

## Short, Medium and Long Term Consequences of Inadequate Defect Fuel Management

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### Abstract

Abstract – Defect fuel pencils result in short, medium and long term consequences to the environment within and external to the nuclear power station. The paper will describe these consequences and specify the Defect Fuel Management Practices required to avoid these consequences.

### 1. Introduction

This paper is written from the perspective of the plant chemist who has accountability for minimizing corrosion of, environmental emissions from, and radiation fields emanating out of nuclear power plant systems. The authors are expert in neither fuel and physics, nor reactor safety, nor in fuel design, rather persons who have operated and overseen the operations of nuclear plants from the chemistry, materials and environment perspectives.

Within the past year or so, unanticipated contamination has been encountered in the heat transport systems of some CANDU reactors. Contamination is expected to be encountered primarily as a result of activated corrosion products and, occasionally, fission products. The contamination encountered in 2009 had low ratios of (total) beta plus gamma ( $\beta\gamma$ ) radioactivity to (total) alpha ( $\alpha$ ) radioactivity. What this means is that at lower ratios of  $\beta\gamma$  to  $\alpha$  the contamination monitoring, contamination control and work execution become much more challenging and time consuming. This obviously is undesirable as maintenance work becomes more involved and costly. The greatest concern, of course, is safety of the station staff.

### Production of alpha emitting radioisotopes

CANDU fuel currently uses natural isotopic uranium (0.7%  $^{235}\text{U}$ ) in the form of uranium dioxide pellets. Fission of  $^{235}\text{U}$  results in fission products, most of which decay by beta and/or gamma emission, and neutrons. Some of the neutrons produced are consumed by fission, some are consumed by nuclei, of fission products and materials of construction, having (thermal) neutron capture cross sections and some are absorbed by nuclei of  $^{238}\text{U}$ . The formation of transuranic elements (including  $^{239}\text{Np}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{242}\text{Cm}$ ,  $^{244}\text{Cm}$ ) many of which decay by alpha emission, is through a combination of neutron activation and transmutation, starting with  $^{238}\text{U}$ . It should be noted that the fissioning of plutonium produces about half the energy produced from the fuel; not all of the plutonium produced is “burned”.

### 3. “Tramp uranium”

There are several definitions of “tramp uranium” which I have encountered. A popular definition is “exterior fuel contamination on new fuel bundles”. The definition of tramp uranium used in this paper is “fuel (fissile/fissionable material) that is outside the fuel cladding but within the heat transport system”. This would also refer to fuel which had previously been “deposited” within magnetite and occasionally reappeared as a result of a “crud burst”.

When a new CANDU reactor goes to power we expect to see small amounts of fission products in both the heat transport system and the annulus gas system. These fission products arise from fission of uranium remaining in the zircalloy alloys from metallurgical refining processes.

The radioisotope  $^{134}\text{I}$  is used as a marker for “tramp fuel” within the heat transport system. This is because when the uranium/plutonium is in the coolant, as opposed to within the fuel pencil, the  $^{134}\text{I}$  is released, in effect, instantaneously into the coolant. Within the fuel pencil, the  $^{134}\text{I}$  is contained within the grain of uranium dioxide and has to be released from the grain. Usually this release is unlikely without the presence of water. The time for diffusion of the  $^{134}\text{I}$  from its creation in the grain, through the grain and into the coolant is sufficiently long that the  $^{134}\text{I}$  has mostly decayed. The half-life of  $^{134}\text{I}$  is 52.5 minutes. For comparison, half-lives of some other observed radioisotopes of iodine are 20.8 hours for  $^{133}\text{I}$ , 6.6 hours for  $^{135}\text{I}$  and 8.05 days for  $^{131}\text{I}$ .

As long as the fission products and fuel remain within the fuel pencil there is no problem. The problems begin when fission products and fuel escape from the fuel pencil. What are known, but perhaps under appreciated, are the immediate consequences of the release of fission products into the coolant. These immediate consequences include increased environmental releases of fission produced radioactive noble gases, increased dose rate on surfaces from depositing fission products, such as  $^{99}\text{Mo}/^{99}\text{Tc}$ ,  $^{140}\text{Ba}/^{140}\text{La}$ , and increased dose rate from the water containing soluble fission products such as radioiodines.

The requirement to approach the reactor face for maintenance purposes is restricted to unit outages. However the mechanical staff, who maintain the fuelling machines, have to get up close and personal with the fuelling machines routinely. This work includes replacing the snout seal ring which requires hands to be placed within the fuelling machine snout. This seal ring, which serves as the pressure boundary between the fuelling machine and the endfitting, is expected to wear and has to be replaced routinely.

Hence the immediate consequences of fuel failure can lead to both increased environmental radioactive emissions and increased radiation exposure to some station staff. A significant increase in radioiodine concentration can result in operating constraints, including unit shutdown.

$^{133}\text{Xe}$  (half-life 5.25 days) is the noble gas produced by fission having the longest half-life; increased radioiodines in the heat transport system and increased dose rate on heat transport system and fuelling machine surfaces result from depositing fission products. Note that whilst the gamma energy of  $^{133}\text{Xe}$  at 80 keV is low, a huge quantity of this radioisotope is produced within the fuel.

It is interesting to note that for those Units at Bruce B the background  $^{131}\text{I}$  and  $^{134}\text{I}$  concentrations are considerably lower than those at Bruce A [1, 2]. This is believed to be a direct indication of the amount of “circulating” fissile/fissionable material in the heat transport system. Also of note is that the higher background in the Units 3 and 4 heat transport systems was observed following the return of those units to power operation in late 2003, early 2004.

## **4. Fuel Defects**

### **4.1 Causes of Fuel Defects**

There are two basic causes of fuel defects. These are fuel manufacturing issues and “foreign material” within heat transport systems.

The fuel manufacturers, generally, do a good job of producing fuel that performs as required. There have been instances when fuel performance has been significantly less than desired such as the fuel that was loaded into the Douglas Point and Bruce Unit 3 reactors in 1983.

Foreign material can be introduced inadvertently as a result of maintenance or construction activities. The latter will be described later for Bruce B. Design changes can also lead to foreign material generation such as when the Bruce B units were changed from fuelling against the flow to fuelling with the flow.

The consequences of fuel defects do not, as many of us previously believed, result in only short term, acute events, but rather long term consequences, as described later. Hence fuel defects must be eliminated, if at all possible.

### **4.2 Fuel defect progression**

Manzer elegantly describes onset of fuel defects and oxidation of fuel in his papers [3, 4, 5]. Once the cladding is penetrated, radioactive noble gases are first released. As a result of corrosion combined with water radiolysis the size of the penetration will grow to the point at which water can enter the fuel pencil. More noble gases and soluble radioisotopes (including delayed neutron precursors,  $^{87}\text{Br}$  and  $^{137}\text{I}$ ) are released from the fuel, initially in small quantities as is fuel, some of which has oxidised. As time progresses the size of the hole in the cladding grows as does the release rate of fission products and fuel particles.

### **4.3 Removal of defect fuel**

The removal of defect fuel from the core has been a priority for those interested in minimizing dose rates, minimizing environmental releases and minimizing the spread of contamination within the nuclear power station. This priority appears not always to be shared with others as evidenced by the apparent lack of urgency to locate and remove defects when they are discovered.

Some CANDU plants, by design, have greater challenges than others in detecting the onset of fuel failure and locating the fuel channel containing the defect, nonetheless those plants

eventually are successful at removing the defect. I would argue that the period between recognition that a defect has occurred and the removal of that defect, from the core, is too long in almost every case at all plants.

There is no question that having to fuel out a defect costs money. New fuel costs money. When specifically trying to remove a defect, fuel is not being added to increase reactivity, rather to eliminate the source of fission products and tramp fuel. In addition, depending on the design of the station, several fuel channels will have to be visited and refueled prior to the successful removal of the bundle(s) having the defected fuel pencil(s). The work of the fuelling engineer can be greatly facilitated by the output of the gaseous fission product detector system and the delayed neutron detection system, minimizing those fuel costs and the valuable time of the Fuelling Engineers spent locating the defect.

There are even those plants which by design were fitted with functioning defect fuel identification and location systems but have either allowed the systems to fall into disrepair or have even to chosen to remove those systems from service!

## **5. Short term effect of fuel failures**

As stated earlier, following onset of a fuel defect increased environmental emissions of radioactive noble gases occurs along with an increase, albeit small, in radiation fields and a greater increase in radiation fields and contamination levels around and in the fuelling machine. The issue is such that one station purposely removes the fuelling machine from service for several days following removal of a fuel defect from the reactor. This action is taken to prevent the possibility of contaminating another unit with radioiodine. An ion exchange capability, which was omitted in the original fuelling machine design, could address this issue.

At Bruce Unit 1 there was a significant increase in radiation fields and contamination levels in 1979 following the P-13 event [6]. During this event a fuel bundle was crushed within the heat transport system. The radiation fields from fission products immediately increased around the heat transport system. Contamination levels within the heat transport system would also have increased.

## **6. Medium term effect of fuel failures**

The medium term effects of fuel failures can result in continuation of the short term effects, described earlier, and an increased difficulty to detect and locate small fuel defects. This is the consequence of fuel particles having left the confines of the fuel pencil. Manzer and Boss suggest that 90% of the fuel that leaves the pencil exits the fuel channel [7].

The 1979 Bruce Unit 1 P-13 event [6], in which a fuel bundle was crushed, resulted in significantly increased fission product contamination and fission product radiation fields around the heat transport system. However, the nuclear community noted with interest that, about one year later, the heat transport system radiation fields, had returned to the levels measured prior to

the event. At the time I recall a feeling of relief that the “problem” had corrected itself. However, other issues had arisen.

It is not clear whether it was Bruce Unit 1 P-13 event, or the consequence of additional events, that led to the significant contamination issues in the fuelling machine maintenance area. Personal protective equipment for access to the fuelling machine maintenance area at Bruce A increased to the requirement to wear double plastic suits. This was a direct consequence of breach of fuel cladding but emphasizes the need for excellence in fuel, and defect fuel management, to minimize and control contamination levels outside the heat transport system. The loss of fuel will lead to contamination challenges.

Fuel that is outside the fuel cladding and is within the core, either as a result of deposition or that released as a result of a crud burst, will undergo fission. Thus fission products will be released directly into the coolant. Should the quantity of this tramp fuel be allowed to climb then the background levels of fission products, in the coolant, will increase. This increase in background of fission products will mask the ability to detect small fuel defects. Thus early detection of small fuel defects will be masked. Hence an additional medium term consequence of fuel failures can be increased difficulty to detect both onset of fuel failure and the ability to locate the defect in a timely manner. This is a condition which, if not remedied, can worsen.

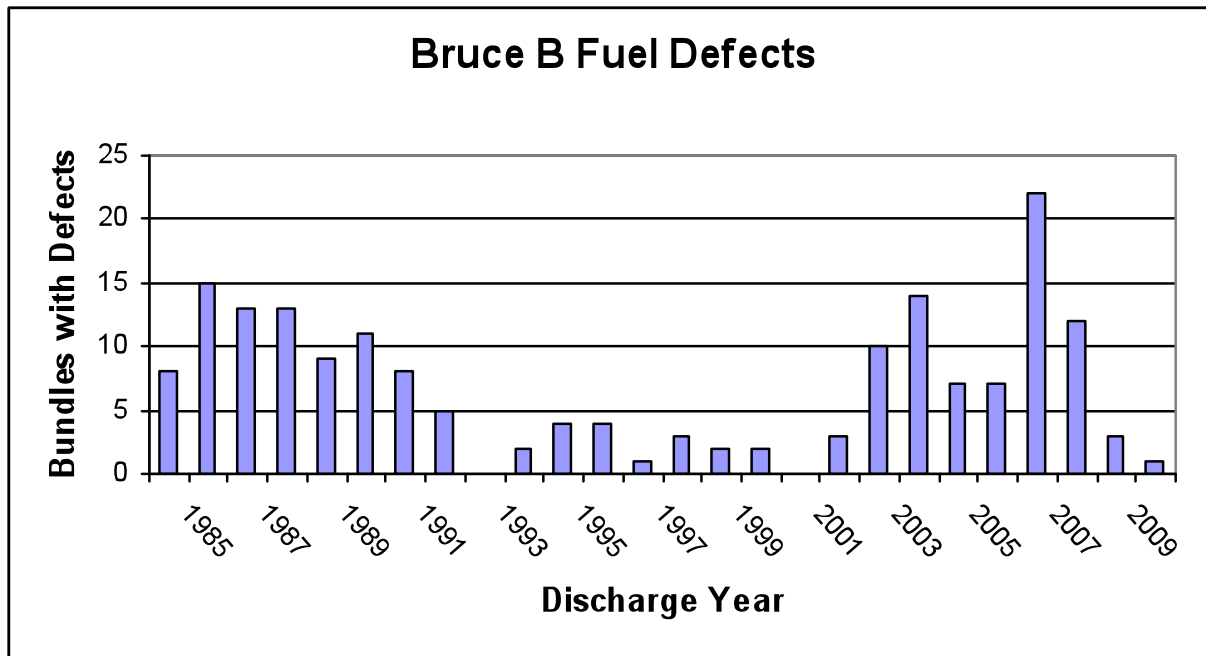
## **7. Bruce B**

I was the chemist supervising the construction and commissioning of Bruce B. The events at Bruce A made me determined that I was not going to allow Bruce B to get into a similar mess with respect to contamination levels in heat transport systems and associated support areas. An undocumented agreement was arrived at between Dan Austman, the then Reactor Physicist for Bruce B, and me. This agreement was that we would do our utmost to identify and remove from the core any fuel defects as soon as possible. At this time my staff looked after the gaseous fission product detection system and Dan’s staff looked after the delayed neutron detection system equipment. The practice of removing defect fuel as quickly as possible was conducted effectively for at least the first six years of operation.

During the construction of Bruce B the cause of failure of the Pickering Unit 1 G16 fuel channel was learned. Garter springs, or spacers were incorrectly positioned allowing fuel channel to calandria tube contact. Bruce Unit 6 had just completed “hot commissioning” and the shutdown guarantees were about to be surrendered to allow the start of “Phase B” commissioning. It was essential that the location of the Bruce B garter springs were determined and, as necessary, repositioned. This work necessitated the removal of the fuel that had been, as per usual practice, dry loaded manually. One morning, I was surprised by two plant operators who were holding what appeared to be swarf. I immediately assumed that they were trying to “pull my leg” and asked if they had visited a lathe in the mechanical workshop. The operators led me into the clean room area of the Unit 6 reactor vault and to the open fuel channels. To my great astonishment I observed swarf in the open fuel channels. The fuel bundle bearing pads had been, in effect, machining the fuel channels. This discovery led to the use of the “shim” for dry loading of fuel in all subsequent CANDU reactors.

Unit 6 was not alone with respect to the swarf problem. Ten channels of Unit 5 were fuelled for hot conditioning as a test to demonstrate whether or not the garter springs would be held in place. It was learned, as anticipated, that the centre two garter springs of those channels containing fuel did not move during operation of the main heat transport system pumps.

Unit 6 achieved “criticality” in May 1984 and Unit 5 in November 1984. Unit 7 achieved criticality in 1986 and Unit 8 in 1987. Units 6 and 5 suffered more than double the number of fuel defects than Units 7 and 8 during their first five years of operation. The reason was the swarf in Units 6 and 5.



**Figure 1 – Histogram showing Bruce B confirmed fuel defects by discharge year [8]**

Figure 1 shows the number of fuel defects experienced at Bruce B since first power. The data show that the number of “confirmed” fuel defects reduced with time and remained low until the design change to implement fuelling with flow occurred. The defect rate has since significantly reduced since the introduction of the new fuel carriers.

	Unit 5	Unit 6	Unit 7	Unit 8
1985	1	7	0	0
1986	7	7	1	0
1987	4	6	3	0

<b>1988</b>	5	1	2	5
<b>1989</b>	3	3	2	1
<b>1990</b>	2	5	0	4
<b>1991</b>	2	2	2	2
<b>1992</b>	2	0	2	1
<b>1993</b>	0	0	0	0

**Table 1 showing the individual number of confirmed defect bundles removed from each of the four Bruce B Units during the first nine years of station operation [8].**

<b>Unit 1</b>	<b>Unit 2</b>	<b>Unit 3</b>	<b>Unit 4</b>	<b>Unit 5</b>	<b>Unit 6</b>	<b>Unit 7</b>	<b>Unit 8</b>
33	33	197	21	44	48	37	50

**Table 2 showing total number of confirmed defect bundles from each of the eight Bruce Units since operation began [8].**

The data for “confirmed” defects removed from Bruce A Units is considered preliminary until paper records have been reviewed. The Bruce B data for defects removed from each Unit is correct [8].

A “confirmed” fuel defect is a defect observed during visual, in-bay, inspection.

## **8. Primary Coolant Gamma Analyses**

Many advantages exist as to having a good gaseous fission product detection (GFP) system. These include analyzing pressurized coolant samples in “real time”. Should the gamma detector of the GFP system have good efficiency up to 2000 keV then much more can be learned with respect to system radiochemistry than if the detector has good efficiency only up to 500 keV. Darlington NGS, for example, made excellent use of this capability during the early years when fuel was being damaged as a result of acoustic vibration induced by the original main heat transport pump impellers. In addition, the samples analysed by the GFP do not have to be depressurized. Thus the GFP system can provide data which otherwise can be very difficult, if not impossible, to obtain.

The samples of primary coolant taken by laboratory staff are depressurised. The depressurisation process does not affect the radioiodine concentrations but does affect the noble gas concentrations. Hence for grab samples, the radioiodine concentrations determined are representative of the heat transport system, whereas the same cannot be stated for the radioactive noble gases.

For both gaseous fission product and delayed neutron detection systems, the lower the background counts for the detectors the easier it is to detect an increase in concentration of fission products.

For comparison the Bruce A (Units 3-4) and Bruce B (Units 5-8) average  $^{134}\text{I}$  concentrations are given:

January 2004 – July 2010	Unit 3	Unit 4
$^{134}\text{I}$ $\mu\text{Ci/kg}$ (average)	44	40

**Table 3: Units 3 and 4 average  $^{134}\text{I}$  concentration since restart [1]**

October 1993 – July 2010	Unit 5	Unit 6	Unit 7	Unit 8
$^{134}\text{I}$ $\mu\text{Ci/kg}$ (average)	11.0	9.4	6.2	5.5

**Table 4: Units 5-8 average  $^{134}\text{I}$  concentration since October 1993 [2]**

The available data suggest that although Unit 4 had half the number of fuel defects experienced by the Bruce B units, the  $^{134}\text{I}$  concentration is higher by a significant margin. This suggests that more fuel was lost into the Unit 4 (and Unit 3) heat transport system than into those heat transport systems at Bruce B. In addition, even though Unit 3 suffered a greater number of defects than Unit 4 the “tramp fuel” concentration would appear to be similar.

A review of the Bruce B  $^{134}\text{I}$  data suggests that the heat transport systems of Units 6 and 5 have more “tramp fuel” than those of Units 7 and 8. The Unit 5 data may have been exacerbated by some defects that had remained in the core for many months. The long residence time of the defects may well have resulted in increased fuel loss into the heat transport system.

## 9. Long term issues of fuel defects

Late in 2009 an issue of alpha contamination was recognized in Bruce Unit 1. This was the consequence of the ratio of total  $\beta\gamma$  to total  $\alpha$  being low. Low ratios have subsequently been seen in other Bruce A units and also at other sites. The explanation offered by some people for the low  $\beta\gamma$  to  $\alpha$  ratio is that it is the consequence of units having been shut down and not operated for a number of years; during this period the total  $\beta\gamma$  to  $\alpha$  ratio would reduce as a result of differences in half-lives with the longer lived alpha radioisotopes predominating. As logical as this argument seems it does not explain the observation reported at another plant which had not experienced a shutdown of several years. Thus the argument would not appear to be decay of the  $\beta\gamma$  emitting radioisotopes.



The explanation is tied, rather, to the fuel that has been lost from defected pencils. The fuel released from the pencil, and channel, is believed to deposit on and/or incorporate into the magnetite film within the outlet feeders, headers, steam generators, headers and inlet feeders. This magnetite offers a vast surface area for adsorption, absorption or other incorporating mechanism.

The Bruce B Units have all experienced internal removal of magnetite from steam generator tubes. Interestingly there has not been a significant decrease in the  $^{134}\text{I}$  concentrations following the magnetite removal process. This suggests that the majority of tramp fuel is not residing within the steam generator tubes. However, the evidence from Bruce Units 1 & 2 suggest that tramp fuel does reside within feeders and steam generators. Another possible explanation is that the chemistries of the transuranic elements behave differently under a given set of heat transport system operating conditions.

Informal discussions with individuals who have researched into activity transport of heat transport systems over the past forty years have revealed that much work was performed on reducing radiation fields, primarily from  $^{60}\text{Co}$ . There appears to have been little, if any, work performed on the chemistry of the transuranic elements under various CANDU heat transport system operating conditions.

There is no question that the dearth of knowledge in this area must to be corrected. Bruce Power has initiated investigations into transuranic chemistry under heat transport system conditions. It is vital that an understanding of the chemistries of these elements is available. This understanding will help to:

- (a) ensure the appropriate priority to remove defect fuel to minimize tramp fuel and transuranic elements within the heat transport system, and
- (b) forecast the radiological conditions to allow appropriate strategies to be used during fuelling machine maintenance, feeder replacement and reactor refurbishments.

## 10. Conclusions

- Fuel defects result in not only acute short term issues, rather long term issues related to contamination and contamination control.
- The low ratio of  $\beta\gamma$  to  $\alpha$  contamination encountered at Bruce, and elsewhere, is related to loss of fuel as a result of fuel defects.
- The low ratio of  $\beta\gamma$  to  $\alpha$  contamination is not the result of decay of  $\beta\gamma$  emitting radioisotopes, rather the result of other phenomena.
- Gaseous fission product and delayed neutron detection systems must be used to their full capability.
- Identification, location and removal of fuel defects must occur with urgency to reduce loss of fuel into the heat transport system to the minimum.
- Current fuel manufacturing processes, and especially those for the future, must improve to eliminate the occurrence of fuel defects from manufacturing issues.

- Design changes, maintenance work, refurbishment and construction activities must eliminate the introduction into the heat transport system of “debris” or “foreign material” otherwise these “debris” and “foreign materials” will cause fuel failures.
- The chemistries of the transuranic elements must be understood to assist with strategies for maintenance, feeder replacement and refurbishment activities.

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