ESTIMATION OF PLUTONIUM CONTENT BY PASSIVE GAMMA SCANNING AND NEUTRON WELL COINCIDENCE COUNTING FOR (TH-PU)O₂ MOX-FUEL.


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ABSTRACT

Advanced fuel fabrication facility (AFFF) has manufactured (U-Pu)O₂ MOX fuels for different types of reactors. AFFF has also made (Th-Pu)O₂ MOX experimental AHWR type fuel pins for irradiation in research reactors at Trombay. Pu-content in mixed oxide fuel is estimated using Neutron Well Coincidence (NWCC) Counting at powder blend stage as a process control check and by Chemical method at sintered pellet stage as a quality control check. Owing to the problem associated with dissolution of thorium-based mixed oxide fuel, an attempt has been made to evaluate the plutonium content at pin stage using Passive Gamma Scanning (PGS) technique. The results obtained from PGS were compared with results of NWCC and chemical method. PGS and NWCC being NDT methods minimize the loss of precious nuclear material and also reduce generation of liquid waste.

Keywords: Passive Gamma Scanning, Neutron Well Coincidence Counting, Plutonium, Thorium, Collimator, Concentric slits, AHWR

INTRODUCTION

Advanced Fuel Fabrication Facility (AFFF) has fabricated Plutonium based mixed oxide (MOX) fuels with varying percentages of Plutonium for different types of reactors. AFFF has also manufactured Thorium oxide based mixed oxide fuel with PuO₂ compositions of 1% and 8% PuO₂ for AHWR type experimental fuel elements for irradiation in research reactors at Trombay. MOX fuel pellets are fabricated by conventional powder metallurgical route [1]. The required quantities of PuO₂ and ThO₂ powder were weighed, mixed and milled in attritor to attain required homogeneity. The mixed and milled powder samples were checked for enrichment using NWCC as a process control check. The accepted powder blend was pre- compacted, granulated, and final-compacted and sintered at high temperature. Chemical analysis of sintered sample pellets was carried out for accurate determination of the PuO₂ composition [2]. The sintered and visually accepted pellets were encapsulated into Zircaloy-2 cladding tubes. The schematic sketch of the pin is shown in Fig. 1. The fuel rods were checked for total integrity by helium leak testing and X-ray radiography of end plug welds. The presence and proper sequence of hardware and MOX column was checked by X-ray gamma autoradiography (X-GAR). X-GAR also revealed cross mixing of pellets, composition fluctuation across the fuel stack and presence of Pu-rich agglomerate [3]. The passive gamma scanning (PGS) has been developed as a faster, non-destructive, qualitative and quantitative test technique for quality control of Plutonium containing MOX-fuel pins and can be utilized for this purpose [4,5]. In the present paper an attempt has been made to use PGS to estimate the Pu-contents and its spatial distribution across the length in (Th-Pu)O₂ fuel pins. The results obtained were also compared with NWCC and Chemical analysis.

Fig. 1: Schematic diagram of AHWR type (1%Pu+Th) O₂ fuel pin
EXPERIMENTAL

Neutron Well Coincidence Counting

NWCC system at AFFF consists of 16-numbers of \( ^3\text{He} \) cylindrical detectors arranged in a circular manner around the central well of diameter 150mm and embedded in the 500mm X 500mm HDPE. The detectors are of 25mm diameter, 500 mm length and 4 atmospheres fill pressure of \(^3\text{He} \) gas. The detectors were connected in parallel to the coincident counting unit Fig.2.

Samples from mixed-milled powder lots were prepared and doubly sealed. The samples were put into the well and Real Count rates (R) for each sample were acquired for 1000 seconds and recorded.

Chemical Analysis

The sintered-sample pellet (one pellet per batch) from each batch was subjected to chemical analysis for Pu-content. The powders were dissolved in Con.\( \text{HNO}_3 \) + 0.05 \( \text{M} \) HF by Microwave heating and titrates were prepared. Titrate from each MOX-batch was analyzed for PuO\(_2\) content by redox-titrimetric method using potentiometric-end-point detection technique. The results were recorded.

Passive Gamma Scanning

Gamma fuel pin Scanner at AFFF consists of 3Ø X 2Ø through hole shaped annular NaI(Tl) detector and single channel based counting system. The detector assembly has been shown in Fig.3. The use of through-hole shaped detector has improved the counting geometry and hence the better counting statistics.
has been achieved. The window of the single channel analyzer was set from 300 Kev to 500 Kev so that the gamma photons of energy 344.96 kev, 375.02 Kev and 413.69 Kev from Pu 239 centered at 384 Kev in NaI(Tl) were counted [6]. The fuel pins loaded with different MOX-pellet batches were scanned and the average count rates over the fuel stack length were recorded.

RESULTS AND DISCUSSION

The count rates acquired from the experiments were analyzed separately for each technique with respect to the % PuO2 (abs.) reported by Chemical method. The regression analysis was used to establish the relation between the count rates and the % PuO2. The relations were obtained and fitted separately using the method of least square with ±3σ limits. The figures 4 and 5 show the correlation between neutron and gamma count rates against Pu-content obtained from chemical analysis respectively. Fig.6 shows the correlation graph of %PuO2 as estimated using NWCC and gamma count rate.

PGS was found to be more sensitive to the change in PuO2 content as the average count rate (7082 counts) was higher than that from the NWCC (3 counts). It was found that variation in %PuO2 of the order of 0.10% could be detected using PGS (σ = 203 counts) where as NWCC could detect a change of the order of 0.11% (σ = 0.5) within ± 3σ limits. It was observed and confirmed that PGS could detect the variation of 0.1% (abs.) in all the AHWR type MOX fuel pins, Fig.7.

The precision of PGS was found to be better in estimating the PuO2 content than NWCC as the relative standard deviation of count rate were 2.87% and 17.12% respectively. As the stipulated PuO2 content for AHWR - type MOX- fuel for experimental studies was 1±0.1%, PGS could be used for estimating the same and its variation more precisely.

CONCLUSION

PGS is a useful non-destructive assay technique for estimating the PuO2 content in MOX at the pin stage. PGS is as good as NWCC in estimating %PuO2 within the specified enrichment band for experimental AHWR-type MOX fuel. Passive gamma scanning could be used as an effective, non-destructive, fast and quantitative test technique for quality control of plutonium-thorium mixed oxide fuel. Being nondestructive, it can be carried out on 100% basis. It has the additional advantage that it avoids generation of liquid waste when compared with chemical method.

ACKNOWLEDGEMENT

The authors would like to thank their colleagues at AFFF for their help in carrying out this work.

REFERENCES