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Non destructive determination of PuO_2 content in MOX fuel pins for fast reactors using Passive Gamma Scanning

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ABSTRACT

Use of Passive Gamma Scanning for non destructive evaluation of PuO_2 content in mixed oxide (MOX) fuels for fast reactors is demonstrated. Experiments have been carried out on MOX fuel pins for the hybrid core of Fast Breeder Test Reactor having nominal PuO_2 content of 44% and MOX pins having nominal PuO_2 content of 21% for the Prototype Fast Breeder Reactor. A comparison of results obtained using a conventional NaI(Tl) detector and that using a through well shaped detector is also presented.

1. Introduction

Advanced Fuel Fabrication Facility (AFFF) of Bhabha Atomic Research Centre at Tarapur, India has fabricated $(U,Pu)O_2$ mixed oxide (MOX) fuel elements for thermal and fast reactors. MOX fuel pins containing 44% PuO_2, have been recently fabricated at AFFF and loaded in the Fast Breeder Test Reactor (FBTR), at Kalpakkam. Fabrication of MOX fuel pins with 21% PuO_2 content for Prototype Fast Breeder reactor (PFBR) is currently in progress at AFFF.

$(U,Pu)O_2$ MOX fuel is fabricated through the powder metallurgical route. Appropriate quantities of UO_2 and PuO_2 powders are mixed and milled to make a batch of MOX powder; nominally of 5 kg in weight. PuO_2 content of FBTR fuel is specified as $44 \pm 1\%$ and that of PFBR fuel is $21 \pm 1\%$ by weight. PuO_2 content in the MOX blend is determined using Neutron Well Coincidence Counting (NWCC) for process control purpose [1,2] by analyzing a sample of 50 g from every batch. After confirming the composition, the batch is processed further. The powder is pre compacted, granulated and compacted. The compacted pellets are sintered in reducing atmosphere of N_2 –7% H_2 at high temperature. Sample pellets from every batch of pellets is subjected to conventional chemical analysis for its PuO_2 content [3].

MOX pellets which conform to the stipulated specifications are stacked into the specified length and loaded into clad tubes. The tubes are hermetically sealed by Tungsten Inert Gas welding with the end plugs at both the ends. The pins are subjected to various quality control checks before final acceptance [4].

2. Objective

PuO_2 percentage is one of the most critical specifications of MOX fuel. Chemical analysis of MOX pellets for PuO_2 content is carried out on one sample pellet per batch. The value obtained is assigned to the batch of pellets of 5 kg. Pellets from one batch of MOX powder are loaded into approximately twenty fuel pins. In practice, some of the pins may contain pellets from more than one batch. It is therefore desirable to estimate the average PuO_2 content of every fuel pin as it is more relevant than assigning the average PuO_2 content of a batch of pellets to all the pins fabricated from it.

The fuel pin for FBTR is approximately 1 m long with a MOX stack of nominal length 430 mm. It contains an axial blanket of nominal length 70 mm made up of UO_2 pellets on the bottom side and a single insulation pellet (UO_2) on top side (Fig. 1a) Fuel pin for PFBR is approximately 2.6 m long with a 1 m long MOX stack and 300 mm long axial blanket (UO_2) on either side (Fig. 1b) [5]. Due to human error, there is a chance of cross mixing of the blanket and fuel pellets while loading of the stacks in FBTR and PFBR fuel pins. Film based technique such as X-Gamma Autoradiography (XGAR) is used for detection of cross mixing of blanket and fuel pellets in a welded fuel pin [6].

During the fabrication of fast reactor MOX fuel pins, the need for a fast and nondestructive technique was felt to confirm the correct loading of pellets and to monitor the uniformity of composition over the length on all the finished fuel pins. Use of Passive

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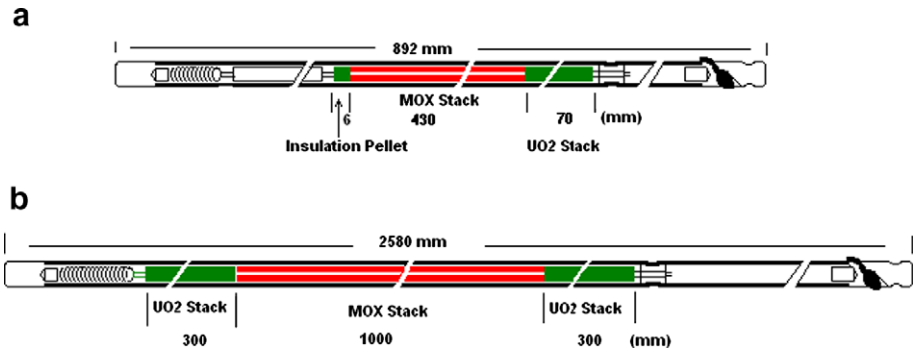

Fig. 1. Schematic diagram of MOX fuel pins for (a) FBTR and (b) PFBR.

Gamma Scanning has been explored for this purpose in our laboratory.

Use of Passive Gamma Scanning (PGS) has been demonstrated to estimate the PuO_2 content in waste drums and monitoring the enrichment of U^{235} in thermal reactor fuel pins [7,8]. Low gamma count rate were the limiting factor to achieve the required accuracy and throughput simultaneously. A combination of Passive and Active gamma scanning has been reported for MOX fuels for Fast Breeder Reactor and Fast Flux Test Facility [9]. Scanning the fuel rods through a Cs^{137} gamma densitometer and/or assaying a nuclide using fission gammas and delayed neutrons by using a Cf^{252} neutron source is also reported [10]. Use of PGS has been demonstrated for estimating the PuO_2 content in MOX fuel pins for thermal reactors with PuO_2 content (0.9–3.25%) [11]. PGS has also been demonstrated for calculating the fissile column length in Mixed Carbide fuel pins with 70% PuC for FBTR [12].

Previously reported studies on fuel pins for fast reactors have not accepted PGS as a standalone technique for estimating PuO_2 content due to the long counting hours required to achieve accurate results. An attempt was made at AFFF to explore the possibility of using Passive Gamma Scanning as a routine quality control technique to estimate the PuO_2 percentage in fast reactor MOX fuel pins with good throughput and high accuracy. Our experiments were based on improved counting statistics and statistical analysis of the scan data. Studies were conducted on fast reactor MOX fuel pins with nominal PuO_2 content of 21% and 44%.

This paper presents the details of the experiments, comparison of results obtained using a conventional detector and an annular detector and its application in the routine quality control of fast reactor MOX fuel pins.

3. Experimental

Gamma Scanner at AFFF consisted of a $3'' \times 3''$ NaI(Tl) detector and single channel analyzer based counting system. The scanner was an automated system, which enabled the movement of the fuel pins before the detector, after loading in the magazine. The detector was shielded to reduce the background noise and the pins were assayed through a collimated window. The speed of scanning was controlled through the software and was fixed to be 1 mm/s. During the scanning, the pin was made to move in front of the detector step by step in synchronization with the counting electronics. The speed of the pin movement, slit width and the counting time were optimized to achieve a relative standard deviation not more than 1.5% at a good throughput. This geometry provided only a tangential coverage to each section and hence the count rate was of the order of 5000 counts/s.

Since the nuclide to be assayed was Pu^{239}, its signature peaks of 332.8 keV, 344.96 keV, 375.02 keV and 413.69 keV, which were registered as a complex peak centered at 384 keV was chosen for counting [13]. Though the 384 keV complex was low in intensity as compared to the low energy peaks of Pu, it was preferred for Pu estimation for our studies since there was no overlapping of uranium gamma energies in the selected window of energy. The window of the single channel analyzer was set to count from 300 keV to 450 keV so that the complete 384 keV complex peak was covered (Fig. 2).

The pin was virtually divided into a number of sections of 2.5 mm (FBTR) or 5 mm (PFBR) width using a collimator and each section was counted for one second as the pin moved. The collection of these counts represented a statistical sample of gamma counts collected under same conditions over the length of the pin. The average of this collection was taken as the average count rate of the pin and a scatter of $\pm 3\sigma$ is considered for better confidence on results. The counting geometry and parameters were set to achieve maximum accuracy without compromising on throughput. The thickness of the collimator was maximised and width of the slit was minimized for reducing the background. Due to this, the experimental set up was applicable only to MOX fuel pins with a minimum of 19.5% PuO_2. In view of the noise level and possible non-linearity at lower values of PuO_2, the validity of the correlations is limited to the range of composition of the pins used for calibration.

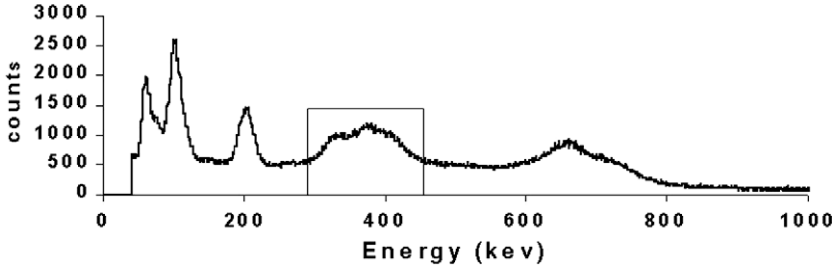

Fig. 2. Gamma spectrum of MOX through NaI(Tl) detector.

3.1. Improved counting geometry

A through well geometry NaI(Tl) crystal scintillator detector was incorporated in the existing scanner to enhance the sensitivity of the system, which was originally based on a conventional $3'' \times 3''$ NaI(Tl) crystal detector [14]. The use of annular detector to estimate the PuO_2 content in MOX fuel pins for thermal reactors with very low PuO_2 content (0.4%) has been previously demonstrated [15].

The annular counting geometry provided a circumferential coverage to the pin and hence a considerable increase in the count rate. The sensitivity of the system was enhanced and hence the accuracy of estimation of PuO_2 content also improved.

The detector was collimated with a pipe shaped lead collimator fitted inside the detector and the pins were made to move through the collimator (Fig. 3). The thickness of the collimator was 10 mm and the length was 30 mm. The collimator had a circumferential slit of arc length 50 mm. The slit width of the collimator was 1 mm for FBTR pins and 3.5 mm for PFBR. It was possible to increase the count rate by almost five times from the small section of the pin moving at the same speed as compared to the conventional scanning system. Due to the increase in count rate, it was possible to achieve a relative standard deviation less than 1%. The calibration pins were used to standardize the system before every real assaying so that slight drifts due to power fluctuations, etc. were eliminated.

4. Results

A number of FBTR and PFBR MOX fuel pins were fabricated with small variation in the average PuO_2 content within the corresponding specified ranges. The pins were used to establish a correlation between the count rates and the average PuO_2 content. The batches were analysed by chemical methods using large number of samples. The chemically determined value of PuO_2 content of the batch was taken as the average PuO_2 content of the pin. All the calibration pins were fabricated using the same lot of PuO_2 powder to avoid any variation in the spectrum due to variation in the isotopic ratio of Pu.

4.1. Conventional detector

A linear correlation was established between the average gamma counts and the PuO_2 content determined by chemical technique.

The correlation between the average count rate of the pin and its chemically determined PuO_2 content in FBTR pins was found to be;

$$\text{Average count rate} = (326.07 * \%PuO_2) - 5395.5 \quad (1)$$

Using the correlation, it was possible to estimate the average PuO_2 content of a fuel pin with an accuracy of ±0.8%(abs.) and confidence of 99.7% (3σ) in the range of 43–45% of PuO_2 in FBTR MOX pins.

The correlation between the average count rate of the pin and its chemically determined PuO_2 content in PFBR MOX fuel pins was found to be

$$\text{Average count rate} = (758.25 * \%PuO_2) - 10,728 \quad (2)$$

It was possible to estimate the PuO_2 content of a PFBR MOX fuel pin with an accuracy of ±0.3%(abs.) and confidence of 99.7% (3σ) using this correlation valid for the range 19.5–23.0% of PuO_2 in PFBR pins. (Fig. 4). The specification of PuO_2 content in both FBTR and PFBR MOX fuel had a tolerance band of ±1% and hence this method can be effectively used for confirming the average PuO_2 content of the fuel pins to be within the specification.

The minimum spatial resolution of detection was 5 mm which is less than the size of a pellet. It was therefore possible to detect any anomalous pellet present in the welded fuel pin.

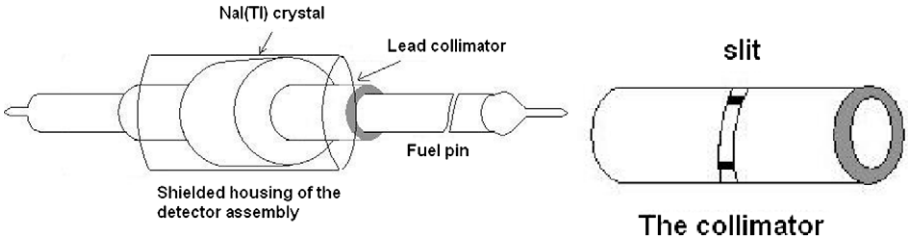

Fig. 3. Schematic geometry of annular counting system.

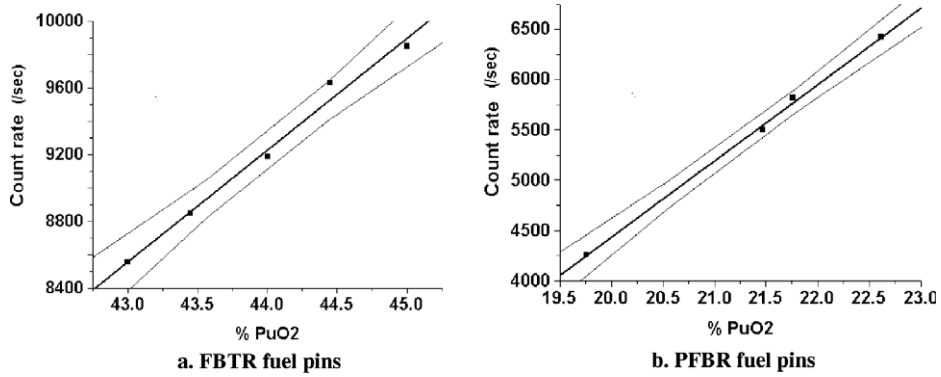

Fig. 4. Calibration graphs using $3'' \times 3''$ NaI(Tl) detector (a) FBTR pins and (b) PFBR pins.

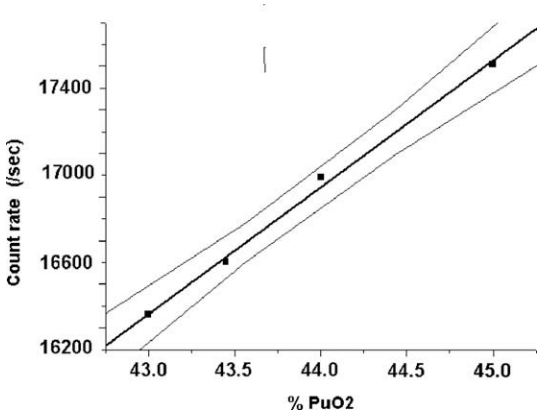

Fig. 5. Calibration graph of FBTR and with annular NaI(Tl) detector.

4.2. Annular detector

FBTR and PFBR pins used earlier were scanned using the annular NaI(Tl) detector. A slit width of 1 mm was used for FBTR pins and 3.5 mm for PFBR pins. Due to higher sensitivity of the annular detector set up, the slit widths were reduced as compared to that for the conventional detector so that the count rates are not more than 20,000. It helped in minimizing the mean noise level and the dead time. The results of the scans of FBTR and PFBR pins are presented in the following paragraphs.

4.3. FBTR fuel pins

PGS using the through well detector showed very high count rates and hence both FBTR and PFBR could not be scanned using the same parameters. FBTR fuel pins were scanned using reduced slit width of 1 mm and scanning speed of 1 mm/s. The correlation between the average gamma count rate and PuO_2 content obtained by chemical analysis in FBTR pins using the set up was found to be;

$$\text{Average count rate} = (581.68 * \%PuO_2) - 8448.9 \quad (3)$$

The correlation is valid for the composition of PuO_2 studied (43–45%) as shown in Fig. 5.

It was therefore possible to estimate the average PuO_2 content of FBTR MOX fuel pins with an accuracy of ±0.6%(abs.) and 99.7% (3σ) confidence. This was better than the results achieved using a conventional $3'' \times 3''$ detector which is ±0.8%(abs). Scans of three fuel pins covering the complete range of enrichment specification for composition (as per chemical analysis values) are shown in Fig. 6. The specification for PuO_2 content was 44 ± 1%.

The relative standard deviation of the counting set up was confirmed to be less than 1% in this case. Any abnormal standard deviations reflected in the scans could be considered as reflection of non uniform distribution of PuO_2 over the length of the pin indicates significant variation of PuO_2 content from pellet to pellet.

4.4. PFBR fuel pins

Since the PuO_2 content of PFBR fuel was less than that of FBTR, the slit width was increased to 3.5 mm with a scanning speed of 3 mm/s. This was helpful in facilitating the scan of the longer (2580 mm) PFBR pin without losing any relevant information. The spatial resolution was still less than the length of one pellet and hence the presence of wrong loading of pellets could be detected. The minimum PuO_2 content for this correlation to be valid is 19.5%. The correlation established for the range of 19.5–23% PuO_2 between the average gamma count and chemically estimated PuO_2 content using the set up is given by;

$$\text{Average count rate} = (3144.2 * \%PuO_2 \text{ content}) - 59,881 \quad (4)$$

and is shown in Fig. 7.

Since the relative standard deviation was less than 1%, it was possible to estimate average PuO_2 content of PFBR MOX pins with an accuracy of ±0.1% and 3σ confidence.

Gamma scans of two PFBR pins with a variation of 0.15% in PuO_2 content are shown in Fig. 8. Gamma Scan of the calibration pin showing the detection of cross mixing of pellets is shown in Fig. 9. The calibration pin was loaded first with a $DDUO_2$ pellets stack (six pellets) with one MOX pellet placed at the middle followed by a MOX pellets stack (six pellets) with one $DDUO_2$ pellet placed at the middle. The pin contained MOX and $DDUO_2$ particles of major dimension 1.5 mm fixed at known locations on the plenum spring. It was possible to detect and discriminate these particles as MOX and $DDUO_2$ using Gamma scanning. Fig. 10 shows scan of a PFBR pin with a MOX pellet in the blanket region. The

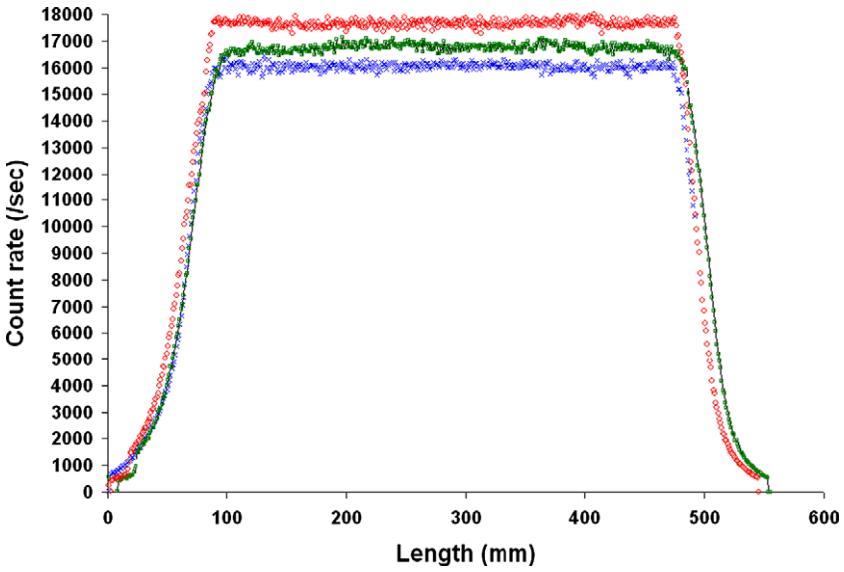

Fig. 6. Gamma scans of FBTR MOX fuel pins with difference in average PuO_2 content (*** 43%, - - - - 44%, ooo 45%).

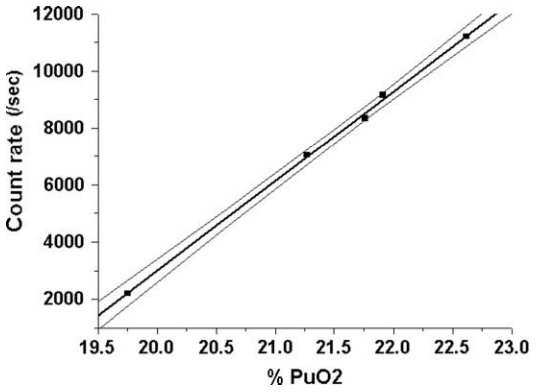

Fig. 7. Calibration graph of PFBR pins using annular NaI(Tl) detector.

difference in count rate being almost 500%, it is certain that, cross mixing of fuel pellet in blanket stack can be detected.

Correlations (1)–(4) hold good for FBTR/ PFBR MOX pins assayed with the same geometrical parameters of the respective experimental set up and fabricated from a PuO_2 powder with the same ratio of Pu^{239} as that used for fabrication of the calibration pins. The assayed gamma peak of 384 keV is from Pu^{239} only and any variation in its ratio would result in a variation in the count rate for the same average PuO_2 content. In case of a different PuO_2 powder lot, the corrected average count rate has to be calculated by normalizing the obtained average count rate. Normalization was done by dividing the obtained count rate with the ratio of Pu^{239}% in the powder to that in the powder used in the calibration pins. Percentage of PuO_2 was estimated from Figs. 6 and 8 after normalizing the average count rates using the corresponding isotopic ratios. PuO_2 content presented in the figures are the chemical values.

5. Conclusions

Passive Gamma Scanning can be effectively employed for estimating the PuO_2 content and its homogeneity over the length of the fuel stack in MOX fuel pins for fast reactors. The accuracy of estimation of PuO_2 percentage (44% nominal) in FBTR MOX fuel pins is improved by 25% by using annular type of detector as compared to a conventional type of detector system. In PFBR MOX fuel pins, the accuracy of estimation of PuO_2 percentage (21% nominal) using annular detector system improved by 30% as compared to a conventional geometry detector system.

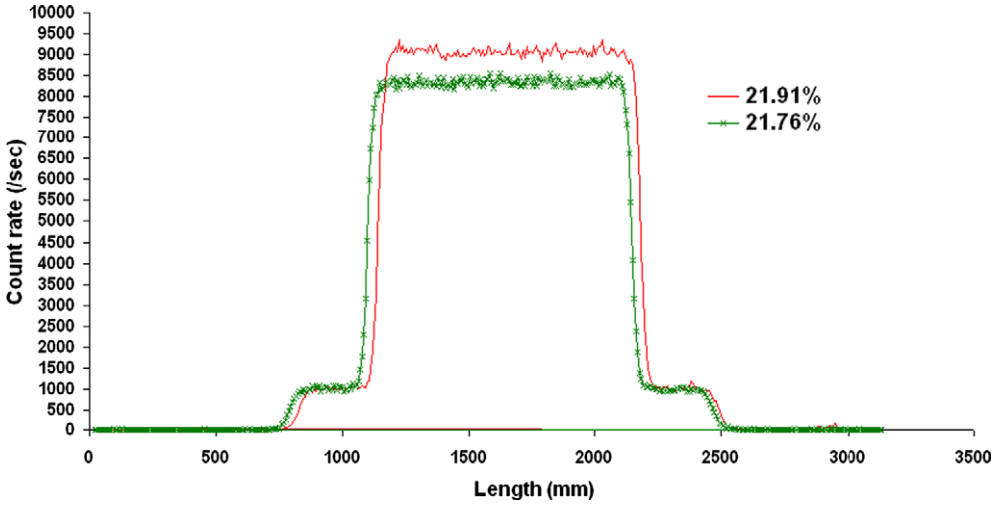

Fig. 8. Gamma scans of PFBR pins with varying average PuO_2 content (21.75%, 21.91%).

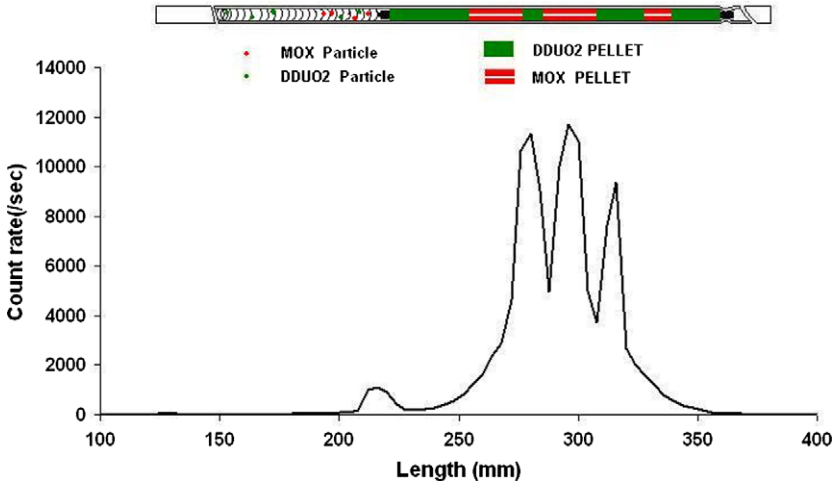

Fig. 9. Gamma scan of PFBR experimental pin showing presence of DDUO2 and MOX particles on plenum spring and cross mixing of MOX and DDUO2 pellets (cross mixing and chips) along with its schematic diagram.

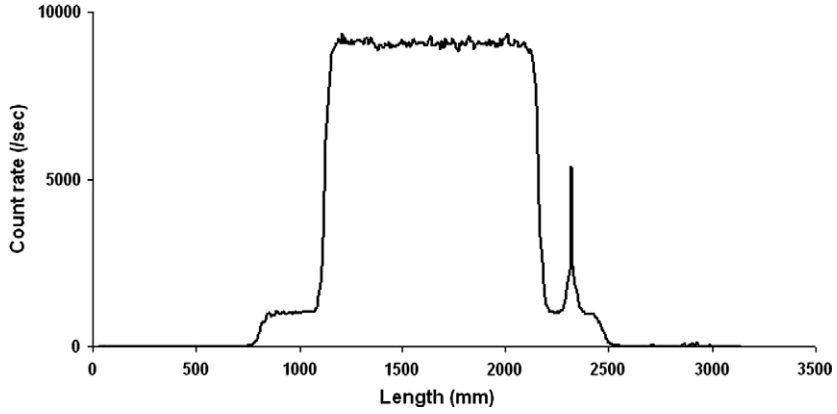

Fig. 10. Detection of a MOX pellet in top axial blanket in a PFBR MOX fuel pin.

It can be employed for detection of anomalous pellets in the fuel and blanket stacks due to cross mixing. It can also confirm the correct loading of the fuel and blanket pellets in the welded fuel pins. The normal scanning time taken for FBTR and PFBR pins are approximately 12 min and any anomalies detected are confirmed by slower scans of the selected portions. Being a non destructive, prompt and easy technique, it can be used routinely on the fuel pins on a 100% basis. This technique can be used efficiently as a screening technique and hence the number of samples for chemical analysis for PuO_2 content can be reduced resulting in reduction of generation of radioactive liquid waste.

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