IMPROVED PERFORMANCE FOR UO$_2$ FUEL

by

J.A.L. ROBERTSON

Chalk River Nuclear Laboratories
Chalk River, Ontario
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ABSTRACT

A program to monitor the performance of fuel in the Douglas Point Nuclear Generating Station revealed a small but significant increase in the fuel failure rate. Co-operation between the utility concerned (Ontario Hydro), Canadian fuel fabricators and AECL has resulted in fuel with improved performance being selected for commercial exploitation. The new fuel is already in production with the first bundles in service at the Pickering Nuclear Generating Station.

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Amélioration de la performance du combustible UO$_2$*

par

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Résumé

Un programme destiné à contrôler la performance du combustible de la centrale nucléaire Douglas Point a révélé une petite mais significative majoration du taux de rupture du combustible. Une étude effectuée conjointement par la Commission électrique ontarienne, les fabricants canadiens de combustible et l'EACL a permis de choisir un combustible à performance améliorée pour les réacteurs de ces centrales. Le nouveau combustible est en voie de fabrication et les premières grappes sont déjà en service dans la centrale nucléaire Pickering.

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Introduction

The outstanding performance of the first three units of Ontario Hydro's Pickering Nuclear Generating Station has focussed attention, both at home and abroad, on CANDU (Canada Deuterium Uranium) reactors. The four 540 MW(e) reactors at Pickering represent the first commercial units of the CANDU reactor system. Pickering-1 achieved criticality in February 1971 and was declared in-service just 22 weeks later. This period was progressively reduced to 15 weeks for Pickering-2 and to 5½ weeks for Pickering-3. The fourth unit is due to be started up in early 1973. In the station's first 12-months of operation, units 1 and 2 had net capacity factors over 80%, the target for a mature station.

The CANDU reactor system is a distinctively Canadian product from initial concept through design, development and supply to construction and operation, whose success is largely attributable to continuing co-operation between AECL, the utilities and Canadian industry. One characteristic of CANDU reactors, reliable fuel at low cost, provides an example of the effectiveness of this co-operation, where a potential problem was identified and a solution found in a remarkably short time.

Douglas Point Fuel Performance

The fuel for all existing and committed CANDU reactors consists of short (50 cm) bundles (Figure 1) of UO₂ fuel pellets in zirconium-alloy sheaths. In the Douglas Point reactor² 19-element bundles fit in 8-cm diameter pressure tubes that contain the coolant, while in the Pickering reactors each bundle consists of 28 elements because of the larger diameter (10 cm) pressure tubes. However, individual elements have approximately the same maximum design power output in each of these reactors — 500 W/cm. The element's power output can be related to the temperature at the fuel axis and, hence, provides a measure of the severity of the operating conditions.

Figure 1 — Fuel bundle for Pickering reactors.

Judged by any standards the performance of Douglas Point fuel has been excellent. Up to mid-1971(¹) less than one percent of all bundles loaded into the reactor had failed, including those suspected of having failed but in which a defect could not be positively identified². At mid-1972 the figure was still under one percent and, indeed, had decreased significantly.

The fuel's performance was being monitored in sufficient detail that an increase in the failure rate, still within the one percent figure, was detected in

¹ A 200 MW(e) prototype power reactor owned by AEC(¹), and operated by Ontario Hydro.

² Much lower failure rates can be obtained by quoting the percentage of all elements that have failed. However, since a bundle must be removed from the reactor to stop the release of activity when an element fails, the bundle failure rate is the more meaningful.
Spring 1970. Since CANDU reactors, with the capability for on-power fuelling, allow failed fuel to be discharged without shutting down the reactor the direct economic penalty of such low failure rates was unimportant. However, the need to discharge specific bundles imposes unwelcome restraints on fuel management and any radio-active release to the coolant impedes maintenance. We therefore decided that the cause of the failures should be identified and, if possible, eliminated.

In CANDU reactors, bundles are moved progressively through the pressure tubes until the fuel has achieved its design burnup. Thus each bundle experiences an increase in power after a prolonged period at relatively low power. Analysis of the Douglas Point records showed that many of the failed bundles had undergone such power increases shortly before giving evidence of failure. Others had apparently failed after the reactor power had been raised to 100% of the design value after several months at 75%. Since fresh fuel has survived irradiation at powers considerably higher than the maximum in the Douglas Point reactor, the prior exposure at low power must somehow be making the fuel more susceptible to failure. In the Douglas Point reactor the failure probability correlated equally well with the magnitude of the power increase and with the final power attained, so that it could not be said which variable was the more important. Also, the fuelling schedule was such that many bundles that passed through the region of maximum power in a matter of minutes exhibited no failures, suggesting that some form of delayed failure mechanism was involved.

Hydriding of the Zircaloy sheath by hydrogenous impurities left in the fuel during its fabrication is a well-known cause of sheath cracks and was, of course, suspected. Indeed, we had demonstrated in experimental irradiations how the deliberate inclusion of impurities could lead to failures, and the failure of some prototype bundles tested in the NRU test reactor at Chalk River had been attributed to this cause. However, the control exercised over the commercial production of CANDU fuel appears to have been adequate to prevent hydriding failures. Although examination of the failed bundles showed much hydriding damage, detailed analysis revealed that the hydrogen in the sheath consisted largely of the deuterium isotope. Thus, the hydrogen must have come from heavy-water coolant that had penetrated the failed element, so that much of the observed damage (Figure 2) was the consequence of the original failure rather than its cause.

![Figure 2 — Close-up photograph of a failed fuel bundle from the Douglas Point reactor showing the nature of the damage.](image)

Similarly, excessive sheath corrosion by the coolant was considered as a possible mechanism for sheath failure, but eliminated. It is known that poor control of the coolant chemistry, particularly in boiling systems, can lead to the build-up of a thick layer of crud on the sheath surface, and hence to high sheath temperatures and accelerated corrosion. Fuel failures in the US Boiling Light Water and the UK Steam Generating Heavy Water reactors have been attributed to this cause. However, the Douglas Point coolant does not boil and its fuel has never exhibited such heavy crud deposits: no signs of excessive sheath corrosion have been seen.

Other potential failure mechanisms that were eliminated included:
- faulty components or welds in the fuel bundles,
- gross overpower of the fuel, say $\geq150\%$ of the design value,
- mechanical damage during fuel handling.

Although each of these has been demonstrated to cause failures under particular circumstances none could account for the bulk of the Douglas Point observations.

Power Increase Tests

The significance of a power increase after an earlier exposure at lower power was confirmed in
well-controlled irradiations in the NRU test reactor. Here, full-size bundles could be irradiated in two water-cooled loops under representative conditions for CANDU power reactors. A mechanical device had been developed that would move fuel bundles within a loop, between positions of low and high power, while the reactor was operating. In a typical experiment a fuel bundle was irradiated for several months in a low power position of the loop, such that outer elements operated at about 250 W/cm. When the fuel burnup reached about 2000 MWd/tonne U, equivalent to a fluence of about $2 \times 10^{20}$ neutrons/cm$^2$ (Energy > 1 MeV) on the sheath, the bundle was removed from the loop, examined in the hot cells to ensure that it was still in good condition, then assembled on the fuel-moving device. When the bundle was returned to the loop it was kept in its original position for at least a day, then it was moved to a high power position (outer element 500 W/cm) in about 20 seconds. When gamma-monitors in the loop circuit detected significant increases in the coolant activity the reactor was shut down and the bundle was removed for examination.

The most obvious conclusion from the series of fuel-moving tests was that increasing the power of fuel that has already been appreciably irradiated does indeed cause failures, confirming what had been suspected from the Douglas Point reactor records. Closer examination of the results showed that the failure probability was very much greater in the NRU tests than in the normal fuelling schedule of the Douglas Point reactor: compared with the failure rate of under one percent in the Douglas Point reactor, all the NRU test bundles failed, and in one of these bundles nine of the sixteen outer elements developed defects. We believe that the difference in behaviour is largely due to movement of $\text{UO}_2$ fragments within the sheath during handling of the bundles between their low and high power irradiations. In the NRU tests the bundles are depressurized, loaded into a shielded container, rotated from vertical to horizontal, transported by truck, transferred into a hot cell, moved intermittently for examination, then returned to the loop: in the Douglas Point reactor the bundle is simply translated a short distance horizontally while remaining pressurized, so that the cracked $\text{UO}_2$ fragments have no opportunity to move. Thus the irradiations in the NRU experimental facilities constitute a much more severe test and we have taken advantage of this fact in searching for improvements.

Further experiments showed that the rapid increase in power achieved by the fuel-moving device was not required to produce fuel failures. Bundles that had been returned to high power positions in the NRU loop after low power irradiations and interim examination also failed.

CANDU fuel uses thin Zircaloy sheaths (thickness to radius ratio of 0.06) which are collapsed onto the $\text{UO}_2$ fuel by the pressurized coolant. Although we do not have much experience of $\text{UO}_2$ fuel in thick-walled sheaths under power reactor conditions, the failure of one such bundle ($t/r = 0.09$) as a result of power increases in NRU tests demonstrated that thick-walled fuel, such as is used in Light Water Reactors, is not immune to this type of failure.

Figure 3 — The delay between increasing the fuel’s power and the resulting increase in coolant activity.

The delay between increasing the fuel’s power and any evidence of a sheath failure was noted in the Douglas Point experience. This effect is most strikingly illustrated in Figure 3, which shows the gamma-activity in the loop coolant during one of the tests using the fuel-moving device. Here the delay was about ten hours; in similar tests, with and without the fuel-moving device, the delay has ranged from a few minutes to a few days. Since fuel elements irradiated at constant power exhibit no evidence of incipient sheath failure, the initial penetration of the sheath presumably occurs sometime during the delay period, but exactly when is not established. In some elements that had been subjected to a power increase there was deuterium in the gases collected on puncturing the
elements, indicating that coolant had entered. However, these same gases contained just as much fission xenon and krypton as was collected from intact neighbouring elements and even the original filling gas was present in full measure. Thus the initial sheath penetration caused by the power increase apparently lets water into the element from the coolant, which is generally at a higher pressure. Subsequent sheath deterioration, accelerated by the water within the element, allows the active fission products to escape, signalling a failure.

Search for Fuel with Improved Performance

Several modifications to the reference fuel were suggested to improve its performance, each intended to counter a postulated failure mechanism. Unfortunately, at that time none of the evidence distinguished unambiguously between the various mechanisms. We therefore decided, in Summer 1970, to test empirically any reasonable modification, suspending critical judgement on its associated mechanism. Because of the very low failure rate in the Douglas Point reactor, several hundred bundles for each modification would have been needed to demonstrate a statistically significant improvement in performance. However, the NRU test, being so much more severe, offered a means of screening the many candidates using only a few elements of each.

Figure 4 — A test bundle with demountable elements, with one element being removed.

Some tests used standard fuel bundles incorporating the modification under study; others used specially developed bundles in which any of the sixteen elements in the outer ring could be replaced during interim examination (Figure 4). These "demountable element" bundles permitted comparison of various modifications, usually with controls of the reference design, under truly identical conditions. When a given element failed it could be replaced, allowing the test to continue. This procedure provided a clear distinction between those elements that survived a particular power increase and those that failed, and thereby ranked the various modifications. Altogether, seventeen modifications were selected for testing but some have not yet been fully assessed.

Figure 5 — The irradiation history of 12 standard elements exposed to a power increase. The asterisk (*) indicates evidence of failure.

Control elements were made according to the reference design for Pickering fuel and irradiated as demountable elements in bundles with the potential solutions. Figure 5 illustrates how after irradiation to 2000 MWd/tonne U at element powers of 310 W/cm the bundles were removed for examination and returned for further irradiation, at 550 W/cm. Five hours after reaching full power, activity in the coolant indicated that one or more of the elements had failed. Examination showed that of the twelve control elements, at least two had failed. Since these results confirmed the behaviour of reference bundles exposed to similar treatment, as already noted, all the control elements were replaced by other elements for continuing irradiation of the bundles.

During a power increase the irradiation-damaged sheath is stressed by thermal expansion of the fuel, with some degree of stress concentration occurring over cracks in the UO$_2$ pellets$^{(12)}$. Also, irradiated zirconium alloys are known$^{(13-15)}$ to be susceptible
to stress corrosion cracking at 300°C in an atmosphere of free iodine, which is a major fission product. Although it could be argued that the resultant sheath strains were insufficient to cause failure and that the iodine should be chemically combined with fission product cesium, possible remedies for both mechanisms were investigated.

Laboratory experiments by Baird and Coleman(16) had shown that a thin layer of graphite on the inner surface of the sheath, presumably by its lubricating properties, decreased the strain concentrations in a sheath expanded over simulated fuel fragments. Other laboratory tests, by Wood(17), showed that the graphite layer also protected the Zircaloy against stress corrosion cracking in an iodine atmosphere. In the standard test, with a single major power increase (Figure 6), all eight demountable elements of this type survived, providing the most significant statistics of improved performance in this series of irradiations. Five of these elements that were exposed to a second period of prolonged operation at low power, survived a second power increase.

Preferred Solution

In these comparative tests several of the modifications exhibited a performance under power increases better than that of the reference design. The graphite layer on the inner surface of the sheath was selected for commercial exploitation because:

- it offered the best statistics (all 8 specimens that were tested survived),
- its power had been increased in a single major step while some of the alternatives had experienced intermediate holds that might have made the test less severe,
- it would increase the fuel fabrication costs very little, and
- it had no effect on the fuel’s neutron economy.

The two Canadian fuel fabricators immediately initiated programs to develop production processes for applying the graphite to the sheath. At the same time discussions were held with Ontario Hydro to agree on the additional tests that should be performed to have the modified fuel accepted into CANDU reactors. As a result, one of the elements that had survived the power increase had a small hole (0.5 mm diameter) drilled in its sheath about half-way along its length, and then the deliberately defected element was further irradiated in coolant representative of CANDU reactor conditions at a power output of 570 W/cm. During its 14 day exposure, its activity release was similar to that from reference elements without graphite operating under the same conditions, and subsequent examination revealed no deterioration of the element.

One of the modified fuel elements that had been irradiated without any hole in its sheath was sectioned and examined metallographically. The graphite layer was generally intact but had separated from the sheath in many places. Meanwhile, Campbell(18) had mounted an in-reactor experiment to measure the gas pressure resulting from the UO₂—graphite reaction, should a piece of the graphite layer fall into the hot central region. This experiment deliberately combined some very unfavourable circumstances: a piece of graphite, whose weight exceeded that of the whole layer in a modified element, was placed on the axis of the fuel element, where the temperatures are highest; the O/U ratio of the fuel, 2.015, was higher than that permitted by standard fuel specifications. Even so, no appreciable increase in internal pressure attributable

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Figure 6 — The irradiation history of 8 elements that incorporated a graphite layer on the sheath’s inner surface. Note change in time scale from Figure 5.

As a variant on the previous modification, four demountable elements were prepared with the graphite layer on the cylindrical surface of the UO₂ pellets, instead of on the inner surface of the sheath. All four survived power increases that occurred in steps. This result can be taken as some confirmation of the beneficial effects of a graphite layer between fuel and sheath but is in itself insufficient to endorse this alternative fabrication route for providing the layer.
to the presence of the graphite was observed until a power equivalent to about 550 W/cm in CANDU power reactor conditions was reached.

The experimental irradiations described in this paper showed that a layer of graphite between the UO₂ and the sheath significantly improved the fuel's performance without introducing any new hazard. Four prototype bundles with the graphite layer were produced for irradiation testing in the NRU reactor, and twelve for the Pickering-2 reactor. These commenced their irradiation during the first half of 1972.

The modified UO₂/Zircaloy fuel that contains a thin graphite layer between fuel and sheath has been designated CANLUB fuel. Although the name suggests that lubrication is involved, it is not clear that this is the only function of the graphite, or even the principal one. From a detailed study of all observed fuel failures, and an analysis of their possible causes, Thomas and Bain(19) concluded that both localized stress concentrations in the sheath and aggressive fission products probably contribute to the failures.

Acknowledgements

In a program of this magnitude acknowledgements are much more than a formality, but the number of people that have contributed in one way or another makes it virtually impossible to recognize everyone individually.

Design and construction of the facilities for irradiating and subsequently examining the experimental fuel were due to the appropriate groups at the Chalk River Nuclear Laboratories. Achieving the required operating conditions in the NRU test reactor would have been impossible without the generous and constructive co-operation of the reactor operators, that we have come to take for granted. Members of CRNL’s Reactor Physics Branch provided estimates of the fuel enrichments required to yield the specified power outputs.

Ontario Hydro staff contributed to the analysis of the Douglas Point reactor’s fuel performance, as did staff from the fuel fabricators, Canadian General Electric Company Limited and Westinghouse Canada Limited. The same individuals also assisted greatly in the transfer of technology from a laboratory technique to an utility-accepted commercial process. As a result of this co-operation, a fuel design with improved performance was introduced into a major power reactor within two years of a problem appearing.

During the period in question, direction of the experimental program was successively in the charge of R.D. Page, W.R. Thomas and A.S. Bain of AECL, Chalk River. Each fuel modification was made the responsibility of a sponsor who was asked to press its potential advantages, regardless of his own convictions. All sponsors, and others, contributed ideas and constructive discussion to the program as a whole. Even in retrospect one could not “predict” confidently which modification should prove successful, as each was open to theoretical criticisms not discussed here. Thus the sponsors of the unsuccessful modifications deserve as much credit as the “winners” in what was truly a team effort.

Finally, tribute should be paid to what may, loosely, be described as luck. Probably only in CANDU reactors with their fuel in individually monitored channels and with on-power refuelling would the identification and discharge of failed fuel be possible so rapidly. Without these capabilities the significance of power increases might not have been realized. Indeed, fuel failures in other reactors, tentatively attributed to other causes, may be due to power increases in the fuel. The use of heavy water as coolant in CANDU reactors resulted in the hydrogen absorbed by the sheath after the initial penetration consisting mainly of the deuterium isotope. We were therefore able to avoid the mistake of blaming the failures on internal contamination by hydrogen-containing impurities.
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