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VeriFicatton of CANDU Spent Fuel in Sealed Storage Casks

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ABSTRACT
The development of cost-effective methods for long-term safeguarding of spent reactor fuel in temporary storage is necessary in order for the IAEA to meet all demands for safeguards resources. To this end, we have developed a measurement system to verify spent CANDU type fuel in sealed long-term storage casks. Given the large number of spent-fuel storage casks to be monitored, we have attempted to minimize the cost of the proposed system while still providing a useful detection signature from the entire storage cask. The detection system described consists of three small $^3$He neutron counters placed inside the storage cask in IAEA reverification tubes. The limiting factor on the size of each detector is a bend in the reverification tube around which the detector must pass. Each detector will be shielded with lead to reduce the gamma dose rate below 30 R/hr while still providing a significant neutron detection signal. Specially designed electronics will be designed at LANL to process the signals from the $^3$He neutron counters and store data. The electronics will be located outside the cask to minimize gamma dose, maximize long-term stability, and provide for ease of maintenance. Monte Carlo calculations of neutron transport through a CANDU spent fuel storage cask suggest that a single string of neutron detectors will provide visibility of better than 90% of the cask contents at a 5% detection level.

INTRODUCTION
The International Atomic Energy Agency (IAEA) has considerable experience in safeguarding spent nuclear fuel. Several techniques have been employed to monitor spent fuel in reactor cooling ponds as well as the transfer of spent fuel from these ponds to reprocessing plants or other storage locations, such as dry storage casks. However, an important aspect of the IAEA safeguards technology that needs further development is cost-effective monitoring of spent fuel in long-term storage containers.

Because of the on-line loading and the large number of spent fuel bundles, CANDU-type reactors require a significant safeguards effort for the IAEA. As the spent-fuel storage pools reach capacity, it is necessary to transfer the spent-fuel bundles to dry storage canisters for long-term storage. The IAEA requires an efficient method to monitor the spent fuel in the concrete canisters with minimal intrusiveness to the facility operator. This study evaluates sensors for potential monitoring of CANDU-type spent fuel contained in concrete canisters.

In light of the large number of CANDU-type spent-fuel storage facilities around the world, a cost-effective means for safeguarding material in long-term storage is desirable. The ability to safeguard the cask contents is a necessary requirement for any proposed system. Safeguards criteria appropriate to spent fuel in large canisters currently do not exist. However, guidance has been developed (see report of the Advisory Group Meeting on Safeguards for Final Disposal of Spent Fuel in Geologic Repositories, IAEA STR-309, 1-5 December 1997) indicating that the appropriate criteria for verification would be detection of a number of pins equivalent to 0.5 SQ. Passive measurements based on neutron and gamma detection have been proposed as verification methods for the detection of spent-fuel diversions from storage casks.

The proposed detection system will be housed within the storage cask in specially-constructed "IAEA reverification tubes" inside the concrete structure of the cask. The system should provide a continuity of knowledge (COK) signature of the spent fuel within the cask. Ultimately, the system will be used in conjunction with surveillance cameras and IAEA seals to verify that the contents of a given cask have not
changed over time. Once an initial measurement of the neutron or gamma-ray count rate has been made on a given cask, accounting for the natural decay of uranium, plutonium, and fission products, the system will report changes in this initial measurement over time. In the case of loss of COK of the monitoring system, it will be necessary to re-establish the spent-fuel inventory. For the case where the spent fuel has been previously transferred to the storage cask prior to the implementation of the monitoring system, it will be necessary to establish a baseline spent fuel radiation signature.

In this paper, we show that a passive neutron detection system provides for better visibility of the cask contents than a passive gamma-ray detection system. This is largely because of attenuation and absorption effects of gamma rays in the high-density, high-Z reactor fuel. Neutrons, on the other hand, have a greater penetrability through this environment. Thus, it is more likely that neutrons from the center or far side of the cask (opposite the detectors) will be counted in the reverification tubes. We propose that a passive neutron detection system consisting of three small 4-atm $^3$He neutron counters, in positions near the bottom, middle, and top of the cask, will provide the necessary detection sensitivity to meet IAEA safeguards requirements while maintaining a reasonable system cost and simplicity.

**CANDU SPENT-FUEL STORAGE CASKS**

The heavy water used to moderate a CANDU reactor permits the use of natural uranium in the reactor core. The reactor fuel pins are grouped into bundles, with each bundle containing approximately 21.3 kg of UO$_2$. Thus each bundle of spent fuel in long-term storage is predominantly natural UO$_2$ with a small amount of Pu (~70 g).

For the long-term storage of CANDU spent fuel, the bundles are loaded into stainless steel storage baskets, typically 60 bundles per basket, in the array shown in Fig. 1. After loading, the baskets are welded closed, transported to the storage yard, and placed into a shielded cask. The spent fuel baskets are stacked 9 deep into a concrete storage cask, shown in Fig. 2. The total height of the cask is approximately 6.4 m while the diameter is about 3.1 m. A thin stainless steel liner rings the inner diameter of the concrete. Two reverification tubes are placed on opposite sides of the cask. There is a bend in each reverification tube (not shown in Fig. 2) around which the detectors must pass. In order to traverse the bend, the maximum size of any detector package is 7.5 in. (l) x 1.5 in. (dia.). A full storage cask contains 540 bundles of spent fuel (9 baskets with 60 bundles each). Therefore, there are approximately 10.14 metric tons (1 metric ton = $10^3$ kg) of uranium and about 4.7 SQ of plutonium in a single storage cask.

**CALCULATIONS**

In order to compare the responses for neutron and gamma-ray detection systems in the reverification tubes, we must use as a guide an expected production rate for neutron and gamma rays from the cask. Of course, each cask will be composed of spent fuel with different burnups and cooling times. Therefore, we have calculated the expected emission rates for natural enrichment CANDU fuel with a variety of burnups and cooling times. However, to simply the argument, we limit the discussion here to the case of a cask uniformly distributed with spent fuel that has a burnup level of 7500 MWD/MTIHM (megawatt days per metric ton of initial heavy metal) and a 10 year cooling period. The abundances of the neutron and gamma-emitting isotopes were calculated by the computer code ORIGEN2 [1], which calculates the buildup and decay of isotopes in the fuel.
The dominant sources of neutrons are from the spontaneous fission of $^{244}$Cm and $^{246}$Pu, and from $(\alpha,n)$ reactions in the fuel, with minor contributions from the spontaneous fission of $^{242}$Pu, $^{238}$Pu, $^{238}$U, and $^{242}$Cm. Summing up all these contributions, the neutron production rate is $2.88 \times 10^6$ n/s per MTIHM. Gamma rays from the spent fuel come from activation products, actinides, and fission products present in the fuel. However, most of the gamma rays come from the long-lived fission products with nearly half coming from $^{137}$Cs. The integrated gamma ray intensity is $1.68 \times 10^{15}$ ph/s per MTIHM. Therefore, for a full cask, the expected emission rates would be $2.92 \times 10^7$ n/s and $1.70 \times 10^{16}$ ph/s.

Calculations of neutron and gamma-ray transport through the cask were performed with the 3D continuous-energy Monte Carlo code MCNP [2]. A detailed three-dimensional model consisting of 540 fuel bundles in 9 stainless steel fuel baskets, the cylindrical concrete structure of the storage cask, and three detectors placed in a reverification tube was constructed to determine the expected signal rates. Three small 4-atm $^3$He neutron counters were placed in one of the reverification tubes vertically near the bottom, middle, and top of the cask. As viewed in Fig. 2, we label the baskets, from bottom to top, as Basket 1 through Basket 9. The detectors, labeled Bottom, Middle, and Top, were placed vertically near Basket 2, 5, and 8, respectively.

In calculations involving neutrons, the performance of the detection system was recorded in an expected count rate (counts/s) for $^3$He tubes, whereas in calculations involving gamma rays, the performance is recorded as an expected dose rate (R/hr) on a detector (similar in size and shape to the $^3$He tube) at the same location in space as the neutron detector.
Assuming a fully loaded cask of spent fuel with a burnup of 7500 MWd/MTIHM and a 10-year cooling time, the MCNP calculations suggest that each of the three neutron detectors (placed vertically near Baskets 2, 5, and 8) would measure a count rate of ~1750 count/s. Gamma-ray calculations show that the expected dose rate on detectors at these locations is ~105 R/hr. However, neutron detectors could be shielded with 0.2 inches of lead to reduce the gamma dose to ~30 R/hr.

However, is all of the fuel actually visible to each of the three detectors? Or, does only a small fraction of the fuel contribute the entire signal? These questions will be answered in the next two subsections by examining the vertical and horizontal response signature of the cask. To measure these responses, a series of calculations were performed in which only a limited number of fuel bundles in specific regions of the cask were allowed to emit radiation. In this way, the effects of attenuation and absorption through the fuel could be more readily ascertained.

**Vertical Profile**
Fig. 3 shows the fraction of the total signal rate for each of the three detectors as a function of fuel basket for a fully loaded cask of uniform burnup. As expected, none of the detectors are able to see the whole cask. For neutron detection, it can be seen that each detector sees approximately 1/3 of the cask. That is, the neutron signal for the Bottom detector originates almost entirely from Baskets 1-3. Similarly, the majority of the neutron signal for the Middle detector comes from Baskets 4-6 while the majority of the neutron signal for the Top detector comes from Baskets 7-9.
Figure 3: Fraction of measured signal for each detector originating from specific baskets. The Bottom detector is placed near Basket 2, the Middle detector is placed near Basket 5, and the Top detector is placed near Basket 8.

In fact, there is a remarkable symmetry in the expected count rate for the three detectors. Each of the three detectors provides excellent coverage of nearest three baskets. For all three detectors, about 55% of the measured neutron signal for each detector comes from the nearest basket with about 20% coming from each of the next-nearest baskets.

For gamma-ray detection, it is seen that each detector effectively sees only one of the nine baskets. For example, about 95% of the signal measured by the Bottom detector originates from Basket 2. The next-nearest baskets combined (Basket 1 and 3 in the case of the Bottom detector) contribute only ~5% of the total signal to each detector. Of course, this means that a minimum of 9 gamma-ray detectors would be needed.

Horizontal Profile
It was just shown that a system of 3 neutron detectors provides better vertical coverage of the cask contents than 3 gamma detectors. It is also of interest to investigate the horizontal sensitivity of the detection system. That is, can the detectors see fuel in the interior or the opposite side of the fuel baskets?

Fig. 4 shows the fraction of the signal for a single detector as a function of position within a basket. The horizontal response of the basket is broken into six regions (wedges) for the case of the neutrons and 2 regions (halves) for the case of gamma rays. It can be seen, for example, that approximately 40% of the neutron signal for a given detector comes from the near third (Wedges 1 and 2) of the nearest basket. About 2% of the neutron signal comes from the far third (Wedges 5 and 6) of the next-nearest basket. Thus,
the far side of the next-nearest basket is visible to neutron detectors in the reverification tubes.

![Graphs showing neutron and gamma-ray sensitivity](image)

**Figure 4:** Fraction of the measured signal (for a given detector) from the nearest and next-nearest baskets as a function of location within the basket.

For gamma-ray detection, comparing Fig. 4 with Fig. 3, it can be seen that, for both the nearest and next-nearest baskets, the near half of the basket contributes almost the entire signal from that basket. That is, gamma rays from the far side of the basket are almost completely absorbed or attenuated before reaching the detector. Thus, fuel bundles from the far side of the basket are not visible to gamma detectors in the reverification tubes.

Further gamma-ray calculations were performed in which only one half of the basket was allowed to emit radiation (Half #1 in Fig. 6) but that half was rotated by 60° with respect to the detector position. The measured gamma-ray signal was the same as when the basket had not been rotated. This suggests that the gamma-ray detector sees much less than one half of the basket. It is likely that the detector sees only the nearest third (or less) of the basket.

**DIVERSION SCENARIOS**

It has now been established which regions of the cask give the strongest and weakest responses to the detectors in the reverification tube. It was shown in the previous section that neutron detectors provide much better visibility of the contents of the cask than gamma-ray detectors. In this section we discuss 3 possible scenarios in which fuel has been removed from the cask. The first involves the removal of an entire basket. The second involves removing half of the fuel (the far-side half). The third involves determining how much material can be removed before the signal decreases by 5% of its original value.

If an entire basket is removed, the most likely scenario would be one which is not right next to a detector. For a 3-detector system, with detectors placed next to Baskets 2, 5, and 8, this would be one of the baskets which has previously been called a next-nearest basket (i.e. Basket 1, 3, 4, 6, 7, or 9). The effect of removing an entire basket on the detector systems is evident from Fig. 3. If a next-nearest basket is removed, the signal rate for one of the neutron detectors would decrease by ~20%. Conversely, the signal rate for one of the gamma-ray detectors would decrease by only ~2.5%.

If half of the fuel assemblies were to be removed (30 bundles from each of the 9 baskets), it is most likely that the far-side half, away from the detectors, would be removed. An inspection of Fig. 4 shows that the signal rate in each neutron detector would decrease by ~19% while there would be almost no decrease in the signal rate (< 0.5%) in gamma-ray detectors.
Finally, how much fuel could be removed from the cask before the detector signal rate decreased by 5% of its previous value? From Fig. 4 we see that for a neutron detector, Wedges 5 and 6 from a next-nearest basket contribute a total of ~2.6% to the signal rate for a given detector. Therefore, Wedges 5 and 6 from both next-nearest baskets contribute ~5% of the total signal for a given detector. Thus, 40 out of 540 bundles (7.4% of the fuel or about 0.35 SQ) from the cask could be removed before the rate for a single neutron detector dropped by 5%.

For gamma rays, it was shown that each detector sees, at most, three baskets, where the next-nearest baskets combined contribute only about 5% of the total signal for that detector. Therefore, the two next-nearest baskets could be removed. (Note that above it was shown that the far-side half of all three visible baskets could be removed with a less than 0.5% decrease in the rate.) If each of the three gamma-ray detector signals dropped by 5%, then 6 of the 9 baskets (or 66.6% of the fuel) could have been removed.

Increasing the number of gamma-ray detectors from three to nine provides better vertical coverage of the nine baskets. However, as evidenced in Fig. 4, it still does not address the problem of seeing spent fuel on the opposite side of the cask. A nine-detector gamma-ray system would need to be placed in both reverification tubes in order to have similar coverage as a three-detector neutron system in one reverification tube.

ASPECTS OF THE PROPOSED NEUTRON DETECTION SYSTEM

In the previous section we outlined the differences in detection capabilities between neutron and gamma-ray detection systems in CANDU spent-fuel storage casks. It was shown that three fixed-position neutron detectors provide much better visibility of the cask contents than three gamma-ray detectors. This is due to neutron multiplication and the higher penetrability of neutrons in the high-Z, high-density environment of the spent fuel. In this section we outline some of the other beneficial aspects of the proposed neutron detection system.

Although summing the three detectors through a single amplifier reduces the cost considerably without jeopardizing the ability to safeguard the cask contents, there is considerable advantage to reading out the detectors independently. This would provide for more flexibility in diagnosing anomalous read outs.

The previously mentioned detection level of 5% should be taken as an example only. A measured statistical precision of better than 1% is easily achievable in short collection times. Furthermore, it is expected that the cosmic-ray neutron background rate in the detectors will be less than 1 count/s so background variations should be negligible.

The proposed neutron detection system could be used in an unattended or attended mode of operation. In an unattended mode, a system would be fielded in each cask and collect data continuously. Each monitoring system would take data for a user-selected period and periodically download data to a central collection computer. Alternatively, the system could be used in an attended mode of operation. If the cost becomes too prohibitive to deploy a system in each cask, a single system could be used to periodically reverify the contents of a given cask. After an initial verification of each cask is performed, a random sampling procedure could be employed to reverify the contents of a few casks in the storage yard. This procedure could be accomplished by carefully repositioning the detectors within the reverification tubes of the casks to be measured. In this measurement scenario, short data collection periods will be sufficient to provide adequate measurement statistics. For a counting rate of 1750 counts/s, the statistical uncertainty on a 1-min run is 0.3%. As a further measure of confidence, in an attended mode of operation, the user could opt to place the detection system in each of the two reverification tubes of the cask individually. Then a strong signature would be measured from each side of the cask ensuring that any low count rate areas of the cask are more strongly sampled.
CONCLUSIONS
We have performed ORIGEN2 calculations to establish the expected neutron and gamma-ray source terms from the CANDU spent-fuel bundles. We then performed Monte Carlo transport calculations of neutrons and gamma rays in a CANDU spent fuel cask to compare the performance of proposed neutron and gamma-ray reverification systems. This was accomplished by investigating the sensitivity of the two systems to different regions of the spent fuel.

A neutron system provides excellent visibility of the spent fuel to safeguard the entire cask. The detector system would consist of three lead-shielded 4-atm $^3$He tubes placed near the bottom, middle, and top of a single reverification tube. The detectors will be read out independently to provide the maximum flexibility in data processing and analysis. Although not required to meet the safeguards criteria, the three-detector system could be placed in each reverification tube individually.

For gamma rays, a three-detector system clearly does not provide the necessary sensitivity to safeguard the entire cask. It has been proposed that 9 gamma-ray detectors or a single detector electro-mechanically driven up and down the reverification tube might be used to reverify the contents of the cask. Although this would provide greater vertical sensitivity to the 9 baskets, it still does not address the problem of seeing fuel assemblies in the interior or on the far side of the cask. Approximately 70% of the fuel bundles could be removed without significantly reducing the gamma-ray response. To provide better visibility of the cask, a nine-detector system would have to be placed in both reverification tubes but may still not meet the safeguards requirements.

If COK were never lost, gamma-ray detectors could be effective in monitoring the constant signal level from the spent fuel. However, after the temporary failure of the continuous monitoring system, or in an attended mode of operation in which casks are measured only periodically, the reveryification of the spent fuel would require a neutron measurement.

REFERENCES