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Evaluation of micro-homogeneity in plutonium based nuclear reactor fuel pellets by alpha-autoradiography technique

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HIGHLIGHTS

- Alpha-Autoradiography of $(U-Pu)O_2$ oxide pellet having 0.4–44% PuO_2 was done.
- Exposure and etching time required for alpha-autoradiography was optimized.
- Size of Pu agglomerate was measured using micro-homogeneity profiling.

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ABSTRACT

Alpha-autoradiography is a fast and non-destructive technique which is used at Advanced Fuel Fabrication Facility (India) to evaluate micro-homogeneity of plutonium in uranium and plutonium mixed oxide $(U-Pu)O_2$ fuel pellets fabricated for both thermal and fast breeder reactors. In this study, various theoretical calculations to understand effect of alpha autoradiography process parameters and limiting conditions for measuring micro-homogeneity of plutonium in the pellets having different concentrations of plutonium were reported. Experiments were carried out to establish the procedure to evaluate micro-homogeneity of plutonium in $(U\text{-}x\%Pu)O_2$ pellets where x varies from 0.4 to 44% and to measure the size of agglomerates, if any, present in the pellet. An attempt had been made to measure plutonium content in the agglomerate using alpha-autoradiography. This study can also be useful for carrying out alpha-autoradiography of spent fuel pellets during post-irradiation examination.

1. Introduction

Advanced Fuel Fabrication Facility (AFFF) is a premier and the only facility in India capable of fabricating plutonium based fuel pellets for nuclear reactors at an industrial scale. AFFF had manufactured uranium and plutonium mixed oxide (MOX) fuel for Boiling Water Reactors (BWRs), Pressurized Heavy Water Reactors (PHWRs), Fast Breeder Test Reactor (FBTR). AFFF is currently engaged in fabrication of $(U-21\%Pu)O_2$ and $(U-28\%Pu)O_2$ MOX fuel for upcoming Prototype Fast Breeder Reactor (PFBR) at Kalpakkam India. $(U-Pu)O_2$ pellets are manufactured using Powder Oxide Pellet (POP) technique at AFFF. It basically involves mixing & milling of UO_2 and PuO_2 powder followed by compaction into pellets and then sintering at 1600 °C for 4 h as shown in Fig. 1.

Since the fabrication route involves mechanical mixing of PuO_2 and UO_2 powders, inhomogeneous mixing may result in fissile rich regions or PuO_2 agglomerate in $(U-Pu)O_2$ MOX pellets. These PuO_2 agglomerates, especially those near the surface of the pellet, are detrimental as they act as hot spots resulting in localized overheating of clad tube. In case of a transient power peaking, temperature gradient due to localized overheating may result in failure of clad tube which, in turn, may lead to release of fissile material into the coolant [1]. Furthermore, during reprocessing of spent nuclear fuel, PuO_2 agglomerates are difficult to dissolve without addition of HF in HNO_3 [1,2]. Usage of HF is avoided in reprocessing of fuel to prevent corrosion of reprocessing equipments which are generally made up of stainless steel. Hence, it is imperative to ensure micro-homogeneity of Pu in $(U-Pu)O_2$ pellet as it affects reliability, safety and ease of reprocessing of spent nuclear fuel.

Micro-homogeneity in the pellet is assessed by alpha-

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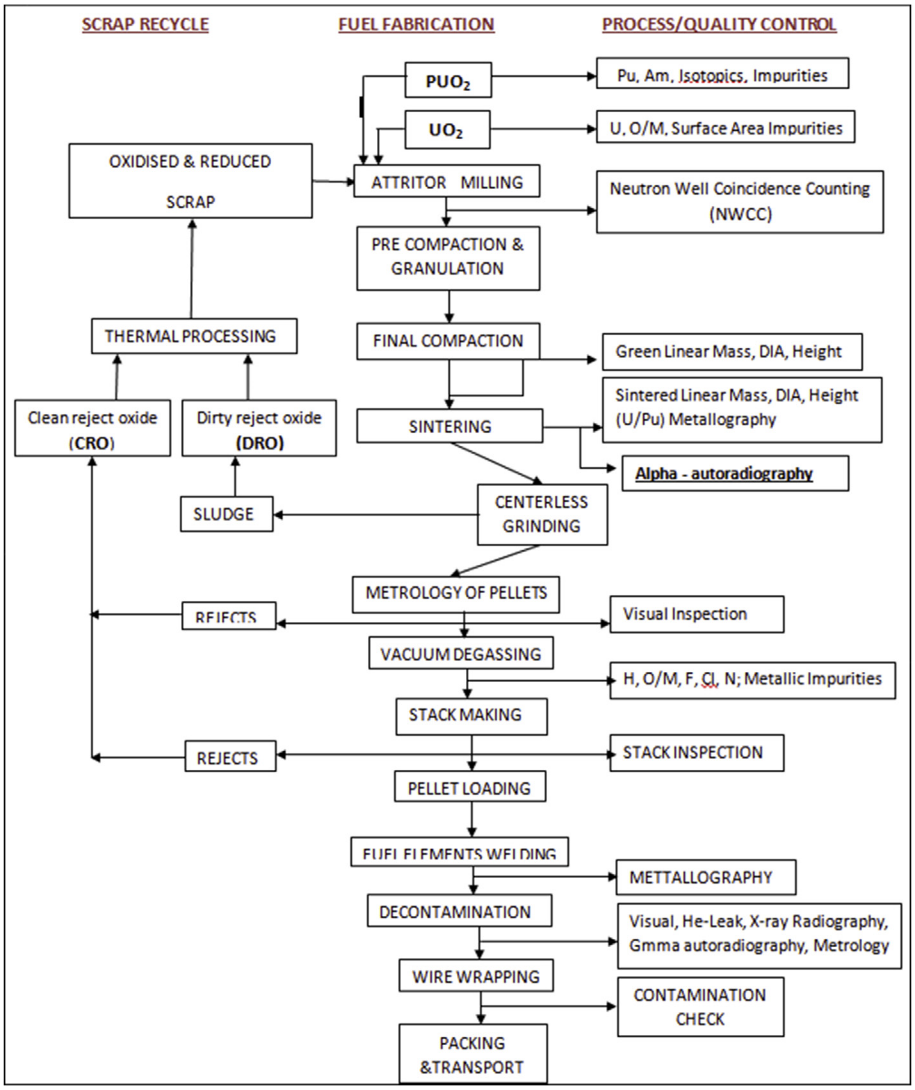

Fig. 1. Flow-sheet for fabrication of (U–Pu)O_2 mixed oxide fuel pellet at Advanced Fuel Fabrication Facility.

autoradiography technique which involves evaluation of tracks generated on alpha autoradiography film by alpha particles coming out of the pellet. Coagulation of alpha tracks at a particular portion on the film indicates the agglomeration of alpha emitting atom at the corresponding location on the surface of the pellet.

Other techniques such as Neutron radiography and Electron Probe Micro Analysis (EPMA) are available for evaluating micro-homogeneity in nuclear fuel pellets. Neutron radiography can be used to detect agglomerates present in the pellet [3]. But Neutron radiography requires huge investment and laboratory setup for handling fast neutron source. EPMA is another effective technique for evaluation of non-uniform distribution of elements on the surface of the samples, but it is expensive and time consuming [4]. In addition, it is difficult to install EPMA inside a glove box to carry out micro-homogeneity evaluation of radiotoxic (U–Pu)O_2 pellet. Alpha-autoradiography is the cheapest technique for evaluation of micro-homogeneity in a MOX pellet. It doesn't require any special equipment or huge laboratory setup and total processing time is less than half an hour. Therefore, alpha-autoradiography is a useful technique for quick estimation of micro-homogeneity in the pellet and hence an appropriate choice for AFFF where approximately 2 million pellets is fabricated annually.

Extensive studies had been carried out regarding use of alpha-autoradiography in determination of Radon and Thoron content in atmosphere, uranium content in soil, Pu level in bio-assay sample and neutron dosimetry [5–23]. But few literature are available providing insight into use of alpha-autoradiography in evaluation of micro-homogeneity in nuclear fuel pellets [4,24–27]. Alpha-autoradiography parameters used for determination of Radon or Thoron cannot be directly employed for alpha-autoradiography of (U–Pu)O_2 MOX pellets because the quantities of alpha emitting elements in the MOX pellet are not in ppm but as high as 45% (see Section 2.1.1). In addition, specific activity of alpha emitting isotopes such as Pu^{239}, Pu^{238}, Pu^{240}, Pu^{242}, and Am^{241} in the pellet is very high as compared to specific activity of U^{235} and Th^{232} atoms; as a result exposure time required will be different as compared to conventional alpha-autoradiography techniques used in other applications.

This paper is a brief account of theoretical calculations carried out for implementation of alpha-autoradiography for measurement of micro-homogeneity in the pellets. This paper discuss the experiments conducted to optimize parameters to use alpha-autoradiography for monitoring of Pu distribution and measurement of Pu agglomerates in (U–Pu)O_2 MOX pellet containing Pu varying from 0.4 to 44%.