

# QUANTITATIVE DETERMINATION OF URANIUM HOMOGENEITY DISTRIBUTION IN MTR FUEL TYPE PLATES

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## ABSTRACT

IPEN/CNEN-SP produces the fuel to supply its nuclear research reactor IEA-R1. The fuel is assembled with fuel plates containing a  $U_3Si_2$ -Al composite meat. A good homogeneity in the uranium distribution inside the fuel plate meat is important from the standpoint of irradiation performance. Considering the lower power of reactor IEA-R1, the uranium distribution in the fuel plate has been evaluated only by visual inspection of radiographs. However, with the possibility of IPEN to manufacture the fuel for the new Brazilian Multipurpose Reactor (RMB), with higher power, it urges to develop a methodology to determine quantitatively the uranium distribution into the fuel. This paper presents a methodology based on X-ray attenuation, in order to qualify the uranium concentration distribution in the meat of the fuel plate by using optical densities in radiographs and comparison with standards.

## 1. Introduction

The restrictions on the HEU uranium (highly enriched uranium - over 90% in  $^{235}U$ ) international marketing (1) led to the need of increasing the quantity of LEU uranium (low enriched uranium – up to 20% in  $^{235}U$ ) in each fuel element. As the maximum enrichment level required 20% in  $^{235}U$ , this led to the development of new fuels using different types of dispersions, allowing the incorporation of large quantities of uranium fuel in each plate (2). From this new generation of dispersion fuels, the  $U_3Si_2$  allowed IPEN to fabricate fuel plates with a uranium density in fuel meat of  $3.0 \text{ gU/cm}^3$ . This is the kind of fuel fabricated nowadays by IPEN-CNEN/SP to supply the IEA-R1 research reactor.

The IEA-R1 research reactor uses MTR type fuel elements with fuel plates fabricated according to the picture-frame technique (3). The distribution of uranium in the fuel meat is evaluated through a simple visual inspection of radiographies, which is compared to a standard pattern to accept/reject the plate, as shown in Figure 1. The standard is the worst radiography in terms of homogeneity to accept the plate. It was chosen among 180 fuel plates irradiated with good performance in the IEA-R1.



Fig 1. X-ray image illustrating the standard of acceptability.

As this qualification procedure is subjective, it always allows discussions about the acceptability of fuel plates tested for the homogeneity of the uranium distribution. Figure 2 shows examples of radiographies where the uranium distribution homogeneity is visually superior to the standard (Figure 2A) and below the standard (Figure 2B), leading to the rejection of the fuel plate.

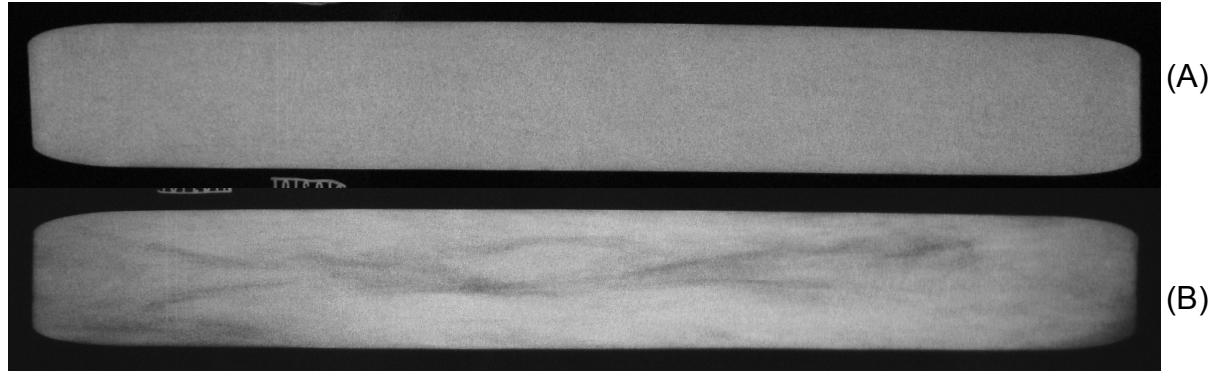


Fig 2. Radiographies illustrating the uranium distribution homogeneity in the fuel meat. (A) superior to the standard, (B) inferior than the standard

The technical specification for the fuel (4) defines values for the surface concentration of  $^{235}\text{U}$ , as shown in table 1, and the range of acceptability of  $\pm 12\%$  for Zone 1 (central zone) and  $\pm 25\%$  for Zone 2 (zone of defects at the ends of the fuel plate meat) of the nominal value specified in the table for the fuel meat, whose geometry is specified in the design of the fuel plate (5).

Tab 1: Values specified for the  $^{235}\text{U}$  density per unit area in the fuel meat.

| Dispersion Material     | Uranium Density              | $^{235}\text{U}$ Surface Density ( $\text{mg } ^{235}\text{U}/\text{cm}^2$ ) | ZONE 1 ( $\pm 12\%$ ) ( $\text{mg } ^{235}\text{U} / \text{cm}^2$ ) | ZONE 2 ( $\pm 25\%$ ) ( $\text{mg } ^{235}\text{U} / \text{cm}^2$ ) |
|-------------------------|------------------------------|--|---|---|
| $\text{U}_3\text{Si}_2$ | $3,0 \text{ gU}/\text{cm}^3$ | 41,2   | 34,1 – 48,2   | 29,1 – 53,3   |

Due to the importance of distribution homogeneity of the uranium inside the meat of fuel plates, mainly for higher power research reactors, as the new Brazilian Multipurpose Reactor (RMB), the objective of present work was to develop a methodology that enables the quantitative determination of  $^{235}\text{U}$  surface density along the fuel plates meat.

## 2. Experimental Procedures

Nuclear Fuel Center of IPEN / CNEN-SP is equipped with an industrial radiography system and an automatic processing for radiographic films. Thus, the attenuation of X-rays was the method chosen to characterize the homogeneity of the fuel meat. The different gray levels of the image provide information on the X-ray attenuation along the thickness direction of the fuel plate. The attenuation is associated with the density of the material traversed and, therefore, in this case, the distribution of uranium in the fuel plate meat. A fuel plate is considered not homogeneous if a uranium concentration is higher or lower as specified limits are detected, as shown in table 1.

The nominal value specified for the  $^{235}\text{U}$  surface density is calculated according to the total mass of uranium that is incorporated in a fuel plate, divided by the nominal area of the meat. The table 1 shows valid density levels for the fuel manufactured by IPEN, which allow quantifying the homogeneity of uranium distribution in the fuel plates meat, as a method of control.

The proposed methodology uses the densitometry evaluation of radiography for fuel meat, made with the aid of an optical transmission densitometer Macbeth TD-904, with window opening 2 mm. The value of optical density is expressed according to Equation (A):

$$OD = \log \frac{I_0}{I} \quad (A)$$

where:

**OD** = optical density;

$I_0$  = Intensity of incident light;

$I$  = Intensity of transmitted light.

The standards were machined from a uranium-aluminum alloy ingot (15.1 mm diameter), manufactured with 30 wt% of uranium. The alloy composition was verified by determination of the uranium concentration by chemical analysis. Nine standards were fabricated with thicknesses between 1.0 and 4.0 mm. All patterns were positioned together with the fuel plate on the table matching with the central area of the radiographic film. An aluminum foil made with the same material (currently Al 6061) and with the same nominal thickness of the fuel plate cladding (0.38 mm) was placed over and under each U-Al alloy standard. The range of thickness of the standards covered the maximum and minimum density of  $^{235}\text{U}/\text{cm}^2$  specified for the fuel meat, allowing the construction of a well characterized calibration curve.

The radiographs were obtained under the conditions of 70 kV, 14.0 mA and 257 seconds (4'17"minutes) for the exposure time. Under these conditions we got the maximum contrast. The radiographs were developed by automatic processing.

The fuel plate meat region in the radiograph was divided into 360 areas of  $1 \text{ cm}^2$ , with 60 rows and 6 columns, as illustrated in Figure 3. A measurement of the optical density was done in the center of each marked square inside the radiograph, as illustrated by the image presented in Figure 3. From each standard, five values of optical densities were determined at four positions around the central point. The arithmetic mean of the readings in each of the standards was used to determine a calibration curve. No optical density of the fuel core was discarded.

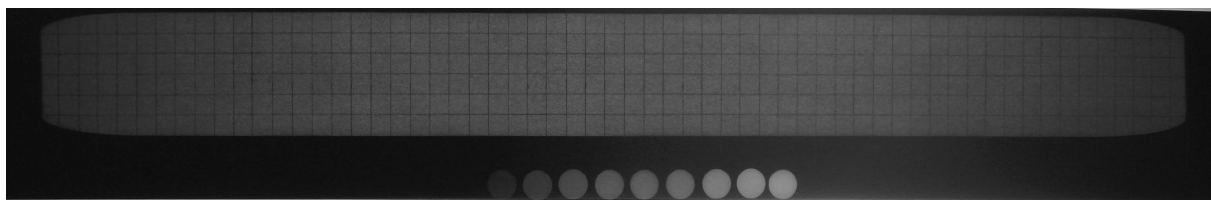


Fig 3. Radiograph with the set of standards, properly drawn to collect measures

Once knowing the uranium concentration in the alloy used to manufacture the standards, the fuel enrichment, mass, thickness and surface area of the standards, it was possible to calculate their  $^{235}\text{U}$  surface density, according to Equation (B). A calibration curve was constructed correlating the optical density measured on a standard with the corresponding  $^{235}\text{U}$  surface concentration. Finally, by using the calibration curve, the optical density values measured on radiographs of the fuel meat can be transformed into values of  $^{235}\text{U}$  surface density ( $D_{US}$ ), as specified.

$$D_{US} = \frac{(m_{\text{standard}} \times C_{U_{\text{Alloy}}} \times C_{^{235}\text{U}})}{A_{\text{standard}}} \quad (B)$$

where:

$D_{US}$  =  $^{235}\text{U}$  surface density ( $\text{mg } ^{235}\text{U}/\text{cm}^2$ ),

$m_{\text{standard}}$  = mass of standard (mg),

$C_{U_{\text{Alloy}}}$  = uranium concentration in the alloy used for standards fabrication (wt%),

$C_{235U}$  = enrichment, or  $^{235}\text{U}$  concentration (wt%),

$A_{\text{standard}}$  = area of standard ( $\text{cm}^2$ ).

### 3. Results and Discussion

The surface density of uranium in each of the standards was calculated according to Equation (B). The optical densities obtained from each of the standards (five measurements) were plotted against their respective  $^{235}\text{U}$  surface densities. The calibration curve was constructed by fitting a curve to the experimental data. A fourth degree polynomial regression expressed the correlation, according to the relationship shown in Equation (C). Figure 4 shows the calibration curve.

$$D_{US} = 247,7 - 286,1 \times OD + 123,8 \times OD^2 - 12,8 \times OD^3 - 2,8 \times OD^4 \quad (C)$$

where:

$D_{US}$  =  $^{235}\text{U}$  surface density,

$OD$  = optical density.

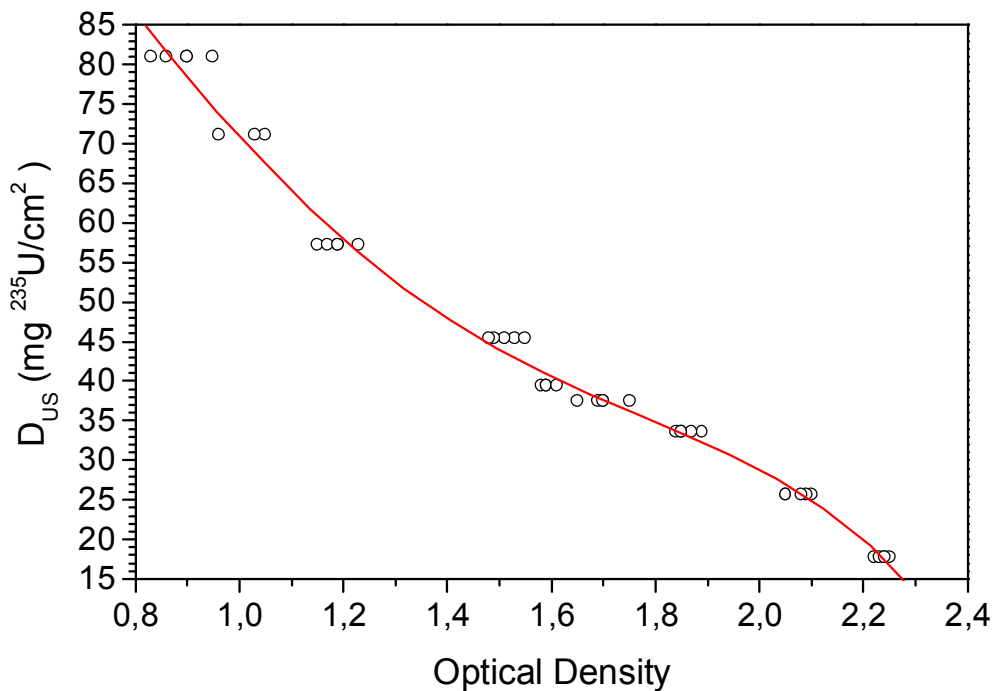


Fig 4. Calibration curve for DO x  $D_{US}$

The range of acceptable variation considering the specifications for the  $^{235}\text{U}$  surface density (5) was incorporated into a chart developed for the final presentation of the data results from this methodology. This graph is shown in Figure 5. This chart shows the maximum (red), medium (green) and minimum (blue) values obtained from six measurements in each of the 60 columns of the radiography of fuel plate meat, as illustrated in Figure 3. These

measurements were performed in a typical fuel plate manufactured by IPEN-CNEN/SP, which was identified as Si-448.

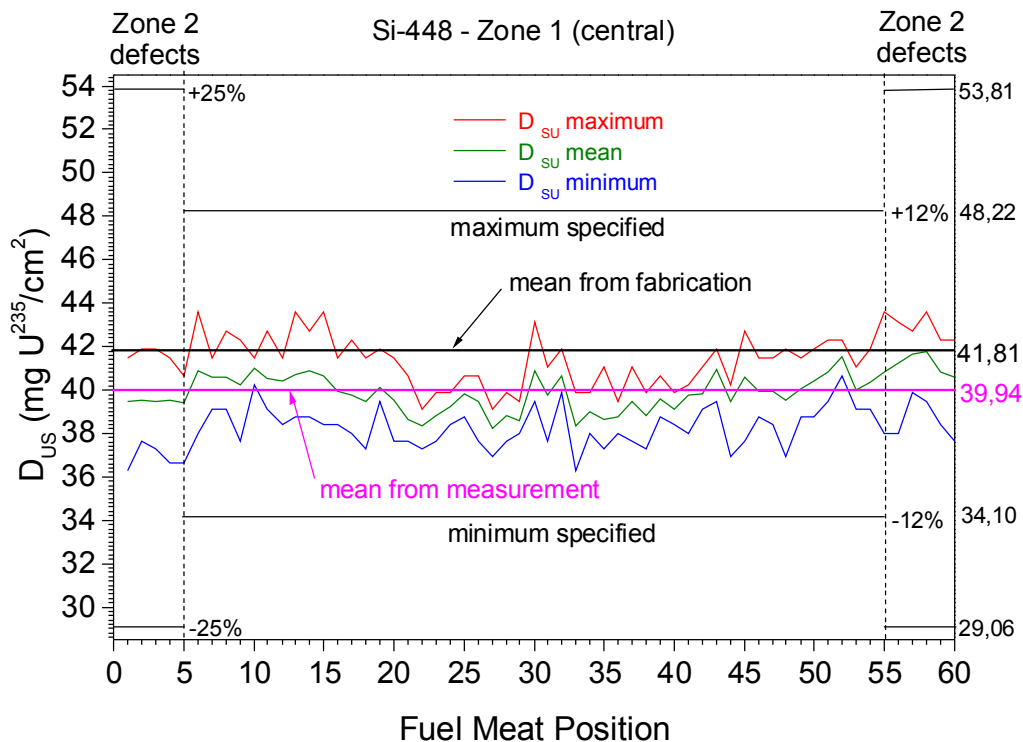


Fig 5. Experimental results for the SI-448 fuel plate.

Note that the nominal  $^{235}\text{U}$  surface density for the fuel plate Si-448 ( $41.81 \text{ mgU}^{235}/\text{cm}^2$ , from fabrication, black line in Figure 5) is very close to the value of the mean for all measured points according to the proposed methodology ( $39.94 \text{ mgU}^{235}/\text{cm}^2$ , magenta line in Figure 5). The nominal value of  $^{235}\text{U}$  surface density of the fuel plate meat is accurate and precise since it is obtained by weighing the uranium incorporated mass in the meat, which is strictly controlled ( $77.6 \pm 0.01 \text{ g}$ ). So, the proximity between these values indicates good accuracy of the method, with an estimated variation of 4.5%. It was concluded that the fuel plate Si-448 met the specification for the current fuel fabricated by IPEN-CNEN/SP to the IEA-R1 research reactor.

#### 4. Conclusion

The technique adopted to develop the meat uranium density methodology showed sufficient sensitivity to generate data that demonstrated the applicability of the proposed method. The method adopted for producing standards also proved to be applicable. The deviation between the real  $^{235}\text{U}$  surface density incorporated into the fuel and the value obtained from the method developed in this work demonstrates variability around 4.5%.

In the future, it is planned to determine the method accuracy and evaluate the possibility of characterizing a larger number of plates simultaneously with the manufacture of other standards sets.

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