Non destructive test nuclear fuel U₃Si₂/Al 4,8 g U/cm³ post irradiation with 60% burn up research reactor

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ABSTRACT

Development of nuclear fuel research on uranium silicide / aluminium with the aim to extend its use in the reactor is done by increasing the fuel density from $2.9~g~U/~cm^3$ to $4.8~g~U/~cm^3$. Performance of fuel in the research reactor is investigated by non destructive tests such as : gamma scanning, visual observation and thickness measurement of the fuel plates. These experimental results show that burn-up distribution of ^{235}U fuel follows a polynomial rank 3 pattern. In case of fuel plate irradiated up to 60 % burn-up in the middle position the earlier value increases to 77 % burn-up with very small swelling (less than 1%) and no corrosion that can potentially release of uranium. It is concluded that uranium silicide fuel elements have good performance.

Keywords: Post irradiation, nuclear fuel, 60 % burn-up.

1. **Introduction**

Multipurpose research Reactor GA Siwabessy in indonesia is widely used for research related to testing nuclear materials and for the production of radiopharmaceutical , which is needed in the health sector. For the effective reactor operation we need a fuel that can be used for a longer time at normal operating power reactor. The reactor fuel uses uranium silicide / aluminum with uranium loading level of 2.96 g / cm³ which is still considered as not optimal $^{[1]}$; then the development of fuel with an increased content of uranium, fuel uranium silicide / aluminum with uranium loading level of 4.8 g / cm³ $^{[2,3]}$ is considered. Nuclear fuel element are irradiated in reactor cores with neutron flux of about $10^{14}\,\text{n}\,/\,\text{cm}^2.\text{s}$. As a result there will be changes in the composition, the chemical properties of the fuel and the fuel cladding. There is a large rate of change in accordance with the power generated by the fuel elements for use in the reactor $^{[8,9,10]}$.

The changes that occur in the fuel are due to fission reaction of ²³⁵U which produces fission product such as radionuclide ¹³¹I (Iridium), Xe (Xenon) and Cs (Cesium) radioisotope. The fuel element in reactor produces fission products, decays and neutron capture. Effect from neutron capture Pu is that ²³⁸U produces ²³⁹Pu and ²⁴¹Pu with a role as fissile. Cladding material will also experience neutron capture reaction; the contact between cladding surface of the reactor and cooling water will cause the oxidation reaction on the surface. Therefore, any change in the composition of the fuel and cladding material will cause a change in the physical properties, such as: a decrease in thermal conductivity and changes in microstructure and increase in the volume of fuel in the fuel elements ^[4,7]. The higher burnup fuel will lead to an increase in changes in fuel composition and physical elements. Enhancing density of fuel will affect the toughness of fuel elements. The rate of change of toughness is influenced by: the exchange of heat, pressure, burn-up, irradiation and other factors ^[2]. In order to determine the level of safety-related changes in the reactor, then

the post-irradiation examination of the fuel elements with non destructive test methodes is envisaged.

Non Destructive Test (NDT) is a central part in the post-irradiation examination. This technique is often used to obtain a picture of the post-irradiated fuel elements rapidly and as a preliminary approach to narrow observation to area degradation of fuel elements after irradiation^[11,12]. Data acquisition results of these tests contribute to determine the sampling areas for more detailed results with destructive testing. The NDT method will produce critical data and important information to determine the decrease of cladding performance, which is the main safety aspect in evaluating the inspection of fuel elements. Fuel plate elements can't be separated, because the majority of toughening phenomenon in fuel element depends on alloy between the fuel and cladding. The purpose of this study was to gain an overview of the performance of fuel element plate U₃Si₂/Al with a loading level 4,8 g U / cm³, by visual inspection, thickness measurement and localized gamma (gamma scanning) after irradiated to 20%, 40% and 60 % burn-up.

2. Experimental Procedure

Sample of fuel plates element U₃Si₂/Al uranium loading level 4,8 g U / cm³ were supplied by PT. INUKI (Persero) nuclear industry of Indonesia and irradiated in the core reactor G.A Siwabessy with varying burn – up. After cooling more than 101 days, the sample was tranferred to hot laboratory RMI – facility using canal channel, as shown in figure 1. in this research the experiments conducted are visual observation using binoculars Nikon and digital cameras DLSR Canon 50D then measurement plate thickness fuel elements each performed at 5-positions on the direction of the long plate and 3-position measurement in the direction of the width plate using a thickness gage Mitutoyo and the last, scanning gamma rays done at a distance of 5 mm each variation with time of enumeration 500 seconds for 20% -burn up, 300 seconds for 40%-burn up and 200 seconds for 60% using a gamma spectrometer ORTEC with liquid nitrogen as a cooling high purity germanium detector (HpGe), ⁶⁰Co as standards for energy calibration and ¹⁵²Eu as standards for detector efficiency calibration.

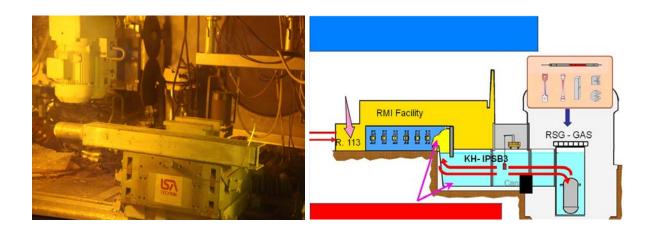


Figure 1. Radiometallurgy Installation (RMI – Facility) Design.

3. Results And Discussion

3.1. Fuel element specifications

Fuel plates element U_3Si_2/al uranium loading level 4,8 g U / cm³ which has been prepared has a specification as in Table 1 ⁽¹⁴⁾, and dimensions plate of fuel elements as shown in Table 2.

| Fuel plates element | | | |
|---------------------|--|--|--|
| | | | |
| | | | |
| | | | |
| | | | |

| Classification | Value |
|---|-----------------------------------|
| Enrichment ²³⁵ U | $19.75^{+0.2}$ % $19.75_{-0.5}$ % |
| Number of fuel element in 1 bundle | 3 Fuel element |
| Composition ²³⁵ U / fuel element | $18,36 \pm 0,30 \mathrm{g}$ |
| Composition ²³⁵ U / bundle | $55,08 \pm 3,80 \text{ g}$ |
| Zona 1 tolerance | nominal \pm 20 % |
| Zona 2 tolerance | nominal + 25 % |

Table 2. Dimension measurement data

| No | Kode Fuel | Length,mm | Width,mm | Height,mm |
|----|-----------|-----------|----------|-----------|
| 1 | CBBJ 249 | 629,00 | 70,70 | 1,40 |
| 2 | CBBJ 250 | 629,00 | 70,71 | 1,39 |
| 3 | CBBJ 251 | 629,00 | 70,70 | 1,39 |

3.2. Hystorical Fuel element irradiation

Fuel plates element U₃Si₂/al uranium loading level 4,8 g / cm³ has been irradiated in the core reactor G.A Siwabessy. Reflector reactor core configuration as shown in Figure 2 illustrates the positions: fuel elements (FE), the control element (CE), Be reflector element (BE), Be reflector element with plug (BS +), the position of irradiation (IP): central irradiation position (CIP), rabbit system pneumatic (PNRs), Rabbit hydraulic system (HYRS). The fuel element for the irradiation process consists of three plates of fuel no3: CBBJ 249 / no 7: CBBJ 250 and no: 19 CBBJ 251, entry into the reactor core at the position of the G-7, and operated 29-11-2008 to 26-05-2009, when EBU CBBJ-249 was released from the G-7 (reactor core). EBU-1 consists of two plates of fuel no: 7 CBBJ 250 and No. 19 CBBJ251 entered the reactor core (at the position G-7) and operated in the reactor core 16-6-2009 to 16-12-2009, when the plate No. 19 CBBJ 251 was issued. EBU-1 consists of first fuel plate no: 7 CBBJ 250 included in the core at the position G-7 to achieve of 60% burn-up.

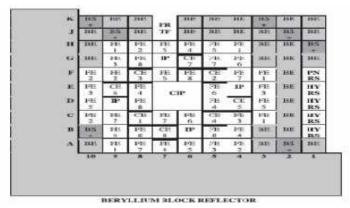


Figure 2. Core-reflector configuration G.A Siwabessy reactor.

3.3. Scanning Gamma (gamma-spektroscopy)

During irradiation fuel elements in reactor cores experience fission reaction of ²³⁵U and neutron capture in ²³⁸U that form ²³⁹Pu. ²³⁹Pu when exposed to neutrons also have fission. In the ²³⁵U and ²³⁹Pu fission reaction energy releases in the form of heat. The probability of occurrence of such reactions depends on the number of neutrons that cause reactions influenced by the position of the plate fuel elements and neutron absorber layout of the fuel element. The fuel element located in the core in a standing position. Distribution of atomic fission reaction is done by measuring fission product that is used as an indicator of the fraction of fuel. Radionuclides election considerations are: (1). Radionuclide that has high fission yield of the fission reaction ²³⁵U. (2). Have small neutron uptake (3) has a long half life. Table 3 shows the specification radionuclides ¹³⁴Cs and ¹³⁷Cs that meet the above mentioned requirements.

Table 3. Radionuclide ¹³⁴Cs and ¹³⁷Cs specification

| Isotope | T _{1/2} (year) | σ (barn) | E (keV) | % yields |
|-------------------|-------------------------|----------|---------|----------|
| ¹³⁴ Cs | 2,1 | 24,3 | 605 | 0,976 |
| ¹³⁷ Cs | 30,1 | 0,05 | 662 | 0, 851 |

Results for localized energy gamma peaks at 605 keV (¹³⁴Cs) and 662 keV (¹³⁷Cs) in various positions on the plate element uranium silicide fuel ²³⁵U yield 20% and 40% burn-up, as shown in figure. 3 and figure. 4. Distribution of ¹³⁴Cs and ¹³⁷Cs in uranium silicide fuel element plate loading level of 4.8 g / cm³ follows a normal distribution pattern and fuel element of 60 % burn up follow a polynomial distribution rank 3 pattern as shown in figure. 5. These measurements have smaller fluctuations than 4% of this value smaller than the deviation of gamma radiation for the gamma spectrometer equipment. Measurements show that the fuel in the plate have normal fission reaction and there is no mismatch fission reaction to cause excessive local heating.

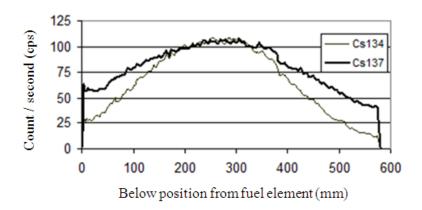


Figure 3. Distribution pattern for 134 Cs and 137 Cs in nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³, with 40% theoretical burn-up.

Burnup calculation of irradiation in a reactor for a plate burning by 40% was performed. Based on these data, the distribution of burnup at each axial location within the fuel element plate is calculated by the following equation:

$$F_{i,j} = \frac{A_{i,j}}{A_{i,r}} F_{i,r}$$

Fi, j : Fraction of fuel calculation position j on the plate i

Fi, r: Fraction of mean fuel on a plate i

A i, j: Activity cesium in position j on the plate i

A i, r: Activity of mean cesium on a plate i

Calculation of fractions at each location based on measurements of ¹³⁷Cs showed that the maximum burnup in the fuel is in the region between 235 mm to 345 mm with a value of 52.7% ²³⁵U burnup, while based on the measurement of cesium-134 burnup area is located at 250 mm up to 320 mm from the bottom of the fuel, with a value of 66.14% ²³⁵U burnup (Fig. 4). Maximum difference in burnup is caused by differences in ¹³⁷Cs source of fission ²³⁵U; ¹³⁷Cs decays more slouly in comparison with ¹³⁴Cs. In addition, absorption value of neutrons for ¹³⁴Cs are greater than ¹³⁷Cs, ¹³⁴Cs at the end regions is indicated by a count of radioactivity- of fuel smaller than ¹³⁷Cs in the range between 130 mm to 370 mm from the bottom plate fuel elements (Fig. 5). Therefore, ¹³⁴Cs radioactivity in the fuel elements is not only determined by the fraction of ²³⁵U in the fuel but are affected by other factors which contribute to the formation of 134Cs. Based on the circumstances mentioned above burnup data from ¹³⁷Cs, and for maximum 60 % burn up in the fuel region between 250 mm to 300 mm are shown in table 4.

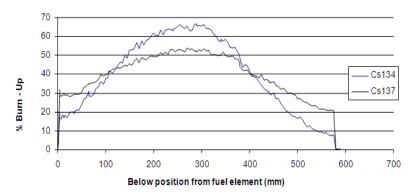
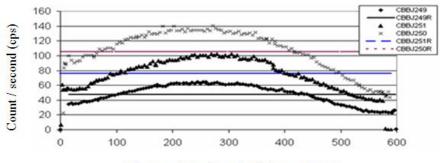


Figure 4. Distribution pattern burn-up nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³ with 40% experimental burn-up.



Below position from fuel element (mm)

CBBJ249 & CBBJ 251 CBBJ: 60% burnp-up

Figure 5. Distribution pattern 134 Cs and 137 Cs nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³ with 60% experimental burn-up.

Table 4. 60 % burn up value nuclear fuel U₃Si₂/Al uranium loading level 4,8 g/cm³

| Measurement position (from below) (mm) | Burn Up Measurement (% ²³⁵ U) |
|--|--|
| 582.5 | 26.65 |
| 250 | 76.35 |
| 6.25 | 47.52 |

3.4. Visual Observation

The results of visual observation of fuel plates element U_3Si_2/Al uranium loading level 4,8 g U / cm³ irradiated in the core reactor with 15 MW power research reactor ,are shown in Fig. 6-8. Cladding surface of the plates have been homogeneously oxidized and do not indicate oxidation pitting on the surface. Surface oxidation does not show any noticeable color differences in the loading area due to overheating as a result of the nuclear reactions in the fuel. The surface oxidation is in the normal range for a plate type fuel element cladding.



Figure 6. Visual data for nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³ with 20% burn-up.



Figure 7. Visual data for nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³ with 40% burn-up.



Figure 8. Visual data for nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³ with 60 % burn-up.

Based on the calculation of the temperature of the fuel reactor in a steady state operating condition, the heat distribution pattern is shown in figure. 9 where the temperature reached a maximum in the region from 30 cm until 40 cm from the bottom position of fuel element.

Maximum difference between high and low temperature overall in the fuel is 40 0 C, it is concluded that the corrosion effect on the surface fuel element (cladding Almg) has relatively the same value.

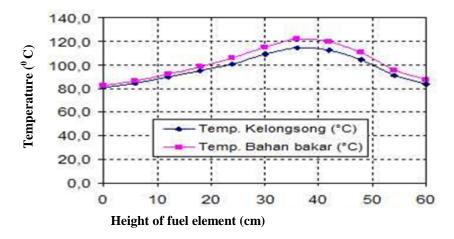


Figure 9. Distribution heat pattern of fuel element and cladding for nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³.

3.5. Mesurement of thickness

In accordance with the level of fission reactions that occur in the fuel, the volume will change constantly in accordance with burnup achieved by the fuel. Nb, Y, Zr, Mo, Ru, Te, Rh, Pd, Rb, I, Ba and Sr can cause swelling in the cladding fuel elements. Swelling due to solid fission product is not affected by temperature. The fission reaction also produces gaseous fission product which will form gas bubbles in the fuel meat matrix. The temperature rise resulting from fission reactions would lead to increased pressure of the fission product gases. The volume increases cause an increase in tension in the cladding at high burnup mainly caused by gaseous fission product, this will cause swelling of the plates fuel elements.



Figure 9.1. Visual data thickness gauge nuclear fuel U_3Si_2/Al uranium loading level 4,8 g/cm³ with 20% burn-up post irradiation.

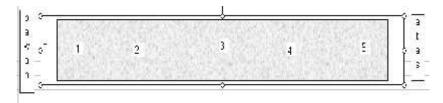


Figure 9.2. Measurement Position thickness of the fuel element

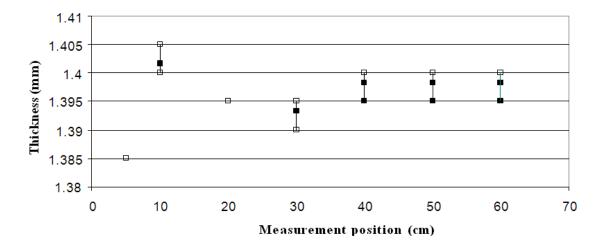


Figure 9.3.. Fuel plate element 20% burn – up

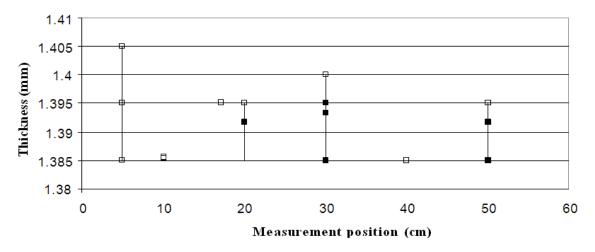


Figure 9.4. Fuel plate element 40% burn – up

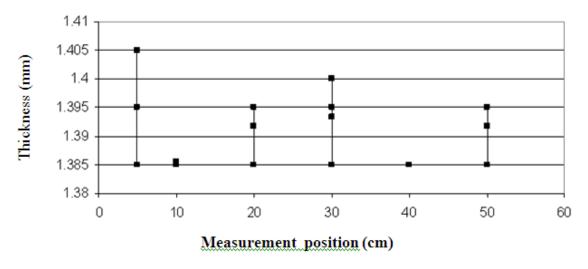


Figure 9.5. Fuel plate element 60% burn – up

Figure 9. Distribution thickness data of nuclear fuel U₃Si₂/Al uranium loading level 4,8 g/cm³.

Uranium in the nuclear fuel U_3Si_2/Al is homogeneously dispersed in the meat fuel (^{235}U 0.051 g/cm²) with an aluminum matrix. In the irradiated fuel element plate. Fission product

of ²³⁵U will be dispersed in a matrix of aluminum according to burn-up in that position. At the center of the plate (burn-up Maximum: 76.5%), this means about 0,76.5 g / cm³ ²³⁵U who have fissioned to form fission products that are bound by the matrix. During irradiation in a reactor, the fuel element temperature changes will be confined by the aluminum matrix in the fuel element, then the increase in volume generated by solid fission products are relatively small when compared to bond aluminum in the fuel matrix. Therefore swelling fuel element caused by solid fission product and gas fission product is very small. The condition is shown in Fig. 9 (Distribution thickness data of nuclear fuel U₃Si₂/al uranium loading level 4,8 g/cm³). The thickness after irradiation with 20%, 40% and 60% burn-up is between 1,385 mm to 1,405 mm compared to thickness before irradiation between 1.39 mm to 1.40 mm, Thus the thickness change is still within the permitted limits (does not disturb the flow of cooling water in the fuel bundle).

4. Conclusion

The post observation above show that: the plate uranium silicide fuel element / Al (U_3Si_2/Al) uranium loading level of 4.80 g / cm³ after irradiation in the reactor core to achieve the theoretical average burn-up of 20%, 40% and 60% indicated distribution of burn-up in accordance with the normal distribution (in the middle of the fuel element with a burn-up of 60%, reaching a burn-up 77%), swelling of plate fuel element is very small (less than 1%) and no corrosion which could potentially release uranium occurs on the fuel plate element.

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