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Fabrication Procedures for Manufacturing High Uranium Concentration Dispersion Fuel Elements

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ABSTRACT

IPEN developed and made available for routine production the technology for manufacturing dispersion type fuel elements for use in research reactors. However, the fuel produced at IPEN is limited to the uranium concentration of 3.0 gU/cm^3 by using the U_3Si_2-Al dispersion. Increasing the uranium concentration of the fuel is interesting by the possibility of increasing the reactor core reactivity and lifetime of the fuel. It is possible to increase the concentration of uranium in the fuel up to the technological limit of 4.8 gU/cm^3 for the U_3Si_2-Al dispersion, which is well placed around the world. This new fuel will be applicable in the new Brazilian-Multipurpose Reactor RMB. This study aimed to develop the manufacturing process of high uranium concentration fuel, redefining the procedures currently used in the manufacture of IPEN. This paper describes the main procedures adjustments that will be necessary.

1. Introduction

The Brazilian demand for radiopharmaceuticals has grown approximately 10% annually over the years. IPEN has worked continuously to increase the primary production of radioisotopes to meet this growing demand. This includes increasing the power of the IEA-R1, from 2 to 5 MW, and its utilization routine, from 64 to 120 hours a week. Moreover, in view of the advanced age of the reactor, which reached more than 50 years of operation, the construction of a new reactor research reactor for radioisotopes production has been discussed by Brazilian authorities as an urgent priority project. This is the Brazilian Multipurpose Reactor - RMB.

Since 1988, IPEN has fabricated the fuel for its pool type research reactor IEA-R1. The current fuel produced at IPEN allows the incorporation of 3 gU/cm^3, by using the uranium silicide technology (U_3Si_2). This uranium concentration is sufficient to operate the IEA-R1 reactor operating until the power level of 5 MW. However, this level of uranium concentration is not enough for the efficient supply of producing radioisotopes reactors with higher and, therefore, higher neutron fluxes, as the Brazilian Multipurpose Reactor - RMB. Another difficulty inherent to the low uranium concentration is the generation of large quantities of fuel depleted by irradiation, or the burned fuel. This is due to low operation life of the fuel with low uranium concentrations, which requires more frequent replacement.

Based on IPEN previous experience in developing and manufacturing dispersion type fuel, the objective of this work is to promote an adjustment to the current fuel manufacturing procedures, allowing the incorporation of higher uranium concentrations to the fuel. The goal is to increase the uranium concentration up to 4.8 gU/cm^3 by using the U_3Si_2-Al dispersion, and 3.2 gU/cm^3 with U_3O_8-Al. These concentrations are the maximum possible to be incorporated into the fuel when adopting the dispersions technology.

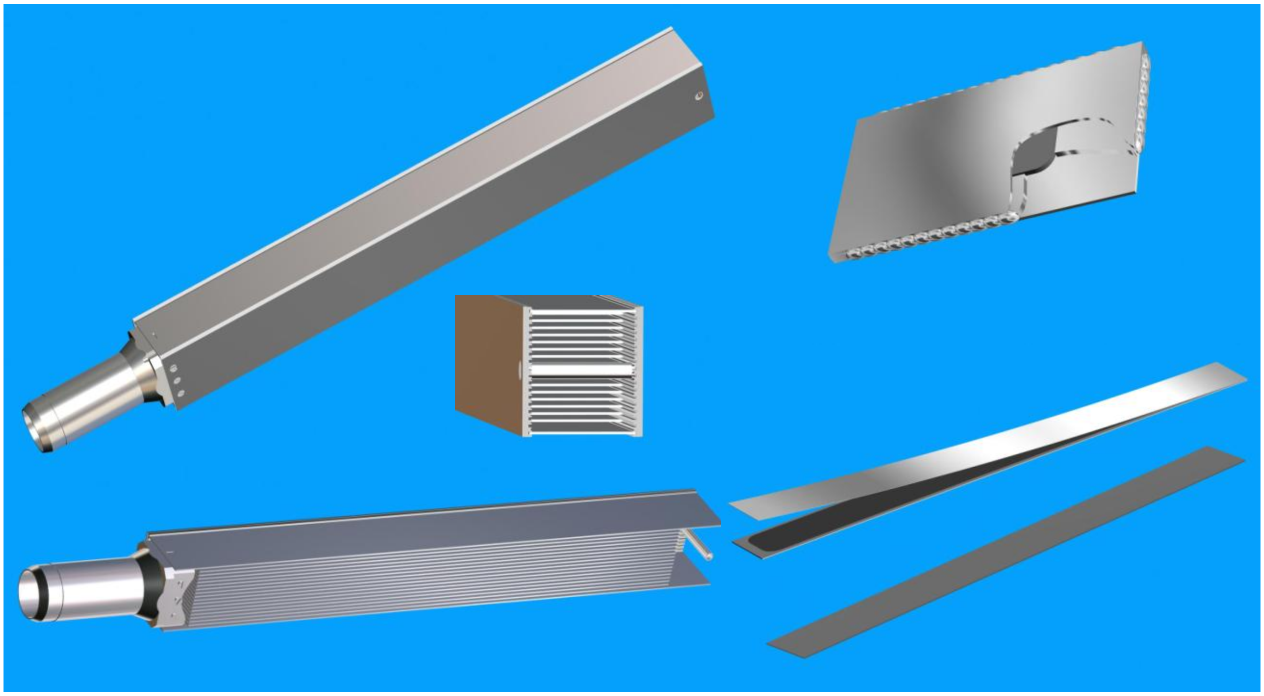
2. Experimental

The fuel elements used by the IEA-R1 research reactor is MTR type (Materials Testing Reactor), which are formed by assembling a series of fuel plates spaced apart, allowing the passage of a stream of water that serves as coolant and moderator. The fuel plates consist of a meat containing the fissile material, which is entirely protected with an aluminum cladding. They are manufactured by adopting the traditional technique of assembling core, frame and cladding, with a subsequent rolling operation. This manufacturing technique is internationally known as the "picture frame technique" [1,2]. Powder metallurgy techniques are used in the manufacture of the fuel meats, the core of fuel plates, which are called “briquettes”. The fuel meats are composed by cermets, or metal-ceramic composites, by using U_3Si_2 or U_3O_8 powder enriched to 20 wt% in the ^{235}U isotope (nuclear fuel material) together with aluminum powder (structural material of the meat matrix). The fuel plates are the most important part of the MTR type fuel, which is nothing more than a set of 18 parallel fuel plates, rigidly assembled to form the fuel element, as illustrated in Fig. 1A.

The manufacturing process of the MTR type fuel elements (U_3Si_2 uranium silicide type or U_3O_8 uranium oxide type) has two main stages, the pressing of briquettes, which are the fuel meats, and the rolling operation for manufacturing the fuel plates. The briquettes are assembled in an aluminum frame with two aluminum cladding plates forming a "sandwich". The set is then hot and cold rolled to get a fuel plate. Fig. 1B illustrates the set ready for rolling and the final fuel plate fabricated. Further details about the basic fuel element fabrication process adopted by IPEN, as well as a history of the manufacturing development of such fuel can be obtained from the literature [3,4].

In this work, the meat compositions were defined based on the maximum uranium density that can be incorporated into the dispersion, which is internationally defined as 45 vol% for the fissile phase. For U_3Si_2-Al dispersions the maximum uranium density is 4.8 gU/cm^3 and for U_3O_8-Al the maximum is 3.2 gU/cm^3.

As a first step, the applied methodology was to produce fuel plates with the highest possible uranium concentration by using exactly the same fabrication procedures currently adopted by IPEN in its manufacturing routine for fuel elements fabrication. Once the difficulties for manufacturing this new type of fuel were determined and studied, a second phase of this study will be to do the necessary technological adjustments in the manufacturing procedures in order to produce high uranium concentration fuel at IPEN. This paper presents the results obtained in the first phase of the work, where the manufacturing difficulties have been found and studied. The causes of the manufacturing problems were identified and the corrective actions were proposed.



(A) (B)

Figure 1. MTR type dispersion fuel. A – fuel element. B – fuel plate.

3. Results and Discussion

From the important parameters for fuel plate qualification, it was found that the length and width of the meat of all produced fuel plates met the specification [5]. Also, the microstructure of the dispersions showed good appearance, as showed in Fig.2. However, difficulties arose regarding to: uniformity in the uranium distribution into the fuel meat and bonding between the meat and the cladding. Also, the cladding and meat thicknesses do not met the specifications. Specification for cladding thickness for the central region of the fuel meat is 0.33 to 0.46 mm and for the extremities of the fuel meat, in the terminal defects region, is 0.25 to 0.46 mm.

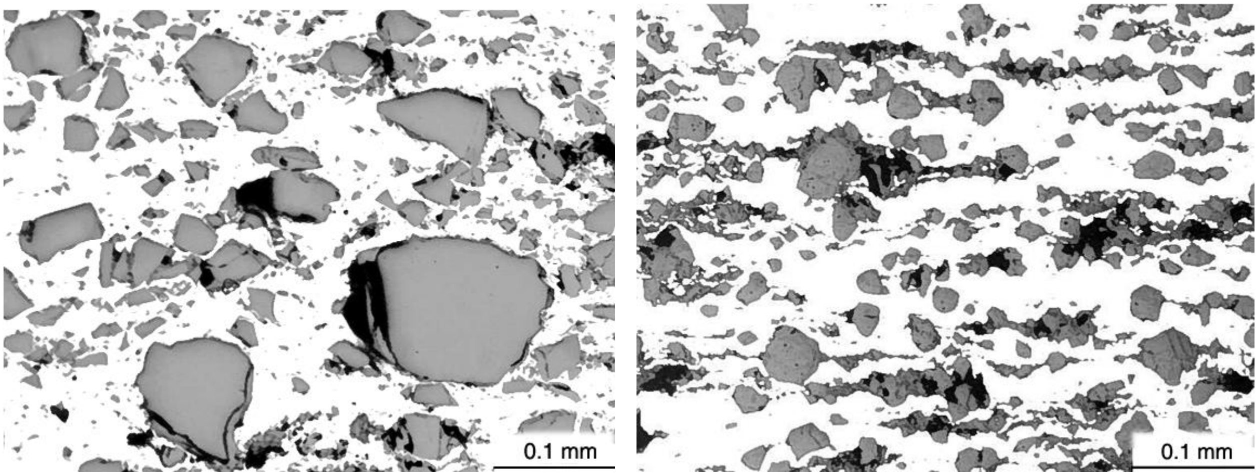


Figure 2. Microstructure of the meat of fuel plates with high uranium concentration. U_3Si_2-Al (left), U_3O_8-Al (right)

The cladding measurement results showed that the cladding thickness for the U_3Si_2-Al dispersion fuel plates was 0.33 mm in the central region, the same value of the minimum specified. In this situation, the entire lot of fuel plates (twenty fuel plates) must be rejected. The reason for this deviation is related to the microstructure of the fuel meat. The specification for the particle size of the U_3Si_2 powder for the fuel based on U_3Si_2-Al dispersion is between 44 and 150 μm, with a maximum of 20 wt% of fines smaller than 44 μm. The specification for the U_3O_8 particle size of the fuel based on U_3O_8-Al dispersion is between 87 and 44 μm, also with a maximum of 20 wt% fines smaller than 44 μm. It can be observed in the U_3Si_2-Al meat that large U_3Si_2 particles are penetrating into the cladding, decreasing its effective thickness. This is a problem, since the aluminum layer over the fuel meat serves to isolate the fuel meat from the reactor environment. Fig. 3 illustrates this phenomenon.

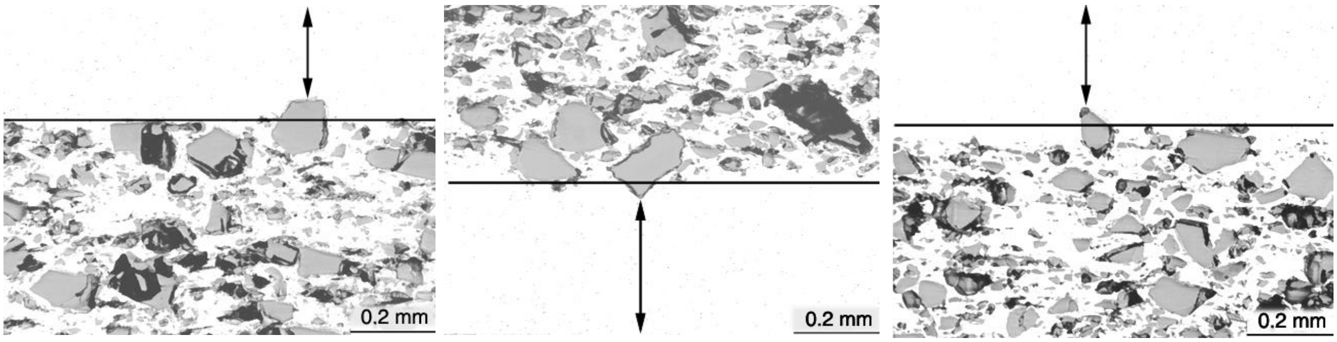


Figure 3. U_3Si_2 particles penetrating the cladding, with reduction of the effective thickness.

Another problem observed refers to the segregation of uranium compound on the underside of the pressed briquette, especially in the case of U_3O_8-Al dispersion. It was observed that during loading the powder inside the die cavity, the much more dense fissile material tends to segregate at the bottom of the cavity, which causes a localized increase in the uranium concentration. If the volume fraction of the uranium compound overcomes the maximum value of 45 vol%, the aluminum matrix of the dispersion is no more continuous, what hinders severely the bonding between the meat and cladding. Fig. 4 illustrates this type of defect.

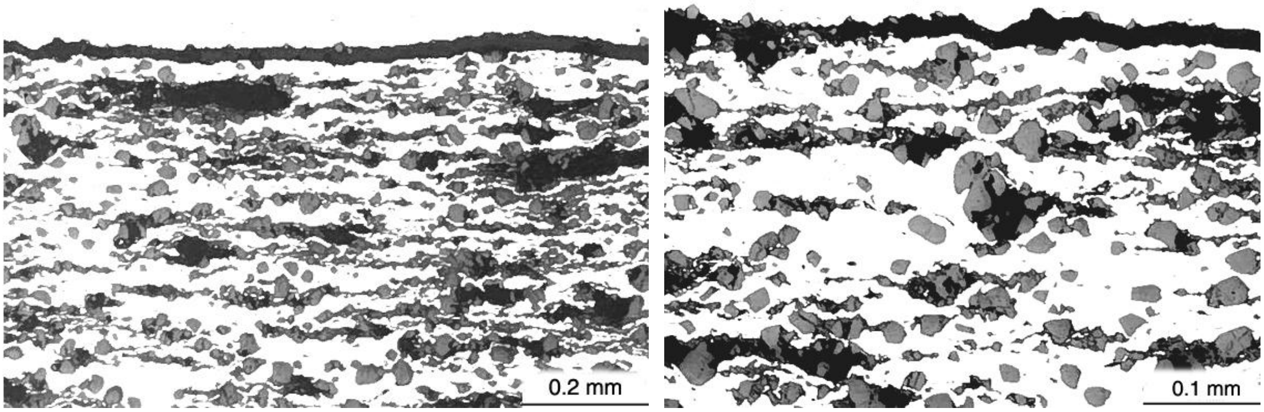
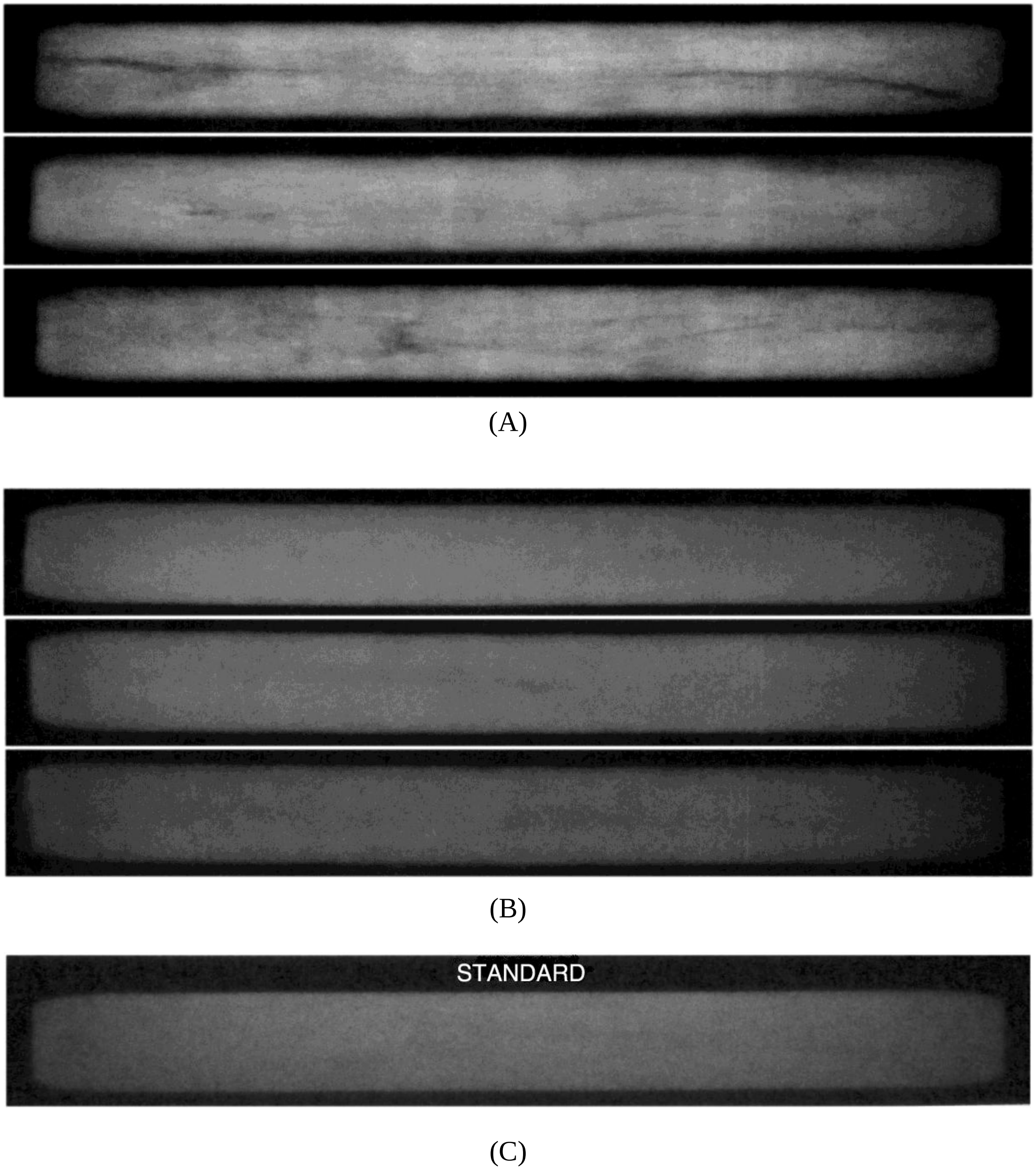


Figure 4. Micrograph illustrating failure in the bonding between meat and cladding, which is caused by the accumulation of U_3O_8 particles on one side of the U_3O_8-Al briquette.

Another difficulty arose from the uranium distribution into the fuel meat. For both U_3Si_2 and U_3O_8, when the volume fraction of the fissile compound is elevated to the maximum (45 vol%), difficulties are found with respect to the homogeneity of the uranium distribution into the fuel meat. This homogeneity is assessed by visual inspection on radiographs. Radiographs are taken from fuel meats and are compared with standard radiographs, which represent the appearance of the minimum level of homogeneity required, according the appearance achieved in the history of traditional manufacturing of fuel plates that have operated in the IEA-R1 reactor with good performance.

Fig. 5 shows radiographs of fuel plates with high uranium concentrations, indicating insufficient homogeneity in the uranium distribution. It was observed that in both cases, U_3Si_2-Al and U_3O_8-Al, that the homogeneity is significantly lower than the standard obtained in the case of fuel plates with low uranium concentrations currently manufactured. The level of homogeneity in the uranium distribution is worse than the minimum acceptable.

The results indicated that the manufacturing difficulties regarding to the bonding quality, uranium distribution homogeneity and cladding thickness are due to the higher volume fraction of uranium compounds present in the fuel meat for either dispersion U_3Si_2-Al or U_3O_8-Al. The problem of segregation during the feeding of the die causes a decrease in the quality of bonding and uranium distribution. Also, the presence of larger amount of particles on the surface of the briquette induces the interpenetration of large diameter U_3Si_2 particles into the cladding, which reduces its effective thickness. Moreover, contrary to what one might expect, the terminal defects (dog-boning) remained within acceptable limits.



(A)

(B)

(C)

Figure 5. Radiographs illustrating the homogeneity of uranium distribution in the core of fuel plates.

A - high concentration U_3Si_2-Al.

B - high concentration U_3O_8-Al.

C - minimum standard of acceptability.

Overcoming these difficulties is the next step of this work, which possibly could be achieved by adjusting the procedure for feeding the die cavity and adjusting the particle size of the U_3Si_2 powder.

4. Conclusion

It was concluded from this work that the procedures currently adopted by IPEN for manufacturing fuel plates with low uranium concentrations cannot be directly applied to the fabrication of fuel plates with high uranium concentration. Fabrication procedures adjustments will be necessary for manufacturing fuel plates with high uranium concentrations. These adjustments are related to the procedure for feeding the die for pressing the briquettes and adjustment of the maximum U_3Si_2 particle size.

Acknowledgments

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