WASTE MANAGEMENT ISSUES AND THEIR POTENTIAL IMPACT ON TECHNICAL SPECIFICATIONS OF CANDU FUEL MATERIALS

J.C. TAIT AND L.H. JOHNSON

AECL, Whiteshell Laboratories, Pinawa, Manitoba, R0E 1L0

ABSTRACT

The technical specifications for the composition of nuclear fuels and materials used in Canada's CANDU reactors have been developed by AECL and materials manufacturers, taking into account considerations specific to their manufacture and the effect of minor impurities on fuel behaviour in reactor. Nitrogen and chlorine are examples of UO_2 impurities, however, where there is no technical specification limit. These impurities are present in the source materials or introduced in the fabrication process and are neutron activated to ^{14}C and ^{36}Cl, which after ^{129}I, are the two most significant contributors to dose in safety assessments for the disposal of used fuel. For certain impurities, environmental factors, particularly the safety of the disposal of used fuels, should be taken into consideration when deriving "allowable" impurity limits for nuclear fuel materials.

INTRODUCTION

Safety assessment studies of the disposal of used uranium oxide fuels have shown that a small number of radionuclides dominate the potential environmental consequence. Among the most significant in their potential radiological effects are ^{129}I, ^{36}Cl and ^{14}C [1, 2, 3]. The latter two nuclides are activation products, arising principally from activation of chlorine and nitrogen impurities in both UO_2 pellets and Zircaloy cladding. Restrictions in these impurity levels thus have the potential to reduce the radiological impact associated with fuel disposal.

The technical specifications for the composition of nuclear fuels and materials used in Canada's CANDU reactors have been developed by AECL and materials manufacturers, taking into account current ASTM standard specifications as well as considerations specific to their manufacture and their operation in reactor. In particular, UO_2 pellet, Zircaloy cladding and pressure tube technical specifications place restrictions on the allowable levels of elemental impurities in source materials and finished products. These restrictions have in the past been established as a result of the known detrimental effects of impurities on the manufacture of materials or on their neutronic or materials behaviour in reactor. AECL is currently in the process of reviewing technical specifications with the fuel manufacturers and end users to reflect all current requirements for Canadian CANDU reactor fuel. For certain impurities, environmental factors, particularly the safety of the disposal of used fuels, should be taken into consideration when deriving "allowable" impurity limits for nuclear fuel materials.

RADIONUCLIDES OF SIGNIFICANCE IN SAFETY ASSESSMENT STUDIES

Two safety assessment case studies have been performed illustrating the performance of hypothetical disposal systems for the disposal of used fuel in plutonic rock of the Canadian Shield. [1, 4]. In each case the disposal system comprised an engineered vault (used fuel bundles in long-lived corrosion-resistant containers surrounded by low permeability sealing materials) excavated in rock at a depth of 500 to 1000 m (Figure 1). The most probable exposure scenario is for an undisrupted vault (i.e. no human or natural phenomenon intrusion) and involves eventual corrosion-induced failure of the containers and leaching of the used fuel by groundwater, followed by slow transport of radionuclides in groundwater to the biosphere.

In the Environmental Impact Statement (EIS) case study [1], the engineered system comprised used fuel bundles in Grade-2 Ti containers, which have a lifetime conservatively estimated to be 10^3 to 10^4 years , emplaced in boreholes in the floor of disposal rooms. Once having failed by corrosion, the containers were assumed to offer no further protection to water ingress and release of radionuclides to the vault. The space between the containers and the borehole wall, as well as the rooms and tunnels, were sealed with clay-aggregate mixtures. The rock surrounding the disposal vault had hydrogeological properties derived from measurements from the Whiteshell Research Area [1]. Groundwater velocities were determined to be extremely slow in the low permeability rock at depths of greater than ~400 m, thus the sparsely fractured rock surrounding the vault was the dominant radionuclide transport barrier in the disposal system.

A second case study [3, 4] examined the performance of an engineered system comprising long-lived ($>10^6$ years) Cu containers in an in-room (as opposed to borehole) emplacement configuration. The containers were assumed to fail with a pinhole defect in the container with release of radionuclides through the defect. The two emplacement concepts are illustrated in Figure 1. In the second case study, radionuclide releases arose only from the small fraction of containers (1 in 5000) assumed to have an undetected manufacturing defect. The rock was assumed to be of a considerably higher permeability than in the EIS case study, thus the engineered barrier system had the greatest impact in limiting the radiological consequence.

The Atomic Energy Control Board (AECB) requires that quantitative estimates of the radiological risk be made for times up to 10,000 years following vault closure [5]. The radiological risk is the probability that an individual or his or her descendants will incur a fatal cancer or serious genetic defect because of exposure to radiation. The risk limit is specified to be 1 in 10^6 per year, calculated without taking advantage of long-term institutional controls as a safety feature. Where predicted risks do not peak before 10,000 years, there must be reasoned arguments that at longer times the rate of radionuclide release to the environment will not suddenly and dramatically increase and that acute radiological risks will not be encountered by individuals.

Results from the case study described in the EIS [1] showed that the radionuclides dominating the dose consequence in the undisrupted vault scenario were ^{129}I and ^{14}C. Subsequent studies [2, 3] showed that ^{36}Cl would also be a major dose contributor. In these studies, release from the fuel pellets (rather than the cladding) was responsible for the estimated doses, and it was emphasized that these three radionuclides are of greatest importance for several reasons, including:

- long half-lives (^{129}I, $t_{1/2} = 2 \times 10^7$ a; ^{36}Cl, $t_{1/2} = 3 \times 10^5$ a; ^{14}C, $t_{1/2} = 5730$ a),
- high solubilities, typically five to eight orders of magnitude higher than the long-lived actinides,
- negligible sorption on geological materials because they exist as anions in groundwater, and,
- "instant release" fractions of from 2 to 10% of their inventories, arising from accumulations in the fuel/sheath gap and at fuel grain boundaries.

The radiological dose estimates from the EIS case study [1], including the ^{36}Cl results [2], indicate that the risk is 6 orders of magnitude below the AECB risk limit at 10,000 years for an undisrupted vault, emphasizing the important role played by the low permeability rock surrounding the vault. The dose rate associated with the AECB radiological risk limit is 5×10^{-5} Sv/a [5].

In the second case study [3] ^{129}I and ^{36}Cl were again shown to be the most significant contributors to dose in the time frame up to 10,000 years, with ^{36}Cl contributing about 4% of the total dose. In this case the average dose rate peaked at $\sim 2 \times 10^{-6}$ Sv/a, a value about 25 times below the AECB criterion (Figure 2a). The much larger estimated dose arises from the relatively high permeability rock assumed for the study, which results in contaminant releases to the biosphere within several hundred years.

MEASUREMENTS OF ^{14}C, ^{129}I AND ^{36}CL IN USED CANDU FUELS

Carbon-14.

Carbon-14 is produced in UO_2 fuel pellets by neutron capture reactions involving $^{14}N(n,p)^{14}C$, $^{17}O(n,\alpha)^{14}C$ and $^{13}C(n,\gamma)^{14}C$ with the first reaction being by far the predominant source. Nitrogen is not listed as a regulated impurity in the technical specification for CANDU UO_2 pellets. Early technical specifications for UO_2 pellet impurities called for a maximum concentration of 100 ppm for any individual "unlisted" impurity, and thus this limit was chosen for nitrogen for the safety assessment calculations.

Carbon-14 may be present in used fuel pellets as carbides, oxycarbides or elemental carbon [6], which would potentially be readily available for release upon contact with groundwaters in a disposal vault. Measurements of ^{14}C release from Light Water Reactor (LWR) fuels leached for several hundred days at 25°C in a low-ionic-strength groundwater indicated releases from bare and clad (defected) fuels ranging from 0.05% to 3.5% of the total ^{14}C inventory [7, 8, 9]. Release measurements at 200°C on LWR fuels [10] ranged from 2.4 to 7.4 % of the total ^{14}C inventory.

The release of ^{14}C from used CANDU fuel pellets has been measured for fuels with burnups ranging from 160 MWh/kg U to 327 MWh/kg U and discharged over the time period 1976 to

1984 [11]. Some fuel samples were clad, 2-cm long segments leached in a borate buffer at 25°C for times up to 90 d. Other fuel samples were 2-cm long fuel segments (cladding removed) that were ground to a powder (5 to 20 μm) in a borate buffer solution to expose all fuel grain-boundaries. The borate buffer was used to impose basic leaching conditions so that loss of ^{14}C would be minimized. In addition, some fuel pellets were completely dissolved and the total ^{14}C inventory in the fuel was determined.

The measured ^{14}C release from leaching was compared to the measured total ^{14}C inventories in the pellets to derive the "instant" release fraction. In addition, the leached inventories were compared to estimates of ^{14}C concentration in these fuels using ORIGEN-S code predictions, assuming a 100 ppm nitrogen impurity in the unirradiated UO_2 fuel [12].

The measured ^{14}C inventories in fuel elements from six CANDU fuel bundles were found to be substantially lower than those calculated using the ORIGEN-S code with an assumed 100 ppm N impurity as the primary source for ^{14}C [11]. The calculated/measured ^{14}C ratio for the fuel averaged about 11.5 ± 3.9 and suggests that the actual ^{14}N impurity in the fuel pellets is probably closer to about 10 ppm. Using either the measured total ^{14}C content, or the calculated total ^{14}C content based on a 10 ppm N impurity, the percentage ^{14}C leached from 14 different fuel elements ranged from 0.06% to 5.04% of the total ^{14}C inventory with an average of 2.7 ± 1.6%.

The ^{14}C releases did not show any correlation with burnup or power-rating of the fuel; however, for a given fuel bundle, there was a consistent pattern of higher release with increase in power (i.e. outer ring elements vs. intermediate and inner ring elements) [13]. It was suggested that the lack of correlation may be due to the fact that the impurity level of N (the main contributor to ^{14}C inventories) is unknown and will be variable in the fuels, potentially masking any such correlation.

The total ^{14}C inventories measured for six CANDU fuel pellets (Bq/g UO_2) were a factor of 2 to 4 lower than values reported for LWR fuels, consistent with the difference in burnup of the two fuel types. The release of ^{14}C from clad fuel pellets was measured over periods of 7 and 90 days and the rate of release was extrapolated to estimate the time required to leach the gap and grain-boundary inventory of ^{14}C. For most of the fuels, this inventory would likely be released to solution in several hundred years if it were conservatively assumed to be accessible to leaching.

Iodine-129.

Iodine-129 is a principal product of ^{235}U and ^{239}Pu fission in UO_2 fuels and is near the upper peak of the fission yield curve. The production of ^{129}I will thus be directly correlated with burnup. Since I is volatile and is incompatible with the UO_2 lattice, a fraction of the inventory will migrate at reactor temperatures to the gap and grain-boundary regions of the UO_2 fuel (probably forming CsI or other ionic compounds), where it will remain readily accessible for release to a disposal vault if groundwater contacts the fuel. Several studies have established a strong linear correlation between both the stable Xe fission gas release and the ^{129}I leached from the gap region of the fuel [14, 15] and the fuel linear power. Subsequently, a study was performed to examine the potential impact of the fuel power on the ^{129}I release to solution from the combined gap and grain-boundary regions [13].

Leaching studies were carried out on either 2-cm used fuel segments with cladding attached, or used fuel pellets crushed to a particle size of 5 to 20 µm to provide access to all grain boundaries. The fuel pellets were simultaneously crushed and leached in borate buffer solution to retain all ^{129}I released [13, 16]. The fuel cladding from the crushed fuels was leached separately in 1 mol/L NaOH. The clad fuel segments were leached in deionized water containing KI as a carrier. The total ^{129}I in the fuel was calculated using the ORIGEN-S code, which has been shown to reliably predict fission product inventories in CANDU fuels [17].

For a total of 14 fuel elements examined, the ^{129}I release varied from 1.8% to 11.0% of the total ^{129}I inventory with an average of $3.6 \pm 2.4(\sigma)\%$, considerably lower than the $8.1 \pm 1\%$ used in the safety assessment studies [18, 19].

It appears that for high power fuels (>44 kW/m), most of the ^{129}I is released to solution in the first few days of leaching and the rate then slows to a value comparable to that from low power fuels (<42 kW/m). Based on the ^{129}I release rates, the combined gap + grain-boundary ^{129}I inventory could be released in a period of about 20 to 30 years for high power fuels, and in 360 to 460 years for low-power fuels [16].

Chlorine-36.

It has recently become apparent that ^{36}Cl, produced by neutron activation of naturally abundant ^{35}Cl impurities in UO_2 pellets and Zircaloy, can be a significant contributor to the total radiological risk in the disposal of used fuel [2]. It was originally anticipated that, due to the high fabrication temperatures of both UO_2 pellets and Zircaloy cladding and the volatility of Cl, that its residual concentration would be negligible. Recent analyses on Zr/2.5Nb pressure tubes [20], however, indicated that Cl impurity levels were typically 1 to 5 ppm (µg/g Zr). This led to speculation that Cl impurities may also be present in the Zircaloy cladding and in the UO_2 fuel pellets at similar levels.

To address this concern, the release of ^{36}Cl and total Cl impurity concentration in used fuel pellets and unirradiated UO_2 pellets have been measured [21]. Used CANDU fuel pellets were analyzed for total ^{36}Cl content by dissolving in nitric acid, precipitating the chloride as AgCl, and analyzing the precipitate for ^{36}Cl using Accelerator Mass Spectrometry (AMS). Total Cl impurities were determined for several unirradiated UO_2 pellets by pyrohydrolytic extraction (thermal decomposition), collection of the evolved gases and analysis by ion chromatography. The leaching behaviour of ^{36}Cl was studied by leaching used fuel pellets in DIW or in a borate buffer solution, precipitating Cl as AgCl and analyzing for ^{36}Cl by AMS.

Analysis of four ceramic powder samples of UO_2 used in the manufacture of Canadian CANDU fuel pellets and a sintered unirradiated pellet showed Cl impurity levels ranging from 1.6 to 3 ppm, with an average impurity level of 2.3 ± 1.1 ppm [21]. The Cl impurity concentration in five irradiated CANDU fuels was derived from the total ^{36}Cl content and

ORIGEN-S code calculations on a fuel with a burnup of 190 MWh/kg U with an assumed 5 ppm Cl impurity. Since ^{36}Cl activation will be directly proportional to burnup, the total Cl impurity in each fuel could be calculated. The derived Cl impurity levels ranged from 0.3 to 3.4 ppm with an average of 1.5 ± 1.1 ppm. Measurements of the release of ^{36}Cl indicated that a significant fraction of the total ^{36}Cl is readily accessible to leaching ("instant" release fraction) when contacted by aqueous solution.

Aqueous leaching measurements indicated that from 0.5 to 20% of the total ^{36}Cl inventory could be released to solution, and that release was directly correlated with both fuel burnup and the stable Xe gas release (and thus fuel power) [21]. For a moderate power and average burnup CANDU fuel, the average ^{36}Cl release to solution would be about 2%. However, for fuels with high linear powers, the release increases substantially. For a fuel with an average fission gas release of 2.2% (based on an analysis of fuel power histories [22]) the average ^{36}Cl "instant" release (i.e. the readily accessible gap inventory) would be about 9%. It appears that the "instant" release ^{36}Cl fraction is present at fuel pellet surfaces and cracks in the UO_2 fuel (gap region) rather than at grain-boundaries. It is likely that since Cl is initially present as an impurity in the UO_2 fuel, segregation of Cl to the gap region at fuel operating temperatures will occur throughout the in-reactor period. This is in contrast to fission products such as I, that diffuse to grain boundaries and the gap region only after they are formed by fission. This likely accounts for the higher percent total release to solution for ^{36}Cl compared to ^{129}I.

POTENTIAL IMPACT OF IMPURITIES ON UO_2 FUEL TECHNICAL SPECIFICATIONS

As previously mentioned, the three radionuclides that are the greatest potential contributors to total radiological dose in environmental assessments of the disposal of used fuel [1, 2, 3] are $^{129}I > ^{36}Cl > ^{14}C$ (Figure 2a). The production of ^{129}I is controlled by the burnup of the fuel and in most cases will provide an upper bound to the risk. The quantity of ^{129}I that would be "instantly released" from UO_2 pellets on contact with groundwaters in a disposal vault can be predicted from the fuel burnup and linear power rating (LPR) for fuels with peak LPR's <47 kW/m. For high power fuels (>48 kW/m peak LPR), the release appears to be much less than that predicted from a linear extrapolation of the low-power fuel behaviour [16]. This would imply that the ^{129}I released from high LPR fuels would not be much greater than that from an average LPR fuel.

The developing trend with future CANDU fuels is to higher burnup, higher power fuels (>48 kW/m peak) where ^{129}I production would increase proportionately with burnup, but where its "instant release" fraction to groundwaters may be approximately the same as that from the highest low-power fuels (<47 kW/m peak). Although this is positive from a disposal aspect, the other contributors to dose (^{14}C and ^{36}Cl) may become increasingly important at high burnup and power, as they are continuously produced from the initial impurity concentration of ^{14}N and ^{35}Cl.

Carbon-14.

Carbon-14 is the third highest contributor to dose in the EIS case study, which used an initial N impurity of 100 ppm for the reference fuel (burnup of 190 MWh/kg U) [1, 23]. As described above, measurements of total ^{14}C content in seven irradiated fuels [11] established that impurity concentrations of N in the fuel were approximately 10 ppm. The second case study postclosure assessment [3, 19] ^{14}C concentrations were derived using the 10 ppm N impurity concentration. This study also indicated that for nuclides whose production is not derived from fission products, ^{14}C continues to be the third largest contributor to dose (Figure 2a).

If N impurity concentrations were indeed typically near the 100 ppm limit assumed in the EIS case study, the dose contribution from ^{14}C would be similar to that from ^{36}Cl (Figure 2a). A strategy may thus be required to assist in the selection of more appropriate impurity limits for N in UO_2 powders or sintered pellets.

The final concentration of nitrogen impurities in the sintered UO_2 pellets will depend strongly on the sintering parameters and the behaviour of N during sintering is not known at present. A strategy for providing a database on the behaviour of N impurities during fabrication would involve analysis of both ceramic powdered natural UO_2 and as-sintered pellets from the manufacturer to develop a history of the behaviour of nitrogen during the sintering process. It is possible that nitrogen impurity limits for ceramic powdered UO_2 could be set higher than those for the sintered product if significant losses occur during the sintering process.

Chlorine-36

As described above, Cl impurity concentrations in used UO_2 fuel pellets and in unirradiated fuel pellets and ceramic powder source materials, have been shown to be less than about 5 ppm. Radiological risk assessments of the impact of ^{36}Cl on the disposal of used fuel [2, 3] indicate that the contribution from ^{36}Cl arising from a reference fuel burnup of 190 MWh/kg U with a 5 ppm initial Cl impurity, would be about 4% of the total dose which is dominated by ^{129}I. The second case study [3] also identified individual simulations where the dose from ^{36}Cl was as high as that from the average ^{129}I dose; this was due to the variability and uncertainty in the parameters chosen for that particular simulation.

Another factor that could lead to high doses from ^{36}Cl is the difference in production pathway for ^{36}Cl compared to ^{129}I and the release behaviour of ^{36}Cl to solution with burnup and fuel power. Chlorine, unlike ^{129}I, is present in the fuel pellets from the beginning of irradiation. As a result, Cl will migrate to UO_2 pellet surfaces and cracks throughout the reactor operational period and the fraction of activated ^{36}Cl available for "instant release" to groundwater will increase with burnup. In addition, higher fuel temperatures (higher powers) will promote migration, also leading to greater ^{36}Cl concentrations at UO_2 surfaces. Thus the release of ^{36}Cl to solution will increase with both fuel burnup and power, in contrast to ^{129}I whose concentration would increase with burnup, but whose release to solution has been shown to be less strongly dependent on power history [16]. This suggests that the radiological risk from ^{36}Cl released to the vault could approach that of ^{129}I as both fuel burnup and fuel power increase. This factor should be considered when selecting appropriate technical specification impurity limits for fuel pellets, particularly when higher burnup/power fuels or enriched fuel designs are proposed for CANDU reactors or if the fraction of fuels discharged with high burnups/powers from current reactors increases significantly.

Another consideration for chlorine impurities is the relatively large Equivalent Boron Content (EBC) factor for Cl which could also play a role in restricting the allowable impurity concentration in fuel pellets. Chlorine has a high cross-section (33.5 barn), about 5% of that for B (764 barn) and higher than many other specified "allowable" impurities in UO_2 pellets.

Currently, there are no technical specifications for Cl impurity concentrations in CANDU fuels (either for powders or sintered pellets). Early AECL technical specifications quoted a limit of 100 ppm for an individual "unlisted" impurity element; however, current specifications do not include "unlisted" elemental limits. Our measurements on Cl impurities in used fuels suggest that the current Canadian manufacturing process typically produces fuels with Cl impurity concentrations less than about 5 ppm [21]. In deriving technical specifications for Cl in both ceramic UO_2 powders and pellets, consideration should be given to establishing a Cl impurity limit at a reasonable but conservative value. The safety assessment case performed for a 5 ppm Cl concentration in the UO_2 fuel has indicated that Cl is the second most important radionuclide contributing to dose and that with higher burnups, its importance could approach that of ^{129}I. If Cl impurity limit concentrations significantly higher than 5 ppm were adopted for the sintered product (and concentrations of Cl were actually present at these limits), the radiological risk would be expected to increase proportionately. For example, if Cl impurity levels were typically 50 ppm, the predicted radiological dose rate for fuel disposal, using the results from the second case study, are estimated as shown in Figure 2b. Were the 50 ppm value to be used to bound the ^{36}Cl dose in a safety assessment, the total dose from all nuclides would increase by about 30% relative to that for a Cl impurity concentration of 5 ppm. From a disposal perspective, it would be appropriate to adopt a specification such that, even if Cl concentrations were typically 50% of the specification, the total risk from all radionuclides would not be increased significantly.

As with N, the final concentration of chlorine impurities in the sintered pellets will depend strongly on the sintering parameters and this behaviour is not known at present. A strategy for providing a database on Cl impurities in powders and pellets similar to that discussed above for N would allow a history of the behaviour of Cl during the sintering process to be developed. It is possible that Cl impurity limits for powdered UO_2 could be set higher than those for the sintered product if significant losses occur during the sintering process.

Other Potential Impurities

The potential impact to the safety of used fuel disposal of the presence of about 80 light element impurities (elements lighter than Pb) in CANDU fuel and the Zircaloy cladding has been reviewed [24]. Other than N and Cl impurities, there were no other impurity elements identified that would significantly impact on the safety assessment for disposal of these materials at reasonable impurity concentrations. For example, potential fuel impurities, such as S and P would produce ^{36}Cl as a result of multiple neutron activation pathways; however, due to their relatively low cross-sections for activation and the multiple neutron events required, the efficiency of production of ^{36}Cl is orders of magnitude smaller than that arising from direct activation from ^{35}Cl impurities [24]. Sulphur as an impurity, however, can have a deleterious effect on the sintering of UO_2 [25] at impurity levels above about 30 ppm.

DISCUSSION AND CONCLUSIONS

The presence of specific impurities in UO_2 fuel pellets can be used to illustrate the necessity to consider the impact of impurities on both fuel irradiation behaviour (fuel neutronics and/or physical and manufacturing characteristics) and radiological and environmental consequences (permanent disposal) when reviewing and selecting impurity limits for technical specifications for fuel pellets and reactor materials. For example, in the case of N or Cl impurities, if fuel irradiation behaviour considerations were considered in isolation, a N or Cl impurity concentration of 100 ppm in UO_2 might be acceptable as it may not affect fuel fabrication or irradiation characteristics; however when environmental consequences are considered, it may be necessary to establish much lower impurity limits.

Conversely, in the case of S, for example, if environmental impacts were considered in isolation, it might be concluded that hundreds of ppm S could be tolerated in the UO_2 fuel pellets before any radiological consequences of significance would arise from the production of ^{36}Cl. However, when taking into consideration physical consequences (grain growth etc.), it would likely be necessary to limit S impurity content in UO_2 pellets to below about 30 ppm [24, 25].

In deriving impurity limits for fuel pellets and bundle/reactor components, a strategy should be applied that considers the potential impact of impurities on all the important factors such as neutronics, fuel behaviour (density, gas release, grain growth, integrity) and radiological consequences of disposal. This may require establishing weighting factors and deriving an evaluation strategy that would allow the most important of these factors to drive the selection of impurity limits. This is likely to become more important in the future, given the trend to higher fuel burnups for CANDU fuels and the development of high burnup advanced fuels for MOX fuel cycles.

In addition, it may be desirable to strengthen the historical database of the behaviour of UO_2 and cladding impurities, such as N and Cl during the fabrication process. Knowledge of typical impurity concentrations in the source materials, for example, would help to establish how their concentrations are affected during the sintering/refining process. If impurity limits were established for the source materials, and their behaviour during processing was well defined through process controls, it could ensure allowable impurity limits would be met in the final product with minimum qualification, and that fuel manufacturing and neutronic factors would be satisfied.

There are both environmental and cost benefits to setting Cl and N impurity specifications at a sufficiently low level that doses from ^{36}Cl and ^{14}C would easily meet regulatory criteria for fuel disposal. These doses should typically be a small fraction of the dose from ^{129}I, normally the most important dose contributor.

The environmental perspective is illustrated in Figure 2, which shows that for the case study describing in-room emplacement of long-lived copper containers of used fuel, the total radiological dose is increased by about 30% if the chlorine level is assumed to be 50 ppm rather than 5 ppm. Additional analyses have also been performed [26], based on a simple model that assumes releases from a pinhole failure in a single container where contaminants are transferred directly to a well used for drinking water and irrigation (i.e., no credit is given for dispersion in the geosphere). In this case doses were less than the *de minimis* dose rate of 50 $\mu Sv \cdot a^{-1}$ (i.e. a dose rate so small as to not warrant institutional control), adopted by the Atomic Energy Control Board [27]. Similarly, were Cl to be present in the fuel at the 50 ppm level, the dose from Cl would be about 30% of the total dose of 1.5 $\mu Sv \cdot a^{-1}$. In the same study [26], it was noted that if well water was used for crop irrigation, the total dose is dominated by ^{14}C and would be 29 $\mu Sv \cdot a^{-1}$ based on an assumed N impurity level of 10 ppm. Clearly, if the N impurity content in UO_2 was an order of magnitude higher, the *de minimis* dose rate would be significantly exceeded for this scenario.

From a cost perspective, appropriately selected specifications for N and Cl would preclude the need for further characterization of fuel for disposal from the perspective of ^{14}C and ^{36}Cl release. A selective characterization of historical fuels has already established typical N and Cl impurity levels in used CANDU fuels. If appropriate N and Cl impurity levels were designated that would satisfy current assessment requirements these limits could be used in future assessments without the need for further analytical verification. Because Canadian CANDU fuel manufacturers appear to produce UO_2 fuel with typically <5 ppm Cl and <10 ppm N, establishing specifications at several times these values is unlikely to have cost implications for fuel manufacturing.

ACKNOWLEDGEMENTS

The authors wish to thank K. Dormuth, R. Sejnoha, P.K. Chan and F.P. Sargent for their review of the manuscript and helpful comments. We also wish to acknowledge funding for a portion of this work from COG Working Party 9.

REFERENCES

1. AECL (Atomic Energy of Canada Limited). "Environmental Impact statement on the concept for disposal of Canada's nuclear fuel waste". Atomic Energy of Canada Limited Report, AECL-10711, COG-93-1 (1994).
2. JOHNSON, L.H., B.W. GOODWIN, S.C. SHEPPARD, J.C. TAIT, D.M. WUSCHKE AND C.C. DAVISON. "Radiological assessment of ^{36}Cl in the disposal of used CANDU fuel". AECL-11213, COG-94-527 (1995).
3. GOODWIN, B.W., T.H. ANDRES, W.C. HAJAS, D.M. LeNEVEU, T.W. MELNYK, J.G. SZEKELY, A.G. WIKJORD ET AL. "The disposal of Canada's nuclear fuel waste: A study of postclosure safety of in-room emplacement of used CANDU fuel in copper containers in permeable plutonic rock. Volume 5: Radiological assessment". Atomic Energy of Canada Limited Report. AECL-11494-5, COG-95-552-5 (1996).
4. WIKJORD, A.G., P. BAUMGARTNER, L.H. JOHNSON, F.W. STANCHELL, R. ZACH AND B.W. GOODWIN. "The disposal of Canada's nuclear fuel waste: A study of postclosure safety of in-room emplacement of used CANDU fuel in copper containers in permeable plutonic rock". Volume 1: Summary. Atomic Energy of Canada Limited Report. AECL-11494-1, COG-95-552-1 (1996).
5. AECB (Atomic Energy Control Board). "Regulatory policy statement. Regulatory objectives, requirements and guidelines for the disposal of radioactive wastes - Long-term aspects". AECB-R-104, June 5, Ottawa. (1987).
6. VAN KONYNENBURG, R.A., C.F. SMITH, H.W. CULHAM AND H.D. SMITH. "Carbon-14 in waste packages for spent fuel in a tuff repository". In Scientific Basis for Nuclear Waste Management X, (J.D. Bates and W.B. Seefeldt, Editors), Mat. Res. Soc. Symp. Proc., Boston, USA, 1986. 84, 185-196 (1987).
7. WILSON, C.N. and H.F. SHAW. "Experimental study of the dissolution of spent fuel at 85°C in natural groundwater". In Scientific Basis for Nuclear Waste Management X, (J.D. Bates and W.B. Seefeldt, Editors), Mat. Res. Soc. Symp. Proc., Boston, USA, 1986. 84, 123-130 (1987).

8. WILSON, C.N. "Results from NNWSI series 2 bare fuel dissolution tests". Report PNL-7169, Pacific Northwest Laboratories, Richland WA (1990).
9. WILSON, C.N. "Results from Nevada Nuclear Waste Storage Investigation (NNWSI series 3 bare fuel dissolution tests"., Pacific Northwest Laboratories Report PNL-7170, Richland WA (1990).
10. NEAL, W.I., S.A. RAWSON and W.M. MURPHY. "Radionuclide release behaviour of light water reactor spent fuel under hydrothermal conditions". In Scientific Basis for Nuclear Waste Management XI, (M.J. Apted and R.E. Westerman, Editors), Mat. Res. Soc. Symp. Proc., Boston, USA, 1987. 112, 505-515 (1988).
11. STROES-GASCOYNE, S., J.C. TAIT, R.J. PORTH, J.L. McCONNELL AND W.J. LINCOLN. "Release of ^{14}C from the gap and grain-boundary regions of used CANDU fuels to aqueous solution." Waste Management, 14:5, 385-392 (1994).
12. TAIT, J.C., I.C. GAULD AND G.B. WILKIN. "Derivation of initial radionuclide inventories for the safety assessment of the disposal of used CANDU fuel". AECL-9881 (1989).
13. STROES-GASCOYNE, S. "Measurements of instant-release source terms for ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{129}I, and ^{14}C in used CANDU fuels." J. Nucl. Mater. 238, 264-277 (1996).
14. STROES-GASCOYNE, S., L.H. JOHNSON AND D.M. SELLINGER. "The relationship between gap inventories of stable xenon, ^{137}Cs and ^{129}I in used CANDU fuel". Nuclear Technology, 77, 320-330 (1987).
15. STROES-GASCOYNE, S., AND D.M. SELLINGER. "The effect of fuel power on the leaching of cesium and iodine from used CANDU fuel". Proc. 1st Int. Conf. on CANDU Fuel, Chalk River, (editor I.J. Hastings) CNS Toronto, 383-400 (1986).
16. STROES-GASCOYNE, S., D.L. MOIR, M. KOLAR, R.J. PORTH, J.L. McCONNELL AND A.H. KERR. "Measurement of gap and grain-boundary inventories of ^{129}I in used CANDU fuels". In Scientific Basis for Nuclear Waste Management XVIII, (T. Murakami and R.C. Ewing, Editors), Mat. Res. Soc. Symp. Proc., Kyoto, Japan, 1994. 353, 625-631 (1995).
17. TAIT, J.C. I.C. GAULD AND A.H. KERR.. "Validation of the ORIGEN-S code for predicting radionuclide inventories in used CANDU fuel". J. Nucl. Materials, 223, 109-121. (1995).
18. JOHNSON, L.H., D.M. LENEVEU, D.W. SHOESMITH, D.W. OSCARSON, M.N. GRAY, R.J. LEMIRE AND N.C. GARISTO. "The disposal of Canada's nuclear fuel waste: the vault model for postclosure assessment". Atomic Energy of Canada Limited Report. AECL-10714, COG-93-4 (1994).
19. JOHNSON, L.H., D.M. LENEVEU, F. KING, D.W. SHOESMITH, M. KOLAR, D.W. OSCARSON, S. SUNDER, C. ONOFREI AND J.L. CROSTHWAITE. "The disposal of Canada's nuclear fuel waste: A study of postclosure safety of in-room emplacement of used CANDU fuel in copper containers in permeable plutonic rock. Volume 2: Vault Model". AECL-11494-2, COG-95-552-2 (1996).
20. AITCHISON, I, AND P.H. DAVIES. "Role of microsegregation in fracture of cold-worked Zr-2.5Nb pressure tubes". J. Nucl. Mater. 203, 206-220 (1993).
21. TAIT, J.C., R.J.J. CORNETT, L.A. CHANT, J. JIROVEC, J. McCONNELL AND D.L. WILKIN. "Determination of Cl impurities and ^{36}Cl instant release from used CANDU fuels". In Scientific Basis for Nuclear Waste Management XX, (I. Triay and W. Gray, Editors), Mat. Res. Soc. Symp. Proc., Boston, USA, 1996. MRS, Pittsburgh Pennsylvania, (in press) (1997).
22. JOHNSON, L.H. AND H.H. JOLING. "Fission product leaching from used CANDU fuel: An estimate of fuel-sheath gap and grain boundary inventories and probable releases after disposal". AECL, TR-00280 (1984).
23. GOODWIN, B.W., D.B. McCONNELL, T.H. ANDRES, W.C. HAJAS, E.M. LENEVEU, T.W. MELNYK ET AL. "The disposal of Canada's nuclear fuel waste: Postclosure assessment of a reference system". Atomic Energy of Canada Limited Report, AECL-10717, COG-93-7 (1994).
24. TAIT, J.C. AND J.R. THEAKER. "Light element radionuclides in used CANDU fuel and their potential contribution to postclosure safety assessment". AECL-11431, COG-95-440 (1996).
25. HOARE, M.R. "UO_2 pellet manufacture for CANDU fuel". Proc. International conference on CANDU fuel. Chalk River Laboratories, Chalk River, Ont. Oct. 6-8. (I.J. Hastings ed.) (1986).
26. AMIRO, B.D. AND K.W. DORMUTH. "A simple analysis of potential radiological exposure from geological disposal of Canada's nuclear fuel waste. Atomic Energy of Canada Limited Report. AECL-11533, COG-96-13.
27. AECB (Atomic Energy Control Board) "Radiation protection requisites for the exemption of certain radioactive materials from further licensing upon transferral for disposal. Atomic Energy Control Board Regulatory Document R-85, Ottawa, ON. (1989).

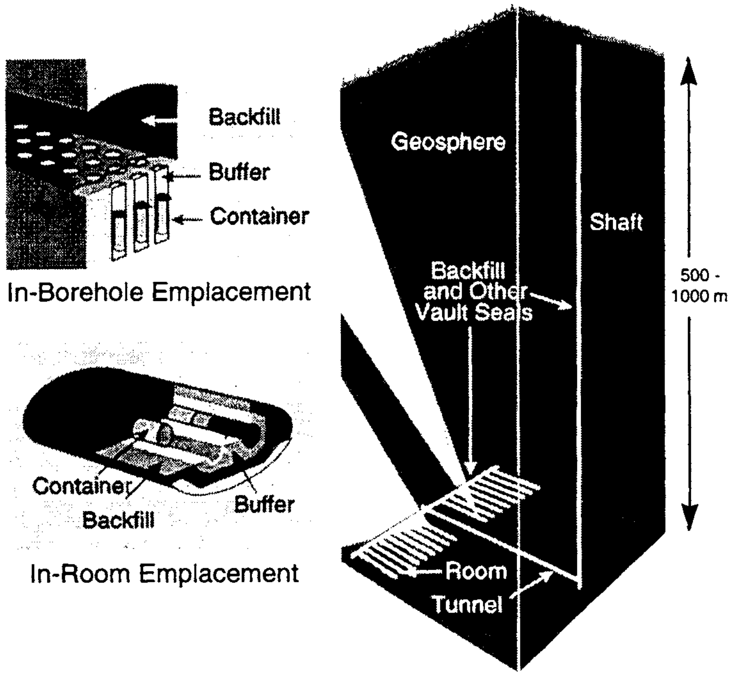
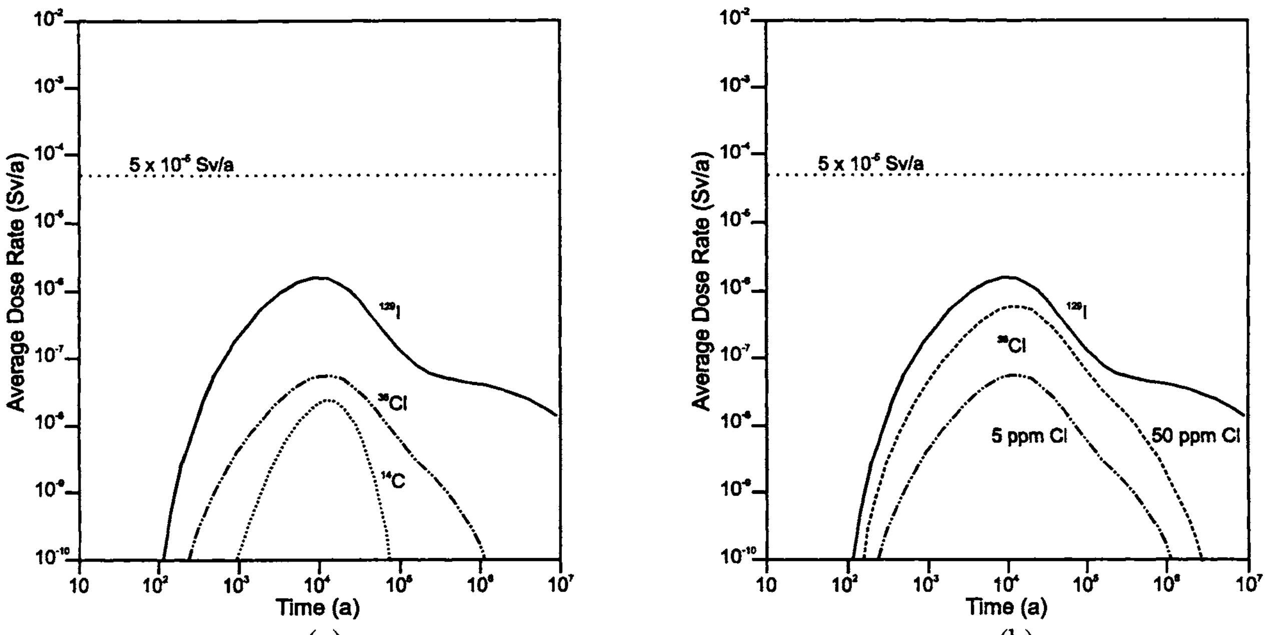


FIGURE 1. LAYOUT OF THE CONCEPTUAL DISPOSAL VAULT FOR USED FUEL SHOWING THE BOREHOLE AND IN-ROOM EMPLACEMENT CONCEPTS.



(a) (b)

FIGURE 2. (a) CONTRIBUTIONS TO AVERAGE DOSE RATE FROM ^{14}C, ^{36}Cl AND ^{129}I IN THE MEDIAN-VALUE SIMULATION. THE HORIZONTAL LINE REPRESENTS THE AECB CRITERION FOR ACCEPTABILITY [27]. (b) COMPARATIVE DOSE RATES FOR USED FUEL CONTAINING AN INITIAL 5PPM AND 50 PPM Cl IMPURITY (FIGURES ADAPTED FROM GOODWIN ET AL. 1996).