



Original Article

Delayed fast neutron as an indicator of burn-up for nuclear fuel elements

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ABSTRACT

Feasibility study of burn-up analysis and monitoring using delayed fast neutrons was investigated at Missouri University of Science and Technology Reactor (MSTR). Burnt and fresh fuel elements were used to collect delayed fast neutron data for different power levels. Total reactivity varied depending on the burn-up rate of fuel elements for each core configuration. The regulating rod worth was $2.07\text{E-}04 \Delta k/k/in$ and $1.95\text{E-}04 \Delta k/k/in$ for T121 and T122 core configurations at 11 inch, respectively. Delayed fast neutron spectrum of F1 (burnt) and F16 (fresh) fuel elements were analyzed further, and a strong correlation was observed between delayed fast neutron emission and burn-up. According to the analyzed peaks in burnt and fresh fuels, reactor power dependency was observed and it was determined that delayed neutron provided more reliable results at reactor powers of 50 kW and above.

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1. Introduction

Burn-up is a crucial parameter for reactor operation, performance, and safety. One of the essential burn-up determination and spent fuel monitoring techniques is the non-destructive analysis (NDA), a well-recognized method that can be implemented using thermal neutrons and photons from fission products [1–5]. Jordan and Perret [6] developed an NDA method with delayed thermal neutron measurement to investigate burn-up credit for PWR fuel pins. Akyurek and Usman [5] obtained burn-up and Pu conversion rates of MSTR fuel elements with delayed neutrons spectroscopy. The primary purpose of this study is to investigate the fast delayed neutrons as a burn-up indicator for both Uranium and Plutonium-based nuclear fuels using a research reactor.

Uranium fuels have been used for energy production in the nuclear industry for decades [7]. Mixed Oxide (MOX) fuel is another type of nuclear fuel that is used in the nuclear industry and gaining popularity [8]. MOX fuel shows many similar performance characteristics as Uranium fuel [9,10]. Therefore, MOX fuel can be analyzed by the NDA method using both gamma and neutron

spectroscopy [11]. The determination of burn-up for a fuel element using gamma spectroscopy has been obtained for more than 50 years [12]. Burn-up indicators are the most critical tool for burn-up credit calculations using NDA gamma spectroscopy. ^{137}Cs , ^{134}Eu , and ^{134}Cs are widely used indicators by various experimental and theoretical methods to obtain burn-up of fuel elements [13–15]. Dennis and Usman implemented a feasibility study for MOX fuel burn-up analysis using of ^{106}Ru [2]. Reilly and co-workers proposed an experimental burn-up determination method using gamma spectroscopy count rate and gamma branching ratio for a burn-up indicator and absolute detector efficiency for the detector used during the experiment [16]. Another burn-up determination was implemented using a simple NDA gamma spectroscopy method using a decay build-up correlation factor based on the irradiation history, fission yield, and specific activity of burn-up indicator [17].

As suggested by Alain and Bignan [18], NDA method can be further improved based on neutron measurements. Würz and co-workers found that it was associated with burn-up and neutron emissions, using combined active and passive neutron measurements [19]. A feasibility study was carried out by Zhao to determine online burn-up credit by taking the neutron count rate for a pebble bed reactor [20]. A combination of gamma and delayed neutron measurement method was implemented to determine fission rate ratios of burnt fuel and fresh fuel element [21]. In addition, an

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experimental delayed thermal neutron counting method was applied to obtain fission rates of different fuel elements having various burn-up values [22].

MSTR (Missouri S&T Reactor) can be used as a tool for feasibility studies of burn-up determination and fuel monitoring using fast delayed neutrons. MSTR is a swimming pool design research reactor operating at 200 kW power since 1961, as shown in Fig. (1). The reactor's core has a movable system that operates in two modes (W: water mode, T: thermal mode). The reactor is in "T" mode when the core grid is close to the thermal column and the reactor in "W" mode when core grid is in the middle of the pool and away from the Graphite walls. The current core fuel configuration of MSTR consists of 15 low enriched uranium (LEU) fuel elements with a mixture of U₃Si₂–Al [23]. A fuel element in the MSTR core consists of 19.75 wt % enriched ²³⁵U and 7.5 wt% silicon. The reactor burn-up and conversion history reported to NRC (Nuclear Regulatory Commission) was presented in the previous study [5].

2. Delay neutrons

Nuclear fission provides many neutron-rich isotopes, which provide delay neutrons. They are essential for nuclear reactor control even though they constitute only 1% of all neutrons from nuclear fission. Brady and England reported [24] that the 271 delayed neutron precursors in their study, and these precursors were grouped based on their similar half-lives. In general, six-groups of delayed neutrons have been used, but eight and twelve groups of delayed neutrons have also been reported for reactor kinetics in the literature [25,26]. Supposing all delayed neutrons are emitted from fission product of ²³⁵U, ²³⁸U, and ²³⁹Pu, the total delayed neutrons can be calculated with Eq. (1) depending on reactor type, while values of A, B, and C vary with fast fission factor, which is discussed in previous work [27].

$$N(t) = (A \cdot \sum_i w_{i-U235} \cdot e^{-\lambda_i t})_{\text{thermal } U}^{235} + (B \cdot \sum_i w_{i-U238} \cdot e^{-\lambda_i t})_{\text{fast } U}^{238} + (C \cdot \sum_i w_{i-Pu239} \cdot e^{-\lambda_i t})_{\text{thermal } Pu}^{239} \tag{1}$$

where t is the time after shutdown, λ_i decay constant, and w_i is the fission yield of i'th precursor. Total fission yields of six-group delayed neutrons are 0.0158, 0.0066, 0.0061, and 0.0412 for ²³⁵U, ²³³U, ²³⁹Pu, and ²³⁸U, respectively [28]. Eq. (1) gives fast fission

factor dependent delay neutrons per fission as a function of time post shutdown. In fact, one would expect that the energy spectrum for each group is not constant in time because each group is made up of a sum of isotopes with different energy spectrum and half-life. The six-group delayed neutrons are for low to medium energy in fact, there is no available information in the literature regarding the energy group of the six-group delayed neutrons. Each group of the delayed neutrons will show different characters since delayed neutrons' energy distribution varies from one group to another. Therefore, delayed neutron counting is not sufficient to investigate spent fuel. The energy-dependent time signature of the delay neutron counts believed to have the information since each precursor's fission yield from Pu and U is different [5].

3. Experimental setup

In order to proceed with the feasibility study for non-destructive burn-up determination using delayed fast neutrons, a liquid scintillator neutron detector (N-Probe) [29] was operated to extract delayed fast neutron spectra from fuel elements at the MSTR beam port in this study. The detector capable of collecting data for both thermal and fast neutrons simultaneously. The detector is also capable of rejecting gamma rays. It can be utilized for data acquisition in nuclear facilities and fuel storage areas [30].

Approximately 6.5 m long beam port presents a link between the reactor core and delayed fast neutron detection system. The beam port outlet dimension in the basement of the reactor is 7.6 cm × 5.1 cm. A 5.8 cm long lead gamma-ray shielding was placed in the beam port to stop unwanted gamma radiation [31]. It is assumed that all the delayed fast neutrons recorded by the detector come from the fuel element (F11 in Fig. 2) directly in front of the beam port entrance. Due to the neutron scattering and the loss of neutron energy, the ratio between the number of neutrons entering the beam port and the number of neutrons exiting is in the order of 10⁴ [31].

The delayed fast neutrons' data were collected for 30 min immediately after shutdown at two different power levels for the four separate fuel elements at MSTR. It should be emphasized that these data were collected after 15 min of reactor operation since neutron precursors would reach their steady-state concentrations during this time for each fuel element. Four different fuel elements were used to collect delayed fast neutrons in "T" mode. Fuel elements F1 and F2 were used (burnt fuels) in MSTR core since 1992, while F11 and F16 fuel elements were mostly stored in the tank next to the reactor pool since 1992 (with little to no burn-up).

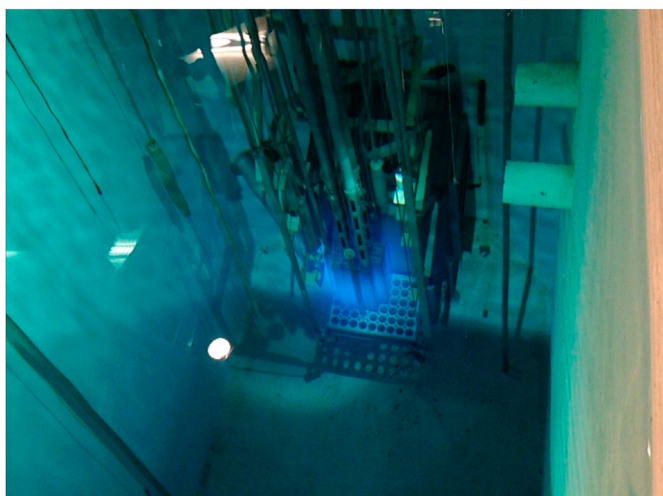


Fig. 1. A picture of MSTR core.

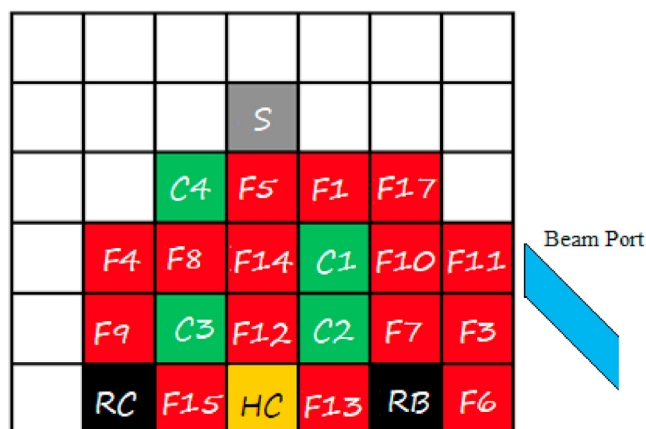


Fig. 2. Current core configuration of MSTR and beam port location (numbers with F represent fuel elements).

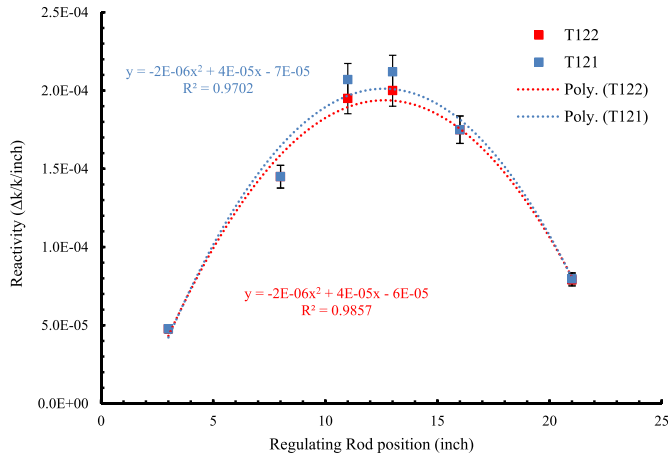


Fig. 3. Regulating rod worth with respect to regulating rod height for T121 and T122 core configuration.

Although F11 fuel element was barely used in the reactor core in 2007 for two months, both fuel elements were considered as fresh fuels in our study. After collecting the delayed fast neutron data of the F11 fuel element in the configuration, as shown Fig. (2), this fuel element was removed from the grid and placed in a storage tank. Then, other fuel elements (F1, F2, F16) were inserted respectively in the grid where the F11 element was located, and their data were taken.

4. Results

In this study, the feasibility of determining burn-up and the Pu–U content for spent nuclear fuel element non-destructively using delayed fast neutrons was examined. The data of delayed fast neutrons were collected for 30 min post shutdown with two fresh (F16, F11) and two burnt (F2, F1) fuel elements. The core configuration of the reactor is named for each fuel change or shuffling. For each core configuration, all safety and calibration analysis were completed as required by NRC (Nuclear Regulatory Commission). Four different reactor core configurations (T119, T120, T121, and T122) were used for burn-up studies using delayed fast neutron data. The reactor core configurations T119, T120, T121 and T122 were when F2, F11, F16, or F1 fuel element respectively was placed in D9 fuel grid without perturbing other fuel elements. Grid location D9 is right in front of the beam port entrance, as shown in Fig. (2).

Figs. (3) and (4) show the regulating rod worth values and total reactivities of T121 and T122 configurations with respect to rod height, respectively. During the experiments, the mid-section of the fuel was providing most of the delayed neutrons for counting. Any vertical movement of the fuel element would have increased the uncertainty in the data and hence no fuel elements were moved during the measurements.

As can be seen from Fig. (3), when a new fuel element is placed in the reactor core, the reactivity in the core increases. The low reactivity in T122 configuration is due to the higher total burn-up of fuel elements. The regulating rod worth was $2.07E-04 \Delta k/k/in$, and $1.95E-04 \Delta k/k/in$ for T121 and T122 core configurations at 11 inch, respectively. A standard deviation error bar (1σ) were included to Figs. (3) and (4) for statistical uncertainties.

Delayed fast neutrons produced different peak heights depending on the reactor powers. Therefore, two separate delayed neutron data sets were taken at low power (10 kW) and high power (100 kW). Figs. (5) and (6) represent the time-integrated energy

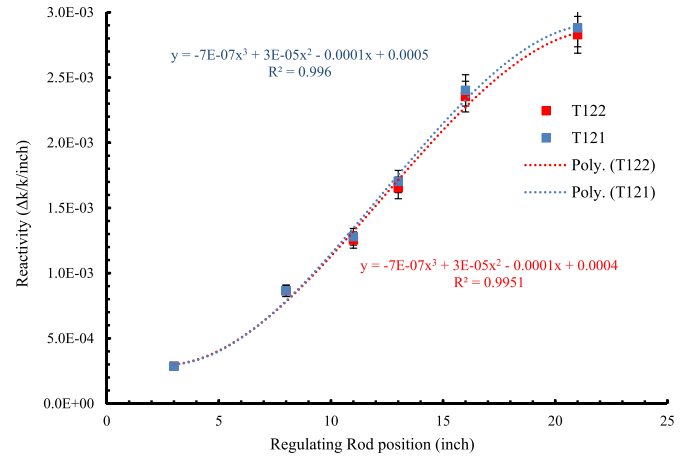


Fig. 4. Total reactivity with respect to regulating rod height for T121 and T122 core configuration.

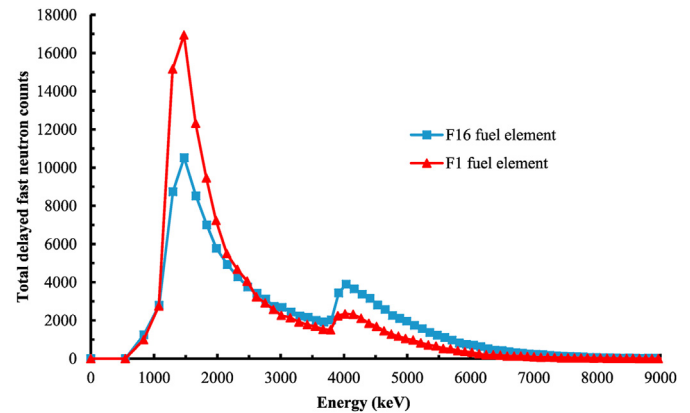


Fig. 5. Delayed fast neutron data for burnt F1 and fresh F16 fuel elements at 10 kW reactor power¹ [5].

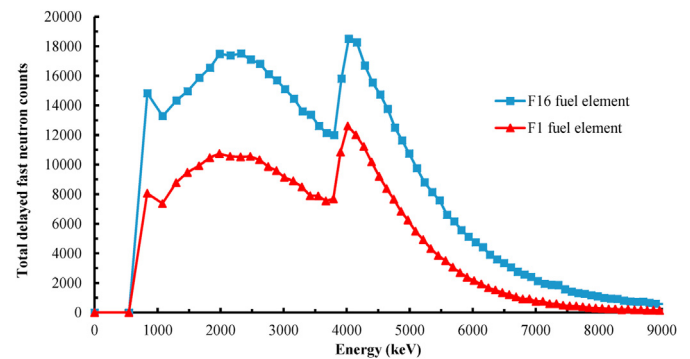


Fig. 6. Delayed fast neutron data for burnt F1 and fresh F16 fuel elements at 100 kW reactor power² [5].

spectra of burnt F1 fuel element and fresh F16 fuel element for 10 kW and 100 kW power runs, respectively. Akyurek and Usman presented these figures in their previous study [5], they are

included here with permission from the publisher. In this study, we investigated in detail time dependent fast neutron spectrum and the precursor population behavior. There was a very pronounced low energy peak (LEP) for the case of low reactor power (10 kW). It can be seen that the peak of the burnt fuel element is dominant compared to the fresh fuel. However, it was observed that the high-energy peak (HEP) of the burnt fuel element at the same reactor power was lower than that of fresh fuel. It is important to note that our experiments showed that the burnt fuel element's LEP is not dominant after irradiation with 50 kW reactor power. Therefore, the LEP is not the proper inspection peak for burn-up feasibility studies due to the power dependence unless an external neutron source is used as a probe simulating low power burnup. A similar situation was observed for burnt F2 and fresh F11 fuel elements [5].

The delayed fast neutron data for the burnt and the fresh fuel elements were investigated for 100 kW power. Although the low energy peak at low power is higher, the opposite behavior is observed at high power level. In addition to first and second peaks, a wide bumpy peak (BP) was formed between these peaks at higher powers. A similar situation was also observed for burnt F2 and fresh F11 fuel elements with 100 kW power [5]. Peak variability at low energy levels indicates the reactor power dependence of precursor production. Besides, fast delayed neutrons obtained at low power level decays faster than those obtained at higher power level. This eventually produces unwanted faulty statistics. Therefore, low-level powers are unreliable for burn-up analysis and Pu conversion calculations using delayed fast neutrons. As shown in Fig. (6), the delayed fast neutron emission rate of the fresh fuel is much higher than the delayed fast neutron emission rate of the burnt fuel. This shows the burn-up history dependence.

The time-integrated spectrum does not provide the full picture of the emission. The low energy peak (LEP) is dominant for times immediately post-shutdown, while the high energy peak (HEP) is dominant for a long time after shutdown. Fig. (7) shows two different count distributions of the F16 fresh fuel element, and F1 burnt fuel element range from 0 to 450 s and from 460 to 900 s at 100 kW power. Interestingly, high energy peaks around 4000 keV emerged around 200 s after the reactor's shutdown and dominated after 450 s. This specifies that the high energy peaks were produced by the precursors with longer half-lives. On the other hand, it was observed that the peaks with low energy level started to disappear after 450 s. The time dependence of the peaks indicated that the source of these neutrons/precursors have distinctly different half-lives. Fig. (8) shows that the low energy peak is dominant at the beginning of delayed fast neutron measurement, and high energy peak is dominant after 10 min for F11 fresh fuel element. This is also valid for all other fuel elements tested at 100 kW power.

At higher reactor powers, an intermediate energy bumpy peak (BP) appears in the middle of the spectrum for each fuel element. This bump-shape peak emerged 50 s after delayed fast neutron count started, as shown in Fig. (9), but they reached a steady-state after 300 s. Small shifts between the peaks in the spectrum are likely the result of detector calibration differences. The emission rate of the new F16 fuel, which was never used before in the core, is also higher than the other fuels in bumpy region. F11 fuel comes right after F16 fuel which was used for a short period of time in the reactor, as mentioned earlier. This bumpy region also reveals burn-up dependence. As it can be seen from Fig. (9), delayed fast neutrons in BP and HEP emerged much later after reactor shutdown,

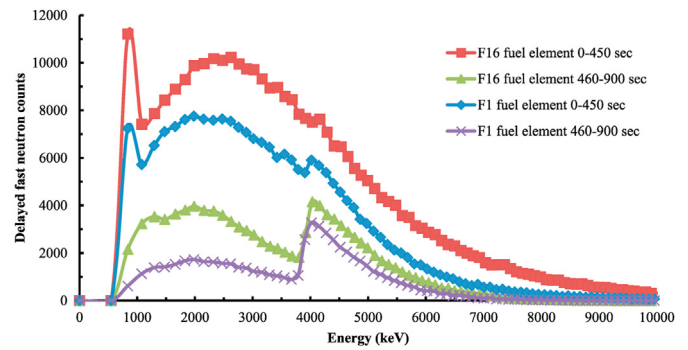


Fig. 7. The count distributions of F1 and F16 fuel elements at different time intervals at 100 kW power.

while delayed neutrons in LEP was increasing. Therefore, delayed fast neutrons for BP and HEP come from long-lived neutron precursors such as ⁸⁷Br (half-life: 55.6 s).

Fig. (10) shows the temporal response of BPs (middle peak) for fresh F16 fuel and F1 burnt fuel elements after 100 kW power shutdown. Similar results were observed for fresh F11 and burnt F2 fuel elements. It is intriguing to see that initial delayed fast neutron precursor build-up was observed for both fuel elements for about 20 s. It was also noticed at low and high-energy peaks and reported in the previous study [5]. Similar behavior was observed by Jordan and Perret using delayed thermal neutrons; they stated that this

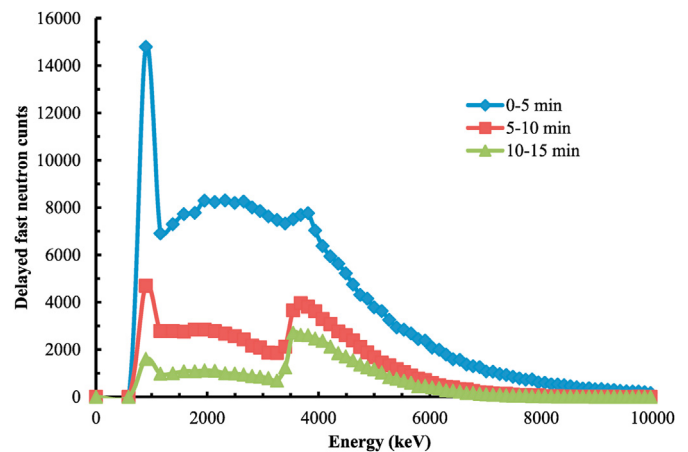


Fig. 8. Delayed fast neutron count distribution of F11 fresh fuel element at different time intervals at 100 kW power.

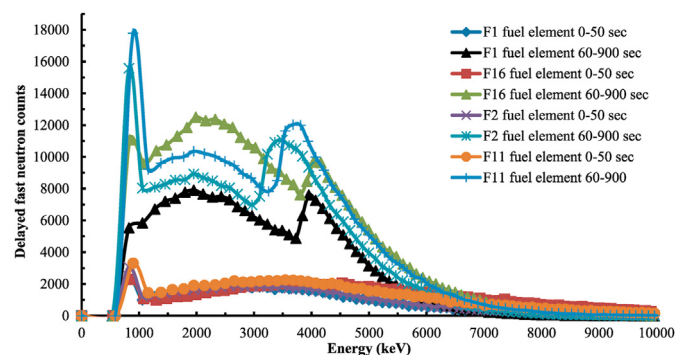


Fig. 9. The count distribution of different fuel elements at different time intervals at 100 kW power.

¹ Fig. 5 was already published in earlier work and permissions granted by publisher and authors to present here.

² Fig. 6 was already published in earlier work and permissions granted by publisher and authors to present here.

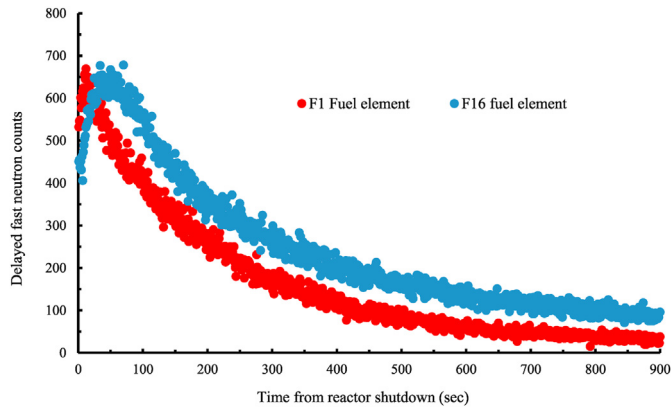


Fig. 10. Delayed fast neutron count rates burnt (F1) and fresh (F16) fuel elements fuel for bump-shape peak after 100 kW shutdown.

precursor build-up originated from fuel transportation [22]. The figure indicates for both fuel elements that there is a strong relationship between delayed fast neutron emission of BP and fuel burn-up.

Even though all investigated fuel elements were inserted into the same core grid and exposed to the same irradiation time for each power run (precursors population depends on both the duration of run and power during those runs), we decided to normalize the peaks to eliminate any dependence that may occur. Figs. (11) and (12) show the normalized LEP/HEP and BP/HEP ratios as a function of time for F16 and F1 fuel elements, respectively. A 10 s sum for the peak taking ratios obtained to remove statistical errors for all steps. As it can be seen from both figures, BPs and HEPs are suitable peaks for burn-up analysis because both peak heights show decisive behavior for burnt and fresh fuel elements, even though they do not appear in the first 50 s after reactor shutdown. BPs and HEPs of fresh fuel elements are always higher than burnt fuel elements, but it should be highlighted that BPs are formed only after 50 kW reactor power. As discussed earlier, LEP for burnt and fresh fuel elements vary depending on the reactor power, hence these peaks should not be relied on for burn-up analysis. Similar results were observed for fresh F11 and burnt F2 fuel elements. These results are consistent with earlier work [32–34], in that the energy spectrum of delayed neutron strongly depend on the fissile material from which neutron originated.

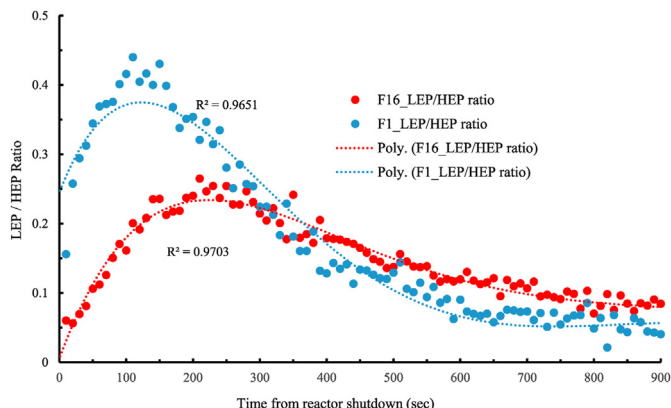


Fig. 11. LEP and HEP with respect to time for F16 and F1 fuel elements at 100 kW.

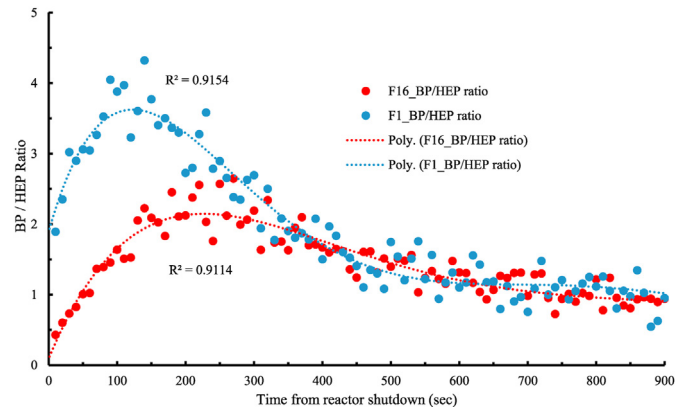


Fig. 12. BP and HEP ratios with respect to time for F16 and F1 fuel elements at 100 kW.

5. Conclusion

An experimental burn-up feasibility study was performed using delayed fast neutrons for MSTR fuel elements and delayed fast neutron data for burnt and fresh fuel elements collected using the non-destructive fuel investigation method at MSTR. It has been observed that delayed fast neutrons are suitable for burn-up analysis and fuel monitoring. Spent fuel investigation and monitoring using delayed fast neutrons would be more effective for MOX fuels than UO₂ because there is almost a 3-fold fraction yield (β_i) difference between ²³⁵U and ²³⁹Pu. Therefore, fuel investigations using delayed fast neutrons would also be a sophisticated tool for distinguishing plutonium and uranium. BPs and HEPs are more prominent for burn-up analysis and monitoring since LEPs depends on power level. Although this study shows a strong relation between burn-up and delayed fast neutron counts, it requires a detailed analysis technique to determine which delay neutron precursors contribute to a specific energy range. As a result, nuclear fuel investigations using delayed fast neutrons are thought to be an alternative non-destructive method. It is important to note that the delayed neutron spectrum can be obtained in a limited time since the delayed neutron data of nuclear fuels can only be obtained when the reactor is completely shut down.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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