Passive, non-destructive enrichment measurement of sintered UO_2 pellets

A. J. Parker, J. Zabalza, M. Bandala, X. Ma, J. Taylor, P. Murray, S. Marshall, and M. J. Joyce

***Abstract*— The measurement of uranium dioxide (UO_2) enrichment in simulated pressurized water reactor fuel pellets has been performed using the infinite thickness technique. The use of this technique is well established on large volumes of uranium compounds, however new advanced reactor fuels require the enrichment assessment of individual UO_2 fuel pellets. Monte Carlo GEANT4 simulations and experimental measurements were performed to collect energy and count data for sintered UO_2 sim fuel pellet slices. Results indicate that the infinite thickness technique can be used on individual fuel pellets for determination of enrichment, by confirming the infinite thickness of sintered UO_2 (4.9 mm), and that the count rate form the characteristic 185.6 keV ^{235}U photopeak increases linearly with fuel pellet enrichment. Working is ongoing to refine the simulated datasets and improve experimental measurements.**

***Index Terms*— Uranium enrichment, infinite thickness technique, passive non-destructive analysis, gamma spectroscopy.**

I. Introduction

THE composition of uranium dioxide (UO_2) used in nuclear fuel, specifically the enrichment of the uranium, is critical to achieving maximum thermal output and efficiency in nuclear power reactors. Deviation from the required enrichment could lead to higher or lower fuel-burnup or unexpected power profiles within a reactor core. These scenarios could lead to reduced reactor efficiency at the least, or an increase the likelihood of serious safety issues at worst.

As such, there is a requirement to monitor the enrichment of uranium during the manufacturing process. In the United Kingdom and elsewhere, assurance is conducted by assessing the enrichment of UO_2 powder following conversion from its chemical precursor uranium hexafluoride (UF_6), prior to processing into a finished fuel pellet [1]. After conversion, UO_2 powder is stored in standard 205 liter drums. It is in this phase that enrichment monitoring takes place, using the infinite thickness technique [2]. Though, Neutron interrogation is the most common method [2], Gao et al. (2020) have presented one of the first methods for measuring enrichment of pellets in a fuel rod [3], but, at present there is no widely adopted use of the technique for measuring individual fuel pellets.

However, the new generation of PWRs will use advanced UO_2 fuel designs where fuel pins within an assembly may contain pellets of differing enrichments. As such, the enrichment of individual pellets will be needed to be measured prior to loading into a fuel-pin. Therefore, this work seeks to develop a proof-of-principle for adopting the infinite thickness technique on individual UO_2 fuel pellets.

II. Materials and Method

This work utilized both Monte Carlo simulations, using GEANT4 nuclear particle code, and γ-ray measurements from UO_2 pellet slices, of natural and depleted enrichments. Both simulated and experimental pellet slices were 9.4 mm in diameter and a UO_2 density of 10.9 g cm^{-3}. An example UO_2 spectrum is shown in Fig. 1, 6.5% enriched pellet of 12 mm in height, highlighted is the photopeak associated with the characteristic185.6 keV emission form the ^{235}U component used for the infinite thickness measurements [4].

Both simulated and experimental measurements utilized a thallium-doped caesium iodide scintillator (Scionix, Netherlands) and a europium-doped strontium iodide scintillator (Scionix, Netherlands). For the experiments, the scintillators were connected via a 14-pin to a digiBASE photomultiplier tube base (Ametek Ortec, USA) and controlled using MAESTRO Multichannel Analyser Emulator software (Ametek Ortec, USA). Sintered UO_2 sim-fuel pellets were acquired from the UK National Nuclear Laboratory.

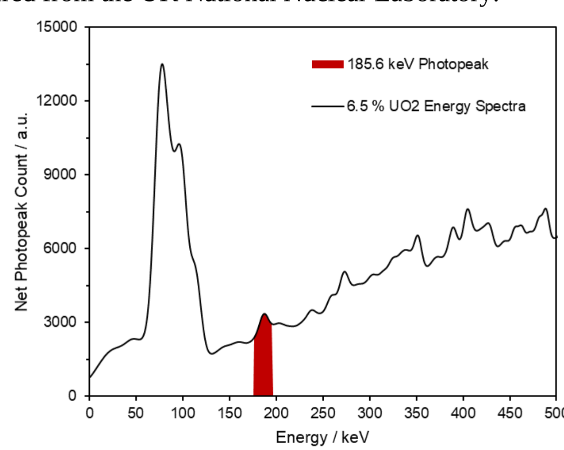

Figure 1. 0-500 keV range GEANT4 energy spectrum from a 12 mm height, 6.5% enriched UO2 pellet. The area highlighted in red is the count associated with the characteristic 185.6 keV γ-ray emission of ^{235}U.

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A. J. Parker, X. Ma, C. J. Taylor, and M. Joyce are with Department of Engineering, Lancaster University, Bailrigg, Lancashire, United Kingdom, LA1 4YR (e-mail: a.j.parker3@lancaster.ac.uk).

J. Zabalza, P. Murray, and S. Marshall are with Department of Electronic and Electrical Engineering, University of Strathclyde, Glasgow, United Kingdom, G1 1XW (e-mail: j.zabalza@strath.ac.uk).

Both the simulations and experiments utilized the same sample geometry, where the UO_2 pellet was placed 10 mm from the detection media, and surrounded by a lead collimator, with a 7 mm diameter circular aperture between the detection media and UO_2 sample.

III. RESULTS AND DISCUSSION

A comparison of the simulated net counts taken from the 185.6 keV photopeak is shown in Figure 2. It can be seen that for the five UO_2 enrichments presented, all are indicative of the count profile associated with the infinite thickness technique. Specifically, that as the pellet height increases from 0 mm, the net count associated with the 185.6 keV emission also increases, until it reaches a maximum – in this case between 3-5 mm in thickness. This is consistent with the stated infinite thickness of sintered UO_2 (4.9 mm) [5]. After reaching the maximum count, the photopeak count for pellets above 5 mm plateaus and, in higher enrichment, shows a gradual decrease. This trend is observed all the data sets collected for different enrichments and pellet lengths – though only a selection are presented. The gradual decrease in photopeak count for higher enriched pellets is likely due to a combination of reasons, principally, the simulated collimator opening (7mm) being too large and an artefact of the empirical post-processing of the simulation data. Subsequent simulations will seek to refine these aspects and confirm the datasets with experimental data.

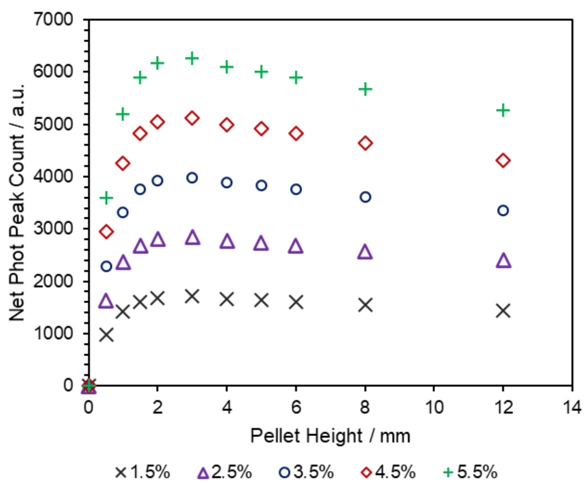

Figure 2. Net count arising from the 185.6 keV ^{235}U photopeak for a selection of different enrichment simulated UO2 pellets of increasing length.

Figure 3 shows the results of the 185.6 keV photopeak count taken from the simulated 5 mm pellets as the uranium enrichment increases. The linear relationship between the net photopeak count and degree of enrichment is apparent. This relationship is characteristic of the infinite thickness method, given that all experimental properties remained the same for the simulations, the only variation was ^{235}U content. As the ^{235}U content in the pellet increases, so does the number of γ-ray emissions, significantly the 185.6 keV emission. Accordingly, the count detected from this emission will also increase. The data from both Fig.2 and Fig. 3 demonstrate than use of the infinite thickness technique can be applied to measuring the enrichment of single fuel pellets. Experimental data (not shown) backs up the simulation findings but, at present, is limited by significant statistical error.

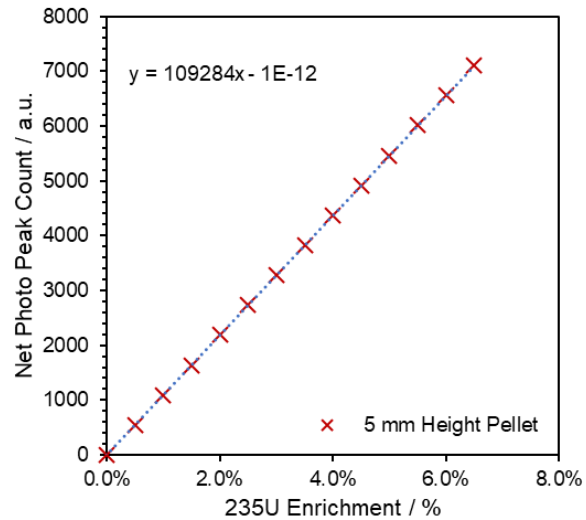

Figure 3. The relationship between ^{235}U enrichment of a UO_2 fuel pellet and the associated count arising from the 185.6 keV photopeak.

The principal limitation of this work is the counting durations to achieve the appropriate counting statistics, limited by the detector performance as found in the experimental measurements. As such, the choice of scintillator is being evaluated along with the detector geometry, to reduce the time taken to acquire the necessary count data and improve spectral energy resolution. Further simulations and experimental measurements are ongoing and will be presented at the conference.

IV. CONCLUSION

This work applied the well-established infinite thickness technique to the measurement of individual fuel pellets enrichment. The preliminary results of this work show that the determination of the infinite thickness of fuel pellets matches the predicted literature value. Furthermore, there is an apparent linear increase in the net count arising from the magnitude of the 185.6 keV photopeak, associated with the linear increase in proportion of ^{235}U in the sample.

The main finding of this work is that the determination of enrichment for a single UO_2 pellet is feasible. Along with the data presented here, a more comprehensive set of data is in preparation necessary to understand the counting statistics associated with counting time.

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