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Neutron Detection Monitoring Outside of Containment of a Power Reactor during Reactor Restart and Establishment of an Equilibrium Core

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Abstract

It is currently a priority objective of the Department of Safeguards within the International Atomic Energy Agency (IAEA) to "strengthen instrumentation capabilities for verification", with the specific research and development need to "develop safeguards equipment to establish and maintain knowledge of spent fuel in shielding/storage/transport containers at all points in their life cycle" [1]. It has been demonstrated that large-area (~0.2 m^2) neutron detectors can monitor reactor fuel cycles at stand-off distances of up to 100 m from a research reactor by measurement of the extremely low neutron efflux, which remains measurable even though significant shielding and infrastructure reduce count rates to near background level. In particular, even at very low measurement levels, the neutron detection rate per unit reactor power has been demonstrated to correlate with changes in fissile isotopic composition of the reactor core [2]. More recently, it has also been demonstrated that the reactor neutron efflux can be measured outside of containment at a power reactor, to the extent of detecting the movement of spent nuclear fuel during online re-fuelling operations [3]. The recent refurbishment and restart of the Unit 2 reactor core (935/881 MWe gross/net) at Darlington Nuclear Generating Station (DNGS) in Ontario, Canada, presented a unique opportunity to monitor, with neutron detectors positioned outside of containment, the changing fissile isotopic composition of a CANDU reactor core as an equilibrium burnup distribution was established some months after restart of the unit-. The results of a nine month measurement campaign at DNGS will be presented.

Introduction

Nuclear reactor safeguards measures are used to verify that nuclear material is not diverted from peaceful uses [4]. It is currently a priority objective of the Department of Safeguards within the International Atomic Energy Agency (IAEA) to "strengthen instrumentation capabilities for verification", with the specific research and development need to "develop safeguards equipment to establish and maintain knowledge of spent fuel in shielding/storage/transport containers at all points in their life cycle" [1]. This includes increasing capability to detect undeclared nuclear material in reactor facilities, which can be accomplished by detecting process emanations from nuclear fuel cycle activities, using newly developed instruments and associated techniques [1].

A prominent emanation from nuclear reactors is neutrons. It has been demonstrated how an array of large-area neutron detectors at various locations outside of reactor shielding (up to 100 m away) could be used as an economical and non-invasive means of tracking the fissile inventory of a reactor core, as well as the movement of nuclear fuel during online re-fuelling [2]. This demonstration was carried out at the National Research Universal (NRU) research reactor in Chalk River, Ontario, Canada. Further experience was gained in monitoring the movement of legacy spent nuclear fuel at

the Fuel Packaging and Storage facility in Chalk River, using the same neutron detection technology at similar stand-off distances, even while the spent fuel is kept inside heavy shielding during movement [3].

The demonstration of reactor monitoring at NRU was at a reactor facility with no containment structure. Most reactor facilities, particularly those at power generation facilities, include additional containment structure that on passing inspection would impede the detection of reactor-sourced neutrons outside of containment. A neutron measurement campaign carried out at Point Lepreau Nuclear Generating Station in Point Lepreau, New Brunswick, Canada demonstrated that it is possible to detect reactor-sourced neutrons outside of containment at a power reactor, with some evidence of re-fuelling operations being detected in changes in average neutron count rate during extended 24-hour measurements [5].

The CANada Deuterium Uranium (CANDU) reactor with its online re-fuelling capability maintains during regular operation an equilibrium core, in which the fissile isotopic content of the core remains relatively constant. The start-up of a CANDU reactor from a fresh uranium core with no Pu present gives a unique opportunity to observe significant changes in the fissile isotopic inventory of the core as the reactor core approaches an equilibrium configuration. Such an opportunity was presented during the recent restart of a newly refurbished Unit 2 reactor at the Darlington Nuclear Generating Station in Ontario, Canada. This paper reports on results acquired in an extended neutron measurement campaign outside containment of the Unit 2 reactor that commenced prior to restart, and followed the progress of Unit 2 towards an equilibrium core.

Methods

Measurements were conducted at Unit 2 of Darlington Nuclear Power Generating Station in Clarington, Ontario, Canada. This is a commercial pressurized heavy water reactor producing 2776 MW thermal power, and 881 MWe net power, and has been in service since 1990 [6,7]. Unit 2 was disconnected from the power grid in 2016 October for refurbishment. After successful completion of refurbishment, Unit 2 went critical on 2020 April 9, and returned to service on the power grid by 2020 June 4 [8]. Measurements were conducted at two locations outside of Unit 2 containment, one near a containment airlock door (about 40 m from the Unit 2 core), and the other facing a primary heat transport pump (PHTP) motor room (about 30 m from the Unit 2 core). Large area neutron detectors were deployed at these locations in 2020 March, some weeks prior to Unit 2 going critical.

The large-area neutron detector facing the PHTP motor room consisted of seven sealed “B10+” stainless steel tubes (2.54 cm diameter, 101.6 cm active length) from General Electric Reuter Stokes (Twinsburg, Ohio, USA) that were lined with an elemental ^{10}B-enriched coating, and filled with 0.75 atm ^{3}He [9]. The detector was encased in a high density polyethylene box for moderation of predominantly fast neutrons streaming from the PHTP motor room. The large area neutron detector positioned near the airlock door consisted of six sealed ^{3}He stainless steel tubes (2.54 cm outer diameter, 101.6 cm active length) also from General Electric Reuter Stokes, that were filled with 4 atm ^{3}He. At the location near the airlock door, the incident neutron spectrum was considerably softer, such that it was not necessary to surround the detector by a moderator box. In the case of each of these two detectors, high voltage was supplied by an NPM3100E neutron pulse monitor (NPM) from Quaesta Instruments (Tucson, Arizona, USA), which also processed pulses through a

charge-sensitive amplifier, a fixed-gain pulse-shaping amplifier, a variable gain amplifier, and an analog-to-digital converter, before using firmware algorithms to analyze the digitized data. The NPM was used to record pulses in data-logging mode. Pictures of the detectors at their respective locations are shown in Figure 1.

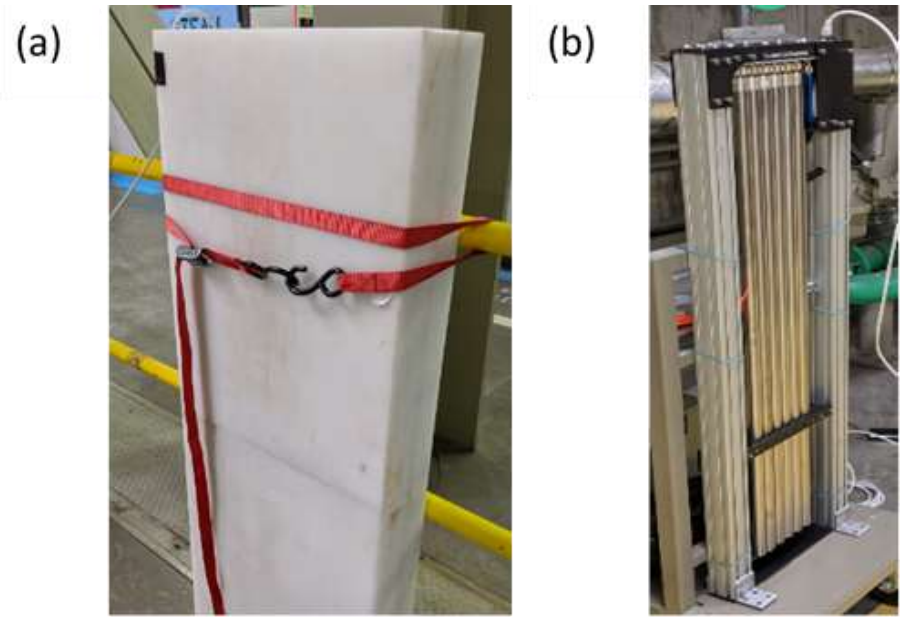

Figure 1. Photos of large area detectors positioned (a) near the PHTP pump motor room, and (b) near the containment airlock door.

Results

Measurements were conducted over a nine-month span of time, from 2020 March 11 to 2020 December 11. Figure 2 shows a survey of the data taken by the two detectors. The clocks of the detectors were synchronized. Between the time period of Unit 2 going critical on 2020 April 9, and Unit 2 going into service at full power on 2020 June 4, the reactor unit underwent a period of testing, which is clearly seen by both detectors. Upon return to service at full power, the detector facing the PHTP pump motor room recorded neutrons at the rate of 76 counts per second (cps), and the detector near the containment airlock door recorded neutrons at the rate of 9 cps. Both detectors also observed a ~50% dip in count rate on 2020 Nov 10, corresponding to a temporary ~50% drop in reactor power. It should be emphasized that although statistically significant neutron count rates were measured outside of containment in the above surveys, this is only due to the high sensitivity of the large-area ^{3}He neutron detector tubes employed. The sensitivity of the neutron detectors employed are such that the ambient dose rate equivalent $H^*(10)$ is ~10 nSv/hr for these count rates [10]. Consequently, the dose consequences of the measured neutron fields are negligibly small.

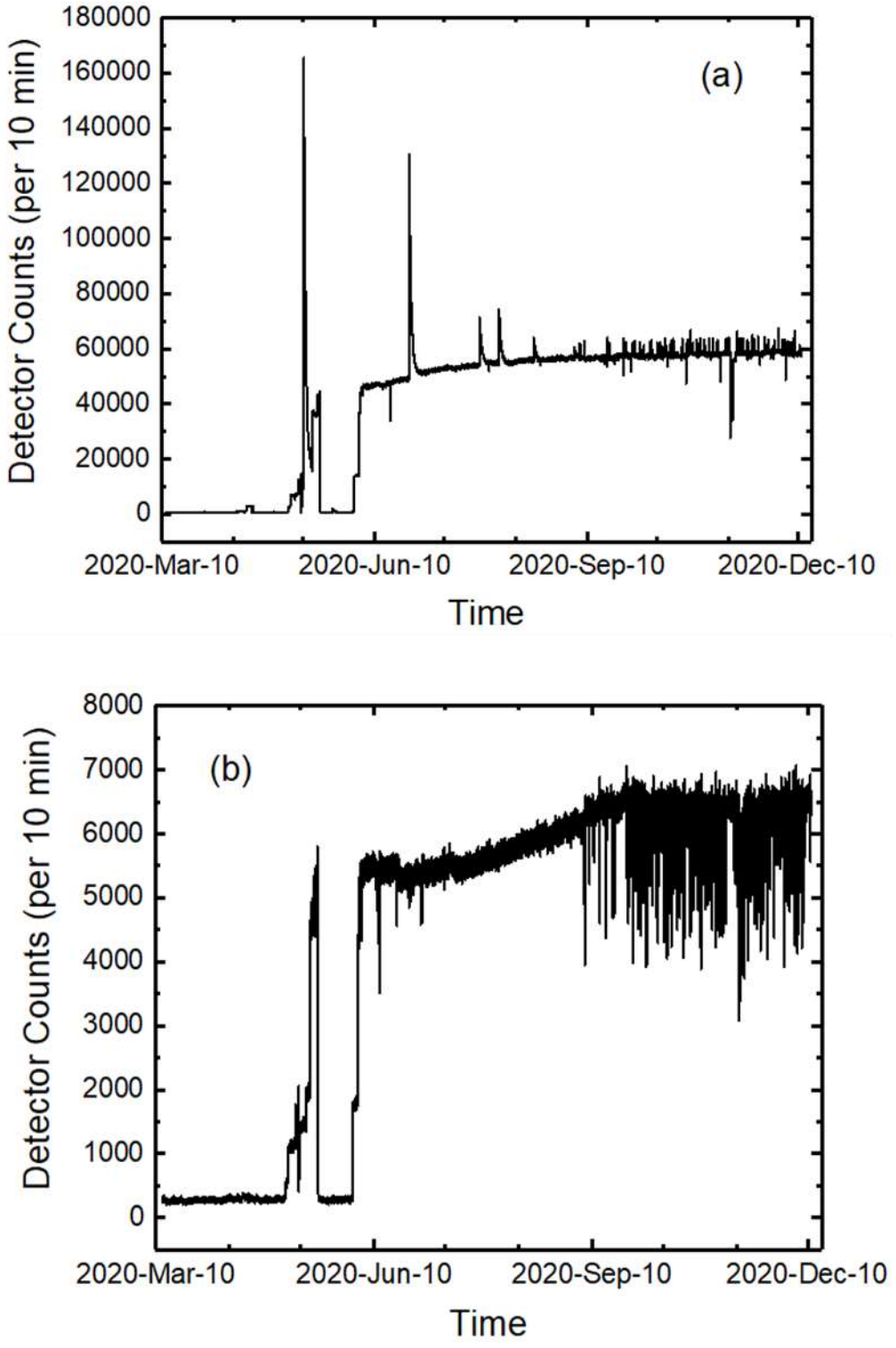

Figure 2. Survey of data taken by the detectors outside of Unit 2 containment (a) facing the PHTP motor room, and (b) near the containment airlock door.

Some distinct differences can be seen between the two plots. The data recorded facing the PHTP motor room features prominent spikes in neutron counts that are absent in data recorded near the containment airlock door. Neutron emission seen from the PHTP system is dominated by neutrons released by neutron activation or photo-induction in the coolant circulating through the PHTP. For a given coolant flow rate through the PHTP, the neutron detection rate is proportional to the neutron flux in the reactor core. However, variations in the flow rate through the PHTP can give rise to the spikes observed in Figure 2(a) during otherwise normal full power operation. The data shown in Figure 2(b) shows regular drops in count rate roughly every two days from 2020 September 6 to 24; the frequency then increases to daily thereafter. The containment airlock door is on level with a reactor end face; the regular drops in count rate likely coincide with fuelling machines temporarily blocking neutron shine from reaching the detector during re-fuelling operations, although this is not confirmed by operational records.

In Figure 2, the most important feature that the detectors have in common is the gradual rise in count rate during full-power operation of the Unit 2 reactor. Both detectors show a monotonic rise in count rate that saturated after approximately three months of data acquisition from the time the reactor returned to service at constant full power. It is notable how Figure 2(a) shows a smoother rise, with greatest slope at earliest times after return to service, while the rise in Figure 2(b) is not as smooth, and may be affected by moderator poison during the approach to an equilibrium core. Typically, a CANDU reactor will use boron as a moderator poison to compensate for excess reactivity in the initial core, when all the fuel in the core is fresh [11]. Gadolinium is often used as moderator poison for reactivity control during the approach to criticality upon start-up [12]. In the case of Darlington's Unit 2, gadolinium was in fact used in the approach to criticality before gradually switching over to boron above 1% full power [13]. As the moderator poison is applied to the moderator within the calandria of the reactor, the signal of the neutron detector deployed near the airlock door would influenced by variations in moderator poison concentration. The moderator poison does not populate the coolant flowing through fuel channels, however, such that the signal of the detector near the PHTP motor would not be sensitive to variations in moderator poison concentration.

Figure 3 shows relative detector count rate, normalized to the count rate measured when Unit 2 returned to service on 2020 June 4. By 2020 September 11, the average count rate had increased by 24% in Figure 3(a), and 19% in Figure 3(b).

Discussion

Following Ref. [14], one can assume a simplified model of a thermal reactor where the neutron flux ϕ in the reactor core is an appropriate space-averaged and energy-averaged value, and the fission cross section for the i^{th} fissioning species is a corresponding average cross section $\sigma_{f,i}$. Here, averaged flux and cross section are used on the grounds that most of the fissions in a thermal reactor (such as a pressurized heavy water reactor [15]) occur in the thermal energy region, where both neutron flux and fission cross section are large. Based on these assumptions, one can arrive at the following expression,

$$\frac{\langle \phi \rangle}{P_{\text{tot}}} = \left[N_A \sum_i \frac{m_i \langle \sigma_{f,i} \rangle E_{f,i}}{w_i} \right]^{-1} \quad (1)$$

where P_{tot} is the total thermal reactor power, N_A is Avogadro's number, and for the i^{th} fissioning isotope species, m_i is the mass, $E_{f,i}$ is the energy released per fission, and w_i is the atomic weight. The neutron detector count rate per unit reactor power is assumed to be proportional to $\langle\phi\rangle/P_{tot}$. The above relationship can be used to quantitatively connect the change in neutron count rate per unit thermal power to the change in fissile content. In particular, as the fuel inventory of a reactor core changes over time, this results in corresponding changes in $\langle\phi\rangle/P_{tot}$ over time, and thus the corresponding neutron detector count rate for fixed reactor power. It is proposed that this accounts for the gradual increase in neutron count rate over time observed over full reactor power operation in Figure 2 and Figure 3.

As a test of this proposal, consider the isotopic composition of the core at the start of full power operation, compared to a near-equilibrium core. Zaysin et al. [16] provide the atom density of important isotopes as a function of burn-up for natural UO_2 fuel in a CANDU reactor, some values of which are provided below in Table 1.

Table 1. Atom density of important isotopes for a natural UO_2 CANDU core at 0 MWd/TU and 2500 MWd/TU [16], together with energy released per fission E_f [17,18], and average thermal neutron-induced fission cross section σ_f [19].

Isotope	U-235	Pu-239	Pu-240	Pu-241
0 MWd/TU (b^{-1} cm^{-1})	$1.8 \times 10^{--4}$	-	-	-
2500 MWd/TU (b^{-1} cm^{-1})	1.0×10^{-4}	3.5×10^{-5}	5.0×10^{-6}	1.5×10^{-6}
E_f (MeV)	201.7	210.0	210.4	212.4
σ_f (barns)	582.6	748.1	0.056	1011.1

The thermal power of Darlington Unit 2 is 2776 MW [7], with 110 MT of U present in a fresh core. Based on this, approximately 2500 MWd/MTU burnup is achieved after 99 full-power days, which spans from 2020 June 4 (the commencement of full power operation) to September 11 (near to an equilibrium core). Using the information provided in Table 1, one can estimate the relative change in $\langle\phi\rangle/P_{tot}$ after this extent of burnup, comparing between the reactor core on September 11 and the reactor core on June 4. Following such a method, a 19% increase in neutron flux over time for fixed power P_{tot} is predicted. This is a core-average calculation. Regions of the core at lower power, such as the ends of fuel channel ends, will have lower burnup, and lower Pu buildup. The above simple core-average calculation may therefore underestimate the Pu fission fraction. This underestimate is nevertheless diminished by the fact that the Pu fission fraction increases more slowly at higher burnup. Overall, the power-weighted calculation may be closer to 24% (observed in Figure 3(a)) than 19% (observed in Figure 3(b)).

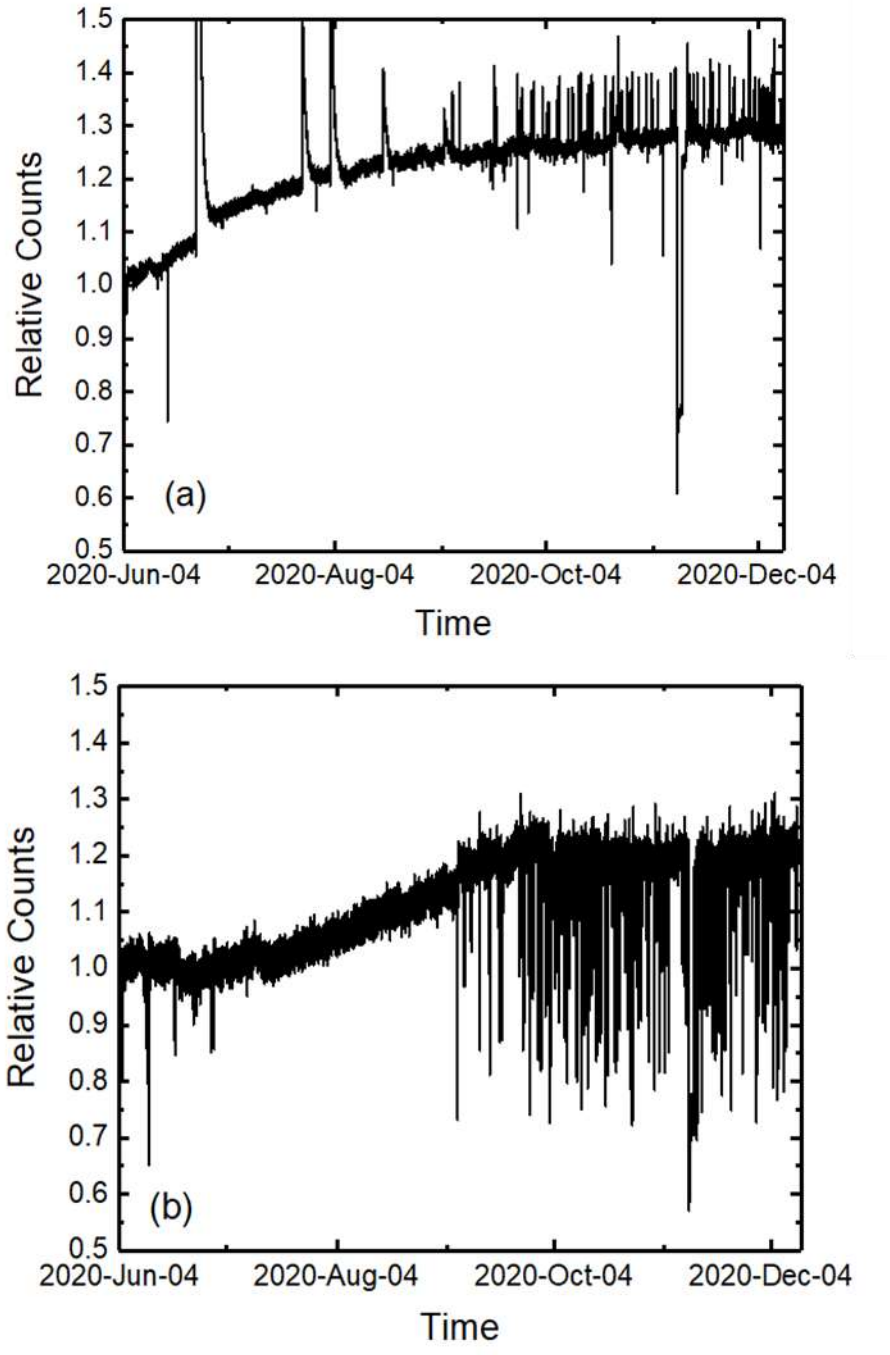

Figure 3. Relative neutron count rate vs. time recorded (a) facing the PHTP motor room, and (b) near the containment airlock door.

Conclusions

The re-start of a CANDU reactor from a fresh fueled core after refurbishment is a unique and rare opportunity to observe substantial changes in isotopic inventory in a reactor that otherwise maintains an equilibrium core with regular online re-fuelling. The measurement campaign

conducted at Darlington's Unit 2 reactor during re-start and return to full-power service demonstrated clear and measurable changes in the neutron count rate measured at two locations outside of reactor containment, during operation at full reactor power. A simple model previously demonstrated to account for relative changes in isotopic composition in the NRU research reactor using neutron detector measurements taken outside of reactor shielding, is also shown here to account for relative changes in isotopic composition of a CANDU reactor with neutron detector measurements taken outside of reactor containment. This shows that power reactor monitoring with neutron detection, even outside of power reactor containment, is a potential tool for providing continuity of knowledge in the safeguarding of nuclear material.

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