Monitoring Defective CANDU Fuel Bundles

E. Corcoran

Masters of Science Student
Royal Military College (RMC) of Canada

Supervisors:

Dr. B. J. Lewis and Dr. W.T. Thompson

Abstract

In 2005, it was proposed that a passive substance such as Nanocrystals could be used to monitor and locate defect fuel elements in-core. The experimental goal was to determine if Nanocrystals could be used for this application. Originally nanocrystals tagging was suggested for current operational CANDU-600 fuel. Other methods, including noble gas tagging, are also being investigated. Moreover, the scope of the project has been extended to include the identification of Dysprosium-doped fuel in the new ACR fuel design. The purpose of this paper is to discuss the experimental progress made at RMC on this project.

1.0 Introduction

An efficient, reliable and cost effective means of locating defected fuel in-core has always been a goal of CANDU reactor operators, especially with the addition of a neutron absorber in the new Advance CANDU Reactor (ACR) fuel design (where the neutron absorber could potentially relocate if a fuel failure were to occur). Thus, a means of locating defective fuel bundles to minimize the potential for fuel loss and a possible increase in coolant activity levels of interest to the reactor operation.

In current CANDU-600 reactors several, techniques have been employed to detect and locate defective fuel, including gaseous fission product monitoring, delayed neutron detection and feeder scanning [1]. However, in recent years, such systems have been degrading and are prohibitively expensive to upgrade or install in new reactors (especially as reactor designers strain to minimize capital costs).

One possibility is the tagging of the fuel bundle with the addition of a passive substance of known properties that could be added to the fuel element during fabrication. In the event of a failure a portion of the tag would be released into the coolant and detected using specialized monitoring equipment. One possibility studied in this work is the use of nanocrystal tagging (NCT).

2.0 Current CANDU-600 Reactors

The **CANadian D**euterium **U**ranium (CANDU) is a Canadian designed **P**ressurized **H**eavy **W**ater **R**eactor (PHWR) that uses natural uranium oxide (UO_2) for fuel and heavy water (deuterium oxide or D_2O) as both coolant and moderator [2]. The reactor core is contained in a large cylindrical calandria tank, made of stainless steel, which holds the heavy water moderator at low temperature and pressure. The ends of the calandria tank are comprised of two thick end shield plates; these end plates support the fuel channels that pass horizontally through the calandria tank. Each fuel channel is composed of an outer calandria tube of Zircaloy-2 and an inner pressure tube of Zr-2.5% Nb. The fuel bundles are housed inside the pressure tube and heavy water is pumped over the bundles to cool them. The current operation CANDU-600 reactors have 380 fuel channels; each channel contains 12-fuel bundles [3].

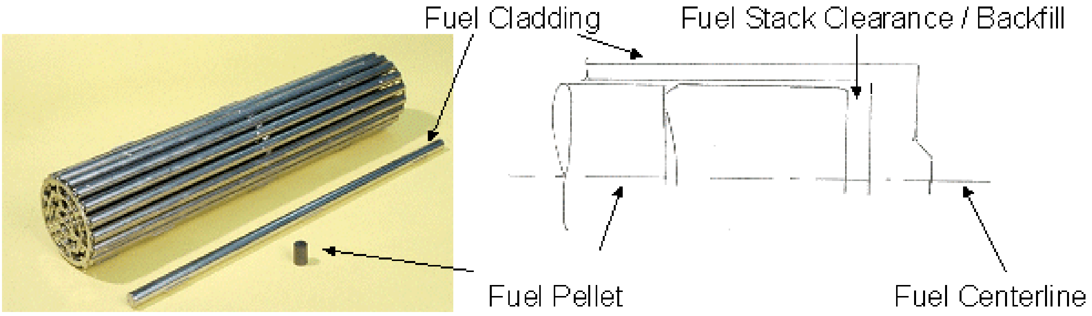

Figure 1. CANDU fuel bundle. [4]

Each fuel bundle is composed of 37 elements. The CANDU-600 fuel element is comprised of a number of sintered UO_2 pellets contained in a sealed tube fabricated from Zircaloy-4, 49.5 cm long and 13.08 mm in diameter, backfilled with helium [5].

3.0 Advanced CANDU Reactor (ACR)

The advanced CANDU reactor (ACR) is the ‘next generation’ of CANDU reactor. The ACR was designed to produce electrical power at a lower capital cost than current CANDU reactors. This reduction in capital cost is attributed to a decrease in reactor size and heavy water (D_2O) inventory. Size reduction was achieved by decreasing the number of fuel channels (284 verses 380 channels) and shrinking the spacing between channels (tighter lattice pitch). The D_2O inventory was reduced 75% by cooling the reactor with light water (H_2O) and using slightly enriched uranium (SEU) fuel. Higher thermal/electrical conversion efficiencies are also obtained by increasing the wall thickness of the fuel channels allowing the coolant to operate at higher pressures and temperatures [6].

3.1 ACR-LVRF Fuel

The ACR is a derivative of the Low Void Reactivity Fuel (LRVF) design conceived in the early 1990's as a means of reducing positive void reactivity in CANDU reactors. This reduction in void reactivity allows the ACR to comply with foreign countries' licensing and creates a fuel that is safer under accident conditions [6].

The ACR fuel has a 43-element design. The elements are arranged in rings of 1, 7, 14, 21 elements (seen in Figure 2). The elements in the ACR bundle have two sizes; the inner two rings have elements with a diameter of 13.5 mm and the elements in the outer two rings have a diameter of 11.5 mm. The difference in element sizes increases the safety margins and increases fuel performance at higher burnups. The outer 42 elements contain Slightly Enriched Uranium (SEU) or uranium that is enriched to 2.1% U-235 to counteract the 7.5 to 15 % of the neutron absorber Dysprosium oxide (Dy_2O_3) in the central element [6].

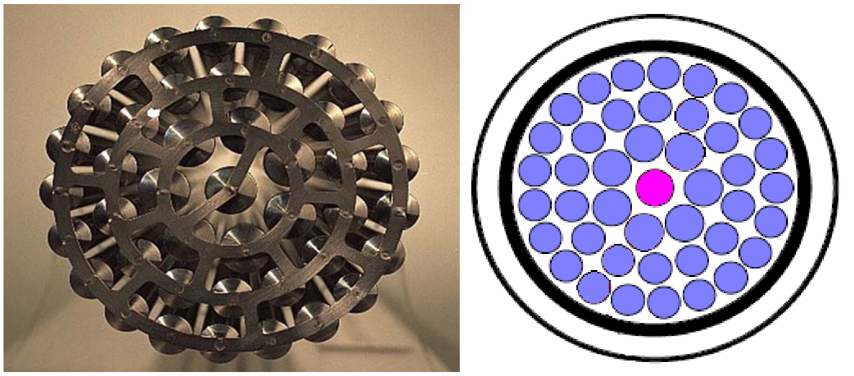

Figure 2. ACR- LRVF Fuel Design [7].

43-element design, the central element contains a neutron absorber (Dy_2O_3) mixed with UO_2, elements in the outer 3 rings contain SEU (2.1% U-325).

The ACR reactor produces 3 times less waste (as compared to current CANDU reactors) because the addition of SEU allows the fuel to be run safely to a burn up of 21 MWD/Kg (3 times current fuel burnup) [6].

The addition of Dy_2O_3 into the center of the fuel bundle reduces the positive void reactivity effect associated with a loss of coolant accident (LOCA) [8]. During a LOCA, with boiling coolant is lost from the reactor creating voids or gas pockets. Fuel bundles within the voids experience an increase in neutron flux followed by an increase in temperature at the center of the fuel bundle. The presence of the Dy_2O_3 neutron absorber in the center element prevents the spike in neutron flux during voiding, increasing safety [6].

Dysprosium (Dy) is a non-radioactive member of the lanthanide series. Dy is deemed a neutron absorber because of its very large cross section (~1700 barns) [9]. Dy was chosen for this application because of its' 'burnable neutron absorber' characteristics. As the fuel is spent and the fission products increase the Dy is converted to other elements with lower cross sections. Therefore, the void reactivity of the fuel bundle remains slightly negative throughout its lifetime in the reactor [6,9].

4.0 Nanocrystal Tagging

Nanocrystal tags are semiconductor Nano-particles that could be added to the helium-filled plenum of the fuel elements contained in a bundle. Unique tags could be used for each bundle and, in the event of a failure of an element, the nanocrystals would be released into the coolant with the fission products. These nanocrystals could be detected using luminescence spectrometer and traced back to the bundle containing the defective element.

4.1 Background

In semiconductors physics, the highest electronic energy level that is occupied by electrons is known as the valence band and the conduction band is an unoccupied level above the valence band. An electron-hole pair (or exciton) is formed when an electron in the valence band gains energy by thermal or photonic absorption and enters the conduction band. The difference in energy between the valence and the conduction bands is known as the bandgap energy (E_g in eV). Also, it is the amount of energy required to create an exciton pair and it is unique to the materials used to form the semiconductor [10].

Nanocrystals are semiconductor materials composed of IV-IV, III-V, II-VI and IV-VI compounds [11]. The difference between bulk semiconductor materials and nanocrystals is that nanocrystals have dimensions that are smaller than the excition Bohr radius (average physical separation between an electron and hole for a given material). When a semiconductor has dimensions smaller than the excition Bohr radius, the energy levels become discretized and the semiconductor ceases to exhibit the properties of a bulk semiconductor. Because of this decretized nature, addition or subtraction of a few atoms has the effect of altering the bandgap energy, which alters the absorbance and photoluminescence of the nanocrystal (Figure 1). Through careful material and size selection, the emitted wavelength from a nanocrystal can be predicted with extreme precision [12].

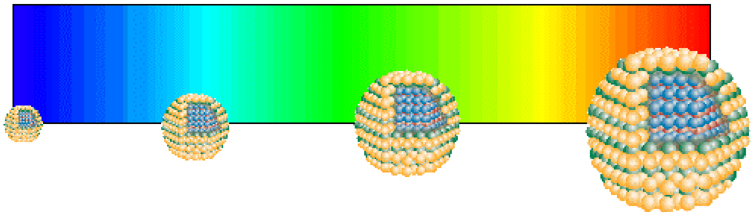

Figure 3. Relationship between nanocrystal size and emitted wavelength. [12]

Nanocrystals have broad excitation profiles and narrow symmetric photoluminescence emission peak profiles. This allows for multi-peaked emission from a signal sample containing a variety of nanocrystals (excited with monochromatic light) [12].

4.2 Experimentation

The experimental goal of this project was to determine if Nanocrystals will survive in the radioactive environment in a CANDU reactor for an extended period of time. Since it is not possible to experiment on Nanocrystals using a commercial power reactor, nanocrystals were irradiated using a neutron flux from a SLOWPOKE-2 (Safe Low POwered C(K)ritical Experiment) research reactor. The SLOWPOKE-2 generates a lower neutron flux (~10^{11} n $cm^{-2}s^{-1}$) as compared to a CANDU (vs ~10^{14} n $cm^{-2}s^{-1}$) thus making it an ideal tool for proof of concept experimentation.

Nanocrystal survival was determined by a comparison of the emission spectra (λ_{ex}=400 nm) in two samples that were irradiated for 8 hours and two samples that were not irradiated and used as a control. Both solutions were stored in the same conditions for the same amount of time and tested on the same day with the same equipment to reduce uncertainty.

For this project a variety nanocrystals were produced in the laboratory at the Royal Military College or obtained from other research groups and companies who fabricate nanocrystals. Aqueous ligand-capped CdTe, CdTe/ZnS nanocrystals were produced using the method outlined by Gaponik et al [13]. Polymer capped nanocrystals were obtained from the Q-dot corporation (USA), and silica capped nanocrystals were donated from the University of Freiburg in Germany [14].

4.3 Procedure

A stock solution of Nanocrystals was divided into four aliquots. Each aliquot was sealed in a 1.5 mL vial containing the nanocrystal solution and double de-ionized water (DDH_2O). Two aliquot vials were placed into one 7 mL vial and sealed. Two 7 mL vials were produced for each type of NC tested. One sample of each type was irradiated for 8 hours and the other was placed in a light-tight box for 8 hours.

After irradiation, both irradiated and non-irradiated samples were placed in a lead box for approximately two weeks. This allowed the irradiated sample to decay back to background radiation conditions and to allow the sample to be safely handled.

After the cooling period, the four 1.5 mL vials were removed from the 7 ml vials. A portion of the sample was removed from the 1.5 mL vial and was diluted to 10 mL in a volumetric flask of DDH$_2$O. This was repeated for all the samples to produce four separate 10 mL solutions. These solutions were then tested using the Perkin Elmer LS 50 B Luminescence Spectrometer. This procedure was repeated twice to ensure reproducibility.

To ensure that the sample solution was stable at low concentration for a two-week period, a dilute solution was created and the emission spectrum was measured using the spectrometer directly following dilution and after approximately two weeks. During the two-week period the sample was stored at room temperature and wrapped tightly in foil to prevent photo bleaching.

4.4 Experimental Results

Figure 4 shows the behavior of the characteristic emission peak at 590 nm for a dilute silica coated nanocrystal sample over a two-week period (without irradiation). Each curve was normalized to a wavelength value at 450 nm.

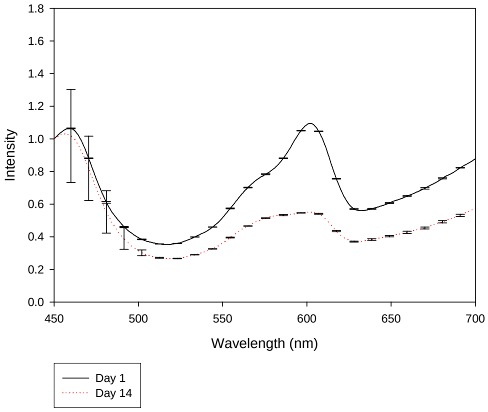

Figure 4. Average emission spectra (λex=400 nm) for a dilute sample (300 μL in 10mL of DDH$_2$O) on day 1(—) and after 14 days (· · ·).

Figure 5 shows a comparison of the original emission spectra results for the irradiated and non-irradiated samples.

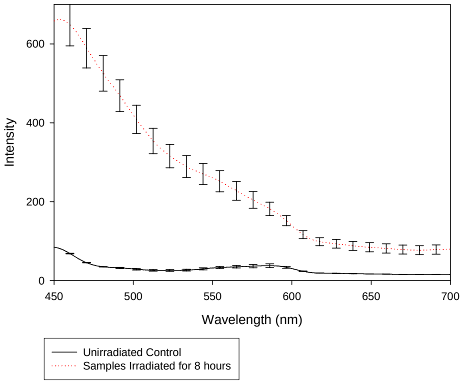

Figure 5. Average emission spectra (λex=400 nm) for sample irradiated for 8 hours (· · ·) compared to the average non-irradiated samples (—).

Figure 6 shows a comparison of the peak profile at 590 nm. Each curve was normalized to the value at 450 nm.

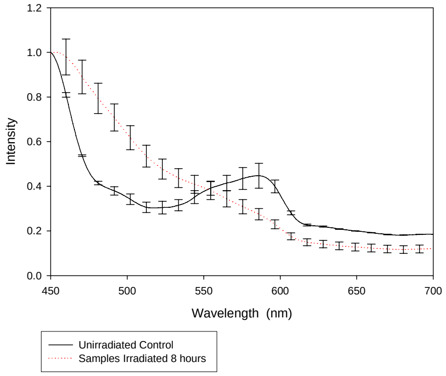

Figure 6. Comparison of peak shape for normalized emission spectra (λex=400 nm) for a sample irradiated for 8 hours (· · ·) compared to a normalized non-irradiated sample

(—).

Figure 4 shows that the characteristic emission peak (590 nm) is slowly decreasing but is still distinguishable after the 14-day storage period. Figure 5 shows an increase in the scattering of the emission for the irradiated sample compared to the non-irradiated sample. However, normalized spectra in Figure 6 reveal the loss of the characteristic peak (at 590 nm) when the silica coated nanocrystal sample is irradiated.

The increase in emission and the loss of the characteristic emission profile in Figure 6 may be the result of an increase in scattering as the nanocrystal degrades in solution. The mechanism for this degradation appears to be the result of the neutron flux and not the result of the storage of the sample at low concentration. Therefore, it can be concluded that the silica coating does not protect the nanocrystal during neutron irradiation.

The CdTe, CdTe/ZnS nanocrystals and polymer-coated nanocrystals followed the same trend as the silica coated Nanocrystals. All of the nanocrystals showed a lost of the characteristic peak after an eight-hour irradiation in the SLOWPOKE-2 reactor at a flux of 5 x 10^{11} n $cm^{-2}s^{-1}$. The CdTe and CdTe/Zn solutions were not stable in dilute solution for more than two weeks. Therefore, it can be concluded that nanocrystals in general are not viable as a passive tag for defective fuel identification in current or future CANDU reactors.

4.5 Future Work

Simpler, alternative tagging systems such as noble gas tagging will also be investigated. Work with the new ACR fuel will include studies to determine if the Dysprosium can be detected by conventional gamma ray spectrometry methods in reactor coolant water in the case of a defect of the central element. It is hoped that the Dy can be detected over the fission product spectrum. If this were the case, then the operators would at least know if there were defective Dysprosium-doped elements.

5.0 Conclusion

Nanocrystals are not a viable material for monitoring defective fuel bundles in CANDU reactors. The nanocrystals degrade in a neutron flux as shown from experimentation with the SLOWPOKE-2 reactor. Although nanocrystal techniques were unsuccessful, further investigation with simpler tagging methods such as noble gas tagging shows promise. In addition, work is continuing on the identification of Dysprosium in the reactor coolant (along with fission products) using conventional gamma-spectrometry methods. Further investigation into these areas could lead to a defective fuel-monitoring tool that is safe, effective and reliable.

6.0 Acknowledgments

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