arbitrary element. Also, it is not necessarily sensitive to missing pins because of the self-shielding effects of the assembly.

In contrast to the in pond and pond-side inspections above, gamma spectrometry applied in hot cells provides the opportunity for a detailed examination of individual fuel pins, but at generally longer cooling times. The focus of the current work is the use of gamma spectrometry in the hot cell environment, with data presented from developments to the hot cell technique.

3. NNL Gamma Scanning Capability

The equipment installed within the Windscale active handling facility (AHF) (Figure 1) is operated by passing, and rotating, the entire length of an Advanced Gas-cooled Reactor (AGR) fuel pin past a gamma spectrometer mounted in the cave wall. One of three Tungsten collimators (0.1, 0.2 or 0.3 mm) is used to restrict the signal reaching the detector and improve the resolution.

The gamma detector electronics, cooling and data acquisition system plus the control system are mounted at the cave face (outside wall of the cave). Although the primary use of the rig is to undertake axial scans of AGR fuel pins, adaptations can be made to accommodate a range of other materials including graphite sleeves and cross sectional pieces of fuel pins.

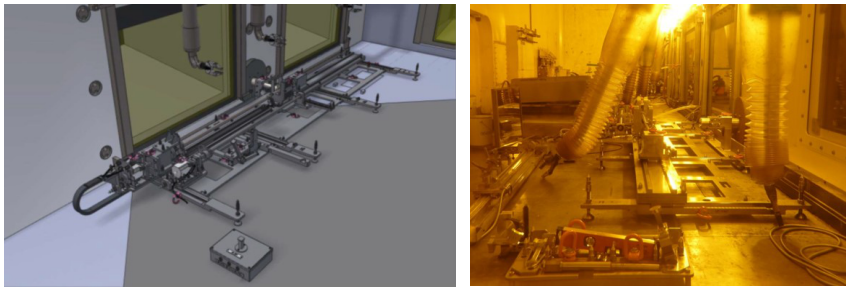

Figure 1. Computer representation and in-cell image of gamma spectroscopy equipment

Gamma spectroscopy is currently used in two different ways within AHF. The first is a long spectral scan of a point on the fuel stack, used to determine the inventory, quantity and location of the gamma emitting isotopes present. The second is to produce a profile of the isotopes along the length of the fuel pin, generated by taking a series of point short spectra scans spaced at small intervals. The intensity of each isotope of interest is then plotted against the position on the pin.

Good resolution is required for positional measurements, both in the features to be examined and energy spectra. Therefore collimation is important. The design of the collimator needs to match the requirements for the sample; allowing an optimal gamma flux to reach the detector, focusing on the area of interest in the sample and reducing the background signal from other sources. Consideration of the construction material, aperture size and design can help achieve this.

The width of the collimator slit is a compromise between increasing counts to enhanced peak resolution (requiring a wide slit) and increasing spatial resolution (requiring a narrow slit). As this balance between count and spatial resolution is dependent on the sample activity, being able to vary the collimator slit size is useful. Historically, the AHF has used a mechanically variable collimator which could be precisely tuned; however reproducibility, precise collimator aperture and periodic servicing issues must be considered.

In place of a variable collimator, a series of interchangeable collimator modules have been developed in the current capability. These can be mounted remotely and allow a limited number of precise collimator slit widths to be set. A second set of collimator inserts with small rectangular apertures have also been created to allow measurements of flat surfaces, for example the metallography sample shown in Figure 2. The flatter surface becomes a point source, allowing two dimensional maps that would not be achievable with the slit formation.

The collimator inserts are machined from tungsten, allowing good attenuation without requiring the same thickness as other alternatives. Additional desirable properties of tungsten are its low reactivity, low toxicity, good stability to high temperatures and (although the mechanical properties make it difficult to machine) minimal deformation during remote handling.

4. Non Destructive Measurements using Gamma Scanning

Measurement of fission gas non-destructively is possible, making use of the isotope ^{85}Kr (half-life 10.78 years). However, a number of factors make use of this isotope difficult:

- ^{85}Kr emits gamma rays at an energy of 514 keV, which is adjacent to two peaks of generally higher intensity; the 511 keV annihilation peak and a peak from decay of 1096Rh at 512 keV.
- Measurement of ^{85}Kr in a region of the fuel stack is confounded by the presence of fission gas held within the fuel matrix as well as that released to the atmosphere.

In the case of LWR fuel, which generally contains an end plenum, it is feasible to assess fission gas release from the direct evaluation of 85Kr activity in the plenum.

For AGR fuel, there is no end plenum and thus no ability to perform measurements of 85Kr in a region absent of fuel. It is therefore necessary to perform assessment of fission gas release indirectly. In addition to the fission gasses xenon and krypton, volatile elements such as caesium and iodine are sensitive to relocation in the fuel matrix at elevated temperatures and burn-ups. The vapour pressure of caesium is ~ 1 atmosphere at 690 °C; previous studies of LWR fuel, conducted by Walker3 have found caesium to migrate through the fuel in a similar way to fission gas (Xenon). Gamma spectroscopy reveals the occurrence of caesium deposition at the ends of the fuel stack and also at pellet-pellet interfaces.

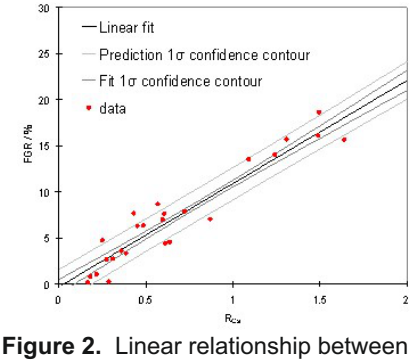

Figure 2. Linear relationship between FGR and the relocation of caesium

A total of 27 UK AGR fuel pins for which the FGR was measured by pin puncture were gamma scanned and the intensity of the end cap caesium measured. The intensity values are compared to fission gas release in Figure 2. A linear fit is applied to the data; the relationship between the calculated gamma spectroscopy based parameter and the pin puncture measurements of FGR is sufficient to permit prediction of puncture FGR using gamma spectroscopy profiles. As an example, the technique predicts FGR of 10 % with a 1 σ confidence level of ±1.8 % absolute. This accuracy deteriorates rapidly for FGR of less than 5 %.

Non-destructive burn up determination using fission products determined from gamma spectroscopy is also possible. Literature almost solely reports the use of a small number of isotopes, namely ^{106}Ru,^{134}Cs, ^{137}Cs and ^{154}Eu, when determining burn-up from gamma spectroscopy data.

It is also worth noting that, when the power distribution in a core shifts during irradiation or if fuel elements are moved in the reactor mid irradiation, the short-lived and long-lived nuclides will differ in their distribution. The long-lived nuclides such as ^{137}Cs and ^{154}Eu retain their whole irradiation history, while a short-lived isotope such as ^{137}Cs reflects the more recent fuel history. .Data from fuel cooled for > 9 years is required in order to fully investigate the accuracy and limitations of the different methodologies, before any robust conclusions can be drawn. It is worth noting that issues may arise with redistribution of fission products to the "cooler" locations in the pin i.e. pin ends, but this is unavoidable in reactor service conditions. A summary of the different techniques for ND burn up determination is shown in Figure 3.

TECHNIQUE	ADVANTAGES	DISADVANTAGES
Absolute count rate of the 662 keV gamma ray from ^{137}Cs..	-Simple linear relationship between ^{137}Cs and burnup. -Half life of 30 years. -Insensitive to variations in reactor power rating and dwell time.	-Absolute measurement requires a well defined and reproducible geometry between the detectors and the fuel assembly.
The nuclide activity ratio: ^{134}Cs/^{137}Cs.	- The ratio method makes it insensitive to geometry.	-2.2 year half life requires significant decay correction and can only be applied to fuel with cooling time < 20 years. - Burnup correlation is dependent on initial enrichment and power rating.
The nuclide activity ratio: ^{106}Ru x ^{137}Cs/(^{134}Cs)^2 .	- Insensitive to geometry. - Independent of enrichment and rating.	- Only useful for fuel < 9 years cooling time (^{106}Ru has a 372 day half life).
Passive neutron measurement (predominantly from ^{244}Cm).	- The neutron signals are received uniformly from all pins in the assembly (gamma measurements are only sensitive to the outer pins). - Good for safeguards applications, as it is sensitive to missing or removed fuel pins.	- ^{244}Cm is a strong function of initial enrichment. - Neutron assay is very geometry sensitive and can also be affected by multiplication and neutron poisons in the pool or within the assembly.

Figure 3. Extract from Spent Fuel Measurements in Support of Burn-up Credit comparing different methods of calculating burn-up[4]

5. Mapping of fuel

Cross sections of AGR fuel pins, shown in Figure 4, are routinely taken for metallographic examination. A hollow bore fuel pellet is surrounded by stainless steel cladding. A characteristic of the cladding is the machined profile to improve the efficiency of heat transfer between the oxide fuel and the gas coolant. Radial cracks can be seen extending across the pellet.

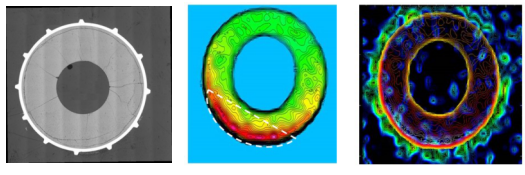

Figure 4. L to R Cross section of AGR fuel; contour map of ^{137}Cs; contour map of ^{60}Co

The mounted fuel sample was scanned with the X, Y, Z table similar to that shown in Figure 1. AGR fuel has a unique burn-up tilt, Figure 3 clearly shows the correlation between the higher 137Cs activity and the side of the fuel with the higher burn-up. These initial results indicate that this technique can distinguish this burn-up tilt when applied to a cross section of a fuel pin. As the collimation used in this work is a slit, consequently it gives the appearance of a false ovality at the fuel bore. Further developments will assess the use of a pin point aperture, along with improving the imaging capability and data analysis route.

This technique gives an insight into isotopic distribution over a large surface area which can usually only be obtained with techniques such as shielded Electron Probe Microanalysis (EPMA). Whilst the resolution achieved through EPMA will be higher than that from gamma scanning, gamma scanning offers an alternative technique when analysing samples with difficult or unique geometry. For example due to the nature of EPMA, careful positioning of the beam is required to avoid porous or cracked areas of the fuelxxvii, whereas this is not an issue with gamming scanning. However, the fine collimation required combined with low gamma emissions from such a small area, would prevent identification of isotopes specific to, for example, a grain boundary region.

The gamma scanning of fuel cross sections also provides the capability to determine isotopic distribution across the diameter of the pin, including studying fuel near the central bore. A standard fuel pin gamma scan taken perpendicular to the pin is unable to provide this detail. This ability to map isotopic distribution on a relatively small scale, in this case 137Cs and 60Co, can be used to gather additional data to support visual examination work. With no recently published estimates of instant release fraction (IRF) available for UK fuel types such as AGR , this technique may also be used to support applications such as the modelling and determination of instant release fractions for disposal scenarios (radiotoxic assessments). It could also be used for Nuclear Forensics and safeguarding purposes.

6. Conclusions and future developments

Applications of data from gamma scanning assessments of spent nuclear fuel include:

- Assessing fission gas release in pins without a plenum using 137Cs.
- Pin mensuration and pin identification
- Non-destructive burn-up analysis of irradiated fuels.
- High burn-up effects in oxide fuels.
- Modelling input into disposal scenarios.

Other applications include non-destructive burn-up analysis of nuclear items with Security and Safeguarding implications

It should be noted that some of the techniques described are reliant on the development of two areas;

- The required resolution and size of the features of interest, and therefore the collimator and spectrometer design discussed earlier.
- Treatment of metallographic samples such that they retain the gamma emitters of interest for analysis.

Once these criteria are further developed, assessing the radial and axial distribution of gamma emitters within a metallographic section will provide a potentially valuable input into models for reactor operation and fuels disposal.

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