CHAPTER 3

NDE OF NUCLEAR FUEL PINS BY RADIOGRAPHIC TECHNIQUES

Experimental studies were made on welded fuel pins containing low Pu enriched mixed oxide fuel pellets with the aim of developing NDE techniques for monitoring plutonium enrichment and detecting plutonium dioxide agglomerates by neutron radiography and gamma autoradiography. Attempt was also made to determine the limit of detection of lack of penetration defect in fast reactor fuel pin welds and tungsten inclusion in fuel pin welds. The effect on radiographic sensitivity due to film fogging was also studied.

This chapter describes in detail the experimental work taken up for monitoring plutonium enrichment and detecting plutonium dioxide agglomerates in nuclear fuel pins using neutron radiography and gamma autoradiography. Results of the investigation of x-radiography of fuel pin closure welds are also presented.

3.1 MONITORING PLUTONIUM ENRICHMENT AND DETECTION OF PLUTONIUM DIOXIDE AGGLOMERATES

3.1.1 Fabrication of MOX fuel pins

A few experimental fuel pins, each containing
MOX pellets with different plutonium enrichment was made at Radiometallurgy Division, Bhabha Atomic Research Centre (Panakkal et al 1985). MOX pellets containing 2.5, 4.0 and 6.0% by weight plutonium dioxide were fabricated in glove boxes by blending, precompaction, granulation, final compaction and sintering in an atmosphere of Ar-8%H. A few uranium dioxide pellets and mixed oxide pellets of annular type was also made. The diameter of the pellets was 12.37 mm (nominal) and the density of the pellets was 94% of the theoretical density.

Mixed oxide pellets containing plutonium dioxide agglomerates ranging in size 125-2000 microns were made. Plutonium dioxide granules in the required size range were mixed with the blended oxide powders (UO$_2$ - 4%PuO$_2$) before compaction and they were sintered at a slightly lower than the normal sintering temperature to avoid total solid solution formation.

The sintered pellets were encased in zircaloy-2 clad tubes (nominal thickness 0.8mm, outside diameter 14mm) by tungsten inert gas welding of endplugs. The pins were subjected to the usual checks such as helium leak test and x-radiography after decontamination.

3.1.2 Characterization of fuel pellets

The composition of the pellets from each category was determined by chemical analysis. The size of plutonium dioxide agglomerates in the pellets was
Fig. 3.1 **PuO$_2$ AGGLOMERATES IN MOX FUEL PELLETS**
confirmed by alpha-autoradiography and ceramography on sample pellets. A typical autoradiograph and the corresponding photomicrograph showing the agglomerates are presented in Fig. 3.1.

3.1.3 Neutron radiography

(a) **Experimental facility.**

Neutron radiography facility set up at APSARA (Dande 1974) was used for the experimental studies. The thermal flux at 540cm from the core was $3 \times 10^6 \text{n/cm}^2 / \text{s}$. The ratio of length to diameter of the collimator used was 90:1. Special fixtures were made to hold the fuel pins during neutron radiography. All the neutron radiography work was carried out by direct technique using gadolinium screen of thickness 25 micron and Agfa Structurix film D2.

(b) **Plutonium dioxide agglomerates and annular pellets**

The neutron radiographs were viewed using a high intensity illuminator. The solid pellets and annular pellets were clearly seen as presented in Fig. 3.2a (Panakkal et al 1985). Most of the agglomerates were also clearly seen. The smallest size of the agglomerates that could be seen was 250 microns (Fig. 3.2b).

(c) **Plutonium enrichment**

The neutron radiographs were scanned using a Joyce-Loebl microdensitometer with an effective aperture of 10 x 500 micron across the image of the pellets
a. SOLID AND ANNULAR PELLET

b. PuO$_2$ AGGLOMERATES

Fig. 3.2 ENHANCED NEUTRON RADIOGRAPHS
(Panakkal et al 1985). The scans were made after normalizing the background density outside the pellets. The density was measured using a calibrated density wedge sensitivity 0.101 per cm. The average difference $\Delta D$ between the density over the pellets and the background density $D$ was calculated from four different scans over each of the three pellets of same composition. The microdensitometric scans are presented in Fig 3.3. The data points were fitted into a straight line

$$\Delta D = 0.95 + 0.16 w$$

where $w$ is the weight% of plutonium dioxide, by the method of least squares with a coefficient correlation of 0.98.

(d) Model for neutron radiography of nuclear fuel pins

In the preceding section, the optical density at the centre of the pellet was correlated to plutonium enrichment. Additional data points were generated from the same neutron radiographs by taking expanded scans (100X) across the diameter of the pellets and the density values $D_j$'s corresponding to thickness segments $t_j$'s were measured. Density values less than 0.8 were not considered. Under these conditions, it was possible to generate 56 data points from pellets of three different enrichment and uranium dioxide pellets. The data points were analyzed to evolve a simple model for transmission of neutrons in neutron radiography
Fig 3.3 Microdensitometer scans of neutron radiographs of MOX fuel pins.
The intensity of attenuated neutron beam is written as

\[
I = I_0 \exp \left\{ - \sum_{i,j} n_i \sigma_{\text{tn}} \left( t_i + \frac{t_i}{\lambda_{c_i}} \right) - \sum_{i,j,k} n_{ij} \sigma_{\text{tn}} \left( t_i + t_j + \frac{t_i + t_j}{\lambda_{c_i} + \lambda_{c_j}} \right) \right\}
\]

\[= I_0 \exp \left\{ - \sum_{j} t_j - \sum_{k} \left( \sum_{i} t_i \right) \right\}
\]

where \( I_0 \) is the incident flux,
\( n \) is the number of nuclei per c.c of type 'i',
\( \sigma_{\text{tn}} \) is the total microscopic cross section of the nuclide 'i',
\( \lambda_{c_i} \), \( \lambda_{c_j} \) are the number of species of nuclei present in the fuel and the clad respectively,
\( t, t \) are the distances traversed by the neutrons in the fuel pellet and clad materials respectively

and

\[
\sum_{\text{fuel}}, \sum_{\text{clad}}
\]

are the total macroscopic cross-section of the fuel and the clad respectively.

The exponential relationship for the attenuation is valid only if neutron removal cross-
section is used. The above equation is rewritten in a general way for the number of neutrons removed as

\[ I - I = f(x) \quad \text{(3-3)} \]

where,

\[ x = \sum_j t \quad \text{(3-4)} \]

assuming the contribution by the clad as a constant.

Optical density \( D \) obtained in a fine grain film with a direct exposure technique bears a linear relationship with exposure (Hawkesworth 1977, Harms 1977) given by

\[ D = K I \quad \text{(3-5)} \]

where \( K \) is a constant. The value of \( K \) depends on the nature of the film characteristic curve, film registration efficiency and yield of secondary radiation in gadolinium.

The difference in density \( \Delta D \) is written as

\[ D = D - D = k f(x) \]

\[ = \phi(x) \quad \text{(3-6)} \]

This functional relationship was determined from the microdensitometric data. The values of

\[ x = \sum_{i=1}^{R} n \sum_{i=1}^{R} t \]

at each of the thickness segment was calculated assuming published values of total macroscopic cross-section of \( -1 \) neutrons of velocity 2200 ms (Mughalghab and Garber
1973). The data was fitted into linear, exponential, logarithmic, power and second degree polynomial. The coefficient of correlation $r$ and standard error of the estimate $\sigma_\xi$ are presented in table 3.1. It has been observed from Fig 3.4 that the scatter of the points is very much reduced with a standard error of the estimate 0.03 for equation of type

$$D = a + b x + \sum_{j} c x^{2}_{j} \quad (3-7)$$

With the aid of one or two radiographs, it is possible to determine the constants of the equation written in matrix notation

$$D = AX \quad (3-8)$$

where $D$ is the density ($\Delta D$) vector,
Fig 3.4 Plot of optical density of the neutron radiograph Vs. product of total macroscopic cross-section and thickness ($\Sigma \tau$).
A is the vector of the constants of equation (3-7)

X is the matrix of powers of x values at different j thicknesses.

Using this, the optical density of plutonium bearing fuel pins of varying composition and diameter in a neutron radiography set-up can be predicted. For example, a density difference of 1.9 is expected for a small diameter (4.18mm) fast reactor mixed oxide fuel (0.7U-0.3Pu)O.

(e) Discussion

Experiments on neutron radiography of welded mixed oxide fuel pins showed that it was possible to detect the presence of annular pellets in the sealed pins. This is useful from the point of view of nuclear safeguards. Normal x-radiography and passive gamma scanning are not useful for differentiating the annular pellets from solid pellets.

Neutron radiographs revealed plutonium dioxide agglomerates down to 250 microns in low plutonium enriched fuel pins. The agglomerates lying in the centre or in the periphery of the pellets could be detected. Image processing of the radiographs was useful in removing the unwanted background noise from the radiographs resulting in better quality radiograph. Neutron radiography is also quite useful for monitoring composition of plutonium bearing fuel pins. A model for transmission of neutrons in neutron radiography of fuel
pins has been proposed based on the analysis of detailed microdensitometric data.

3.1.4 Gamma autoradiography

(a) **Experimental**

Industrial X-ray films were loaded into PVC cassette and the fuel pins were kept in contact with the cassette. The time required to achieve a density of 1.7 over a pellet of 6.0% PuO₂ enrichment was 17 h for D10 film. The film in the cassette was wrapped around the fuel pins containing annular pellets and plutonium dioxide agglomerates. The exposure time was 17h for D10 film and 65 h for D7 film (Ghosh et al. 1984).

(b) **Discussion**

The autoradiograph of fuel pins with different composition was scanned using a microdensitometer (aperture size 15 x 1000 microns and wedge of sensitivity of 0.101 D per cm). Fig 3.5 presents the microdensitometric scans for which a linear calibration graph was obtained. The minimum difference in weight percentage (1.5%) of plutonium dioxide in the pellets investigated corresponds to a density difference of 0.35D. The accuracy of measurement of enrichment is governed by the noise due to film graininess and error in the density measurement. A value of 0.01 (equivalent to 0.005% by weight of plutonium dioxide) is attributed to these factors. Error due to difference in processing conditions can be eliminated by taking the
Fig 3.5 Microdensitometer scans of gamma autoradiograph of MOX fuel pins:

a) 6.0% PuO₂-UO₂
b) 4.0% PuO₂-UO₂
c) 2.5% PuO₂-UO₂
autoradiograph of the standard fuel pin along with the production batch. In practice, the difference in enrichment normally encountered in a fuel fabrication laboratory is not less than 0.5% by weight and hence the method is useful as a simple and inexpensive technique for detecting pellets of incorrect composition.

Gamma autoradiograph of the fuel pin containing the agglomerates revealed plutonium dioxide agglomerates down to 250 microns lying in the outer region of the pellets. The autoradiographs were compared with neutron radiographs of the same fuel pins. Gamma autoradiography, however, could not distinguish the annular pellets from solid pellets because of the self absorption of the gamma rays by the fuel.

3.1.5 Comparison between neutron radiography and gamma autoradiography

On the basis of the results obtained, the usefulness of neutron radiography and gamma autoradiography for NDE of mixed oxide fuel pins are compared (Panakkal, Ghosh and Roy et al 1988). Neutron radiography revealed clearly the presence of annular pellets while gamma autoradiography failed due to its inherent limitation. Both neutron and gamma radiographs could detect plutonium dioxide agglomerates down to 250 microns. Neutron radiography could detect them irrespective of the location. Alpha autoradiography is the standard quality control check
for homogeneity of plutonium dioxide carried out on a sample basis. The occurrence of the agglomerates is statistical in nature and hence it is probable that some of them may lie near the surface and will be detected by gamma autoradiography. Gamma-autoradiography is simple and inexpensive to carry out on all fuel pins compared to neutron radiography which is not economical and practical to carry out on a 100% basis. The exposure time of 17 h is not very inconvenient since the test is to be carried out at the final stage of fabrication.

Both neutron radiography and gamma-autoradiography have more or less the same sensitivity for monitoring plutonium enrichment. Neutron radiography, however, gives information about the whole cross-section of the fuel pellet.

3.2 X-RADIOGRAPHY OF FUEL PIN WELDS

X-radiography is used to check the integrity of the endplug welds made by TIG welding. Defects like lack of penetration, porosity and inclusions are revealed in radiography.

3.2.1 Use of a defect standard

X-radiography of fast reactor fuel pin endplug welds (5.10mm diameter, thickness 0.37mm, material SS316) is performed using a shape correction block which consists of a rectangular block with drilled holes to accommodate the fuel pins. A defect standard
for quantitative evaluation of defect size detectable by radiography was proposed (Ghosh et al 1983a). A modified plug containing grooves of depth 0.025mm and varying width from 0.04 to 0.2 mm was used as a defect standard (Fig 3.6a). The radiographs were scanned along the plug clad gap using an effective aperture size 10 x 300 microns of the microdensitometer and a calibrated density wedge of sensitivity 0.019D per cm (Fig 3.6b).

A more objective assessment of the weld radiographs comparing the defect signals with those from defect standard was possible using microdensitometry. The operator dependence is reduced by adopting microdensitometry. Grooves of size 0.025 x0.042 mm was detected during the radiography of FBTR fuel pin welds. However, it must be realized that the defects in a typical production welds would not be smooth and regular as in a defect standard.

3.2.2 Detection of tungsten inclusion

The specification of fuel pin endplug welds do not allow any indication of tungsten inclusion in the radiograph. Experiments were carried out to estimate the minimum size of radiographically detectable tungsten inclusion simulated by tungsten powder and wires on fast and thermal reactor fuel pin endplug weld configuration (Panakkal, Chandrasekharan and Ghosh 1985). Tungsten wires of diameter 50 and 100 microns and powders whose larger dimensions were 50,100,150 and 200 microns were
Fig 3.6  a) Defect standard used for FBTR weld inspection.

b) Microdensitometer scans.
used. Stainless steel block of thickness 5.5 mm and zircaloy block of thickness 15.5 mm were used to evaluate the detectability of tungsten inclusion. In the fast reactor fuel pin weld configuration (5.5 mm steel), it was possible to detect 50 micron wire and 100 micron particle. Contrast sensitivity $C_w$ was calculated from the scans as

$$C_w = \frac{\Delta D}{\Delta X} \quad (3-8)$$

where $\Delta D$ is the density difference caused by tungsten wire of diameter $\Delta X$ and are presented in table 3.2.

<table>
<thead>
<tr>
<th>Material</th>
<th>Film</th>
<th>Film density</th>
<th>Contrast sensitivity $C_w$/mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.5 mm steel</td>
<td>D2</td>
<td>2.1</td>
<td>7.0</td>
</tr>
<tr>
<td></td>
<td>D4</td>
<td>2.0</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td>D7</td>
<td>2.1</td>
<td>6.0</td>
</tr>
<tr>
<td>15.5 mm zirconium alloy</td>
<td>D4</td>
<td>2.1</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>D7</td>
<td>2.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Figure 3.7 presents microdensitometer scans over tungsten wire over 5.5 mm steel. Tungsten wire of 100 micron diameter was clearly seen in thermal reactor fuel pin weld configuration. Deterioration of the contrast sensitivity compared to stainless steel observed was due to the higher voltage used. The image of 200 micron
Fig 3.7 Microdensitometer scans of radiograph of tungsten wire kept on a 5mm stainless steel block.
powder (larger dimension) was barely seen in D4 film and not seen in larger grained D7 film. But it was possible to see 100 x 300 micron wire in both the films and was taken as the limit of detection. This apparent discrepancy may be attributed to the assumption that the thickness of the particle was the same as the larger dimension. With the advent of microfocus x-ray systems, the minimum size of detectable tungsten inclusion is expected to be less than the values reported in this work.

3.2.3 Effect of film fogging on radiographic sensitivity

In nuclear fuel fabrication facilities using high burn-up plutonium, the chances of film fogging by radiation from the pellets cannot be ruled out (Smith 1973). This may affect the radiographic sensitivity during the radiography of welds.

Experiments were conducted to see the deterioration of sensitivity due to pre-exposure. Commercially available industrial radiographic films (Agfa Gevaert, Kodak) of different speeds were exposed to different extent of radiation to get the required density if processed. The exposure required for this purpose was determined separately. A 5mm stainless steel block simulating fast reactor fuel pin welds was kept over the pre-exposed film and given the required exposure for radiographing FBTR endplug welds. The exposure for each category of the film was such that a
density of 2.2 was achieved on the processed normal (not pre-exposed) film. The density on the processed film will be total of the density due to normal exposure and that due to pre-exposure. Radiographic sensitivity was assessed using image quality indicators of DIN type and ASTM type and the results are given in Table 3.3 (Ghosh et al 1983b).

As judged by the discernability of 0.1 mm diameter and larger wires, there was no significant variation in the radiographic sensitivity upto a pre-exposed density of 2.0 for fine grain films like Agfa Gevaert D2, D4 and Kodak MX. There appears to be a slight decrease in detail sensitivity if ASTM penetrameters are used as criterion. Microdensitometer scans, however, showed deterioration in contrast with increase in pre-exposure, which was not noticed in normal viewing by eye.
### Table 3.3

#### Radiographic Sensitivity of Films Pre-Exposed to Different Densities

<table>
<thead>
<tr>
<th>Film</th>
<th>Pre-exposed density</th>
<th>Radiographic exposure mA.min</th>
<th>Total density</th>
<th>Penetrometer Visibility</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Diameter of the smallest wire seen (mm)</td>
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<tr>
<td><strong>D2</strong></td>
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<td></td>
<td></td>
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<tr>
<td></td>
<td>0.0</td>
<td>38</td>
<td>2.0</td>
<td>0.1</td>
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<td></td>
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<td>3.1</td>
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