High Polymer Composites for Containers for the Long-Term Storage of Spent Nuclear Fuel and High Level Radioactive Waste

H.W. Bonin, V.T. Bui, J.-F. Legault,
A.R. Davey, T.D. Douglas, C.C.S. Duriez and G.B. MacGregor

Department of Chemistry and Chemical Engineering
Royal Military College of Canada / Collège militaire royal du Canada
P.O. Box 17000; Station FORCES
Kingston, Ontario Canada K7K 7B4

ABSTRACT

The feasibility of using polymeric composite materials as an alternative to metals in the design of a nuclear waste disposal container was examined. The disposal containers would be stored in deep underground vaults in plutonic rock formations within the Canadian Shield for several thousands of years. The conditions of disposal considered in the evaluation of the polymeric composite materials were based on the long-term disposal concept proposed by Atomic Energy of Canada Limited. Four different composites were considered for this work, all based on boron fibre as reinforcing material, imbedded in polymeric matrices made of polystyrene (PS), polymethyl methacrylate (PMMA), Devcon 10210 epoxy, and polyetheretherketone (PEEK).

Both PS and PMMA were determined as unsuitable for use in the fabrication of the storage container because of thermal failure. This was determined following thermal analysis of the materials in which heat transfer calculations yielded the temperature of the container wall and of the surroundings resulting from the heat generated by the spent nuclear fuel stored inside the container. In the case of the PS, the temperature of the container, the buffer and the backfill would exceed the 100 °C imposed in the AECL's proposal as the maximum allowable. In the case of the PMMA, the 100°C temperature is too close to the glass transition temperature of this material (105°C) and would cause structural degradation of the container wall. The other two materials present acceptable thermal characteristics for this application.

An important concern for polymeric materials in such use is their resistance to radiations. The Devcon 10210 epoxy has been the object of research at the Royal Military College in the past years and fair, but limited, resistance to both neutrons and gamma radiation has been demonstrated, with the evidence of increased mechanical strength when subjected to moderate doses. Provided that the container wall could be sufficiently shielded from the radiations emitted by the spent nuclear fuel or other high level radioactive waste, this material may well be an interesting candidate for this application. More recent work at RMC on the effects of radiations on PEEK has demonstrated that this high polymer thermoplastic material was even superior to epoxies under radiation environments. Part of this research concentrated on the estimation of the doses accumulated in the container wall over the years using three basic models for the container: one without filling material, one with glass beads as proposed by AECL, and one using thorium dioxide (ThO_2) as filling material. This choice is based on the excellent physical and chemical properties of this compound (resistance to corrosion in particular) and to the expected low cost since thorium is usually discarded in the tailings of uranium mine concentrating plants. The dose calculations were carried out using the Microshield™ software and showed that both the epoxy and the PEEK could maintain structural integrity provided that they are shielded sufficiently against the radiations emitted by the high level radioactive waste. This research investigated also the resistance to the mechanical forces to which the container walls would be submitted in the underground vaults and it was concluded that these materials displayed sufficient mechanical strength for such application. It also permitted the identification of several aspects of the design of the storage containers that need closer investigation.

INTRODUCTION

High level waste generated by the nuclear industry in Canada is mainly composed of spent CANDU nuclear reactor fuel and solidified high level waste (HLW). The latter comes for the most part in the form of discarded radioactive sources and activation products, including, in a minor way, a small number of spent nuclear reactor fuel elements treated or reprocessed for research purposes. The question of the ultimate disposal of highly radioactive material is currently the object of intense debate. Canada started a multi-year research and development programme several years ago, which has resulted in a proposal [1-6] by Atomic Energy of Canada Limited (AECL) to store indefinitely the high level waste in deep underground vaults carved in plutonic granite rock formations within the Canadian Shield (Figure 1). The proposal relies essentially on a conservative approach mostly based on proven technology, although some aspects involve advanced materials such as the titanium alloy proposed for the storage container, and is presently subjected to the public scrutiny throughout Canada.

STORAGE CONTAINER

The fabrication of the spent fuel/high level waste storage container is the object of this particular engineering research project which evaluates the potential use of high-performance or advanced polymer composites. These materials offer excellent resistance to corrosion in addition to their light weight and superior mechanical strength [7-20]. For practical purposes, this research considers spent CANDU fuel bundles as the HLW, but it is understood that application to other types of waste would be straightforward. A typical container, as shown in Figure 2 would store 72 CANDU bundles and, once fully loaded and hermetically sealed, it would be placed in a cavity dug in the floor of an underground disposal vault or in a chamber carved in the wall of the vault located at some 500-1000 m deep. The vault would be connected to other vaults by a network of tunnels bored deep in a plutonic rock formation within the Canadian Shield. The choice of a such rock formation comes from the fact that plutons are homogeneous granite monoliths that have "survived" intact (or with little damage) the numerous earthquakes and other geologic movements of the earth's crust that have occurred in the last billions of years. Their homogeneity also limits greatly underground water movement and seeping.

Once inside the cavity or the chamber in the disposal vault, the container would be surrounded by buffer material typically containing clay and intended to limit corrosion rate of the container material and the rate of dissolution of the waste form should ground water manage to seep into the container. Furthermore, the buffer material would prevent or retard the movement of the contaminants in the event that they are released from the container. The chamber (or the cavity) , the disposal vault and, in turn, the access tunnels would then be filled with backfill material made of cement, clay or concrete. The function of this is again to prevent or retard the movement of any contamination escaping through the buffer layer, and to firmly secure in place the containers and the surrounding buffers. Once all the storage sites within the disposal facility are occupied, all of the remaining tunnels and access shafts and other boreholes would be definitely sealed with clay-based or cement-based materials intended not only to prevent or retard the migration of radioactive contaminants toward the surface, but also to prevent access to the radioactive waste by humans or animals. The concept is therefore based on the multi-barrier approach adopted by AECL to protect the biosphere from the radioactive contamination: nuclear fuel matrix and fuel cladding, filler material (glass beads are proposed by AECL), container wall, buffer material, backfill material and 500-1000 m thick plutonic granite layer.

MATERIALS PROPOSED FOR THE CONTAINER WALLS AND FILLERS

This research focused on evaluating the feasibility of using high polymer composite materials for the fabrication of the storage container wall. The AECL design was retained as for the dimensions and capacity of the container: 2.246 m height, 0.633 m diameter and 6.35×10^{-3} m thickness, for a 72 CANDU fuel bundle capacity. Four polymeric composites were examined: polystyrene (PS), polymethyl methacrylate (PMMA), Devcon #10210 Epoxy, and polyetheretherketone (PEEK). For all these materials, the reinforcing material was boron fibres in either a 50% (mass) or a 70% (mass) mixture. Several characteristics of these composite materials such as mechanical strength, resistance to corrosion, heat transfer properties and resistance to radiation were investigated and the performance of these materials was analysed and compared with the titanium alloy proposed by AECL and with copper [6] (proposed by the Chemical Institute of Canada [21]).

Polystyrene (PS) is an amorphous, clear, brittle polymer that can be injection molded extruded, filament wound and blown to produce containers. Below its 100°C glass transition temperature, it possesses considerable mechanical strength and a high resistance to water and several other chemical agents. Polymethyl methacrylate (PMMA) is also used in extrusions, molding and as protective coatings and adhesives. It displays an excellent resistance to corrosion and only concentrated acids or alkalies can attack this polymer. Its glass transition temperature is only 105°C and may retain its glass state properties if used as a radwaste container. Again, the mechanical strength of this polymer is

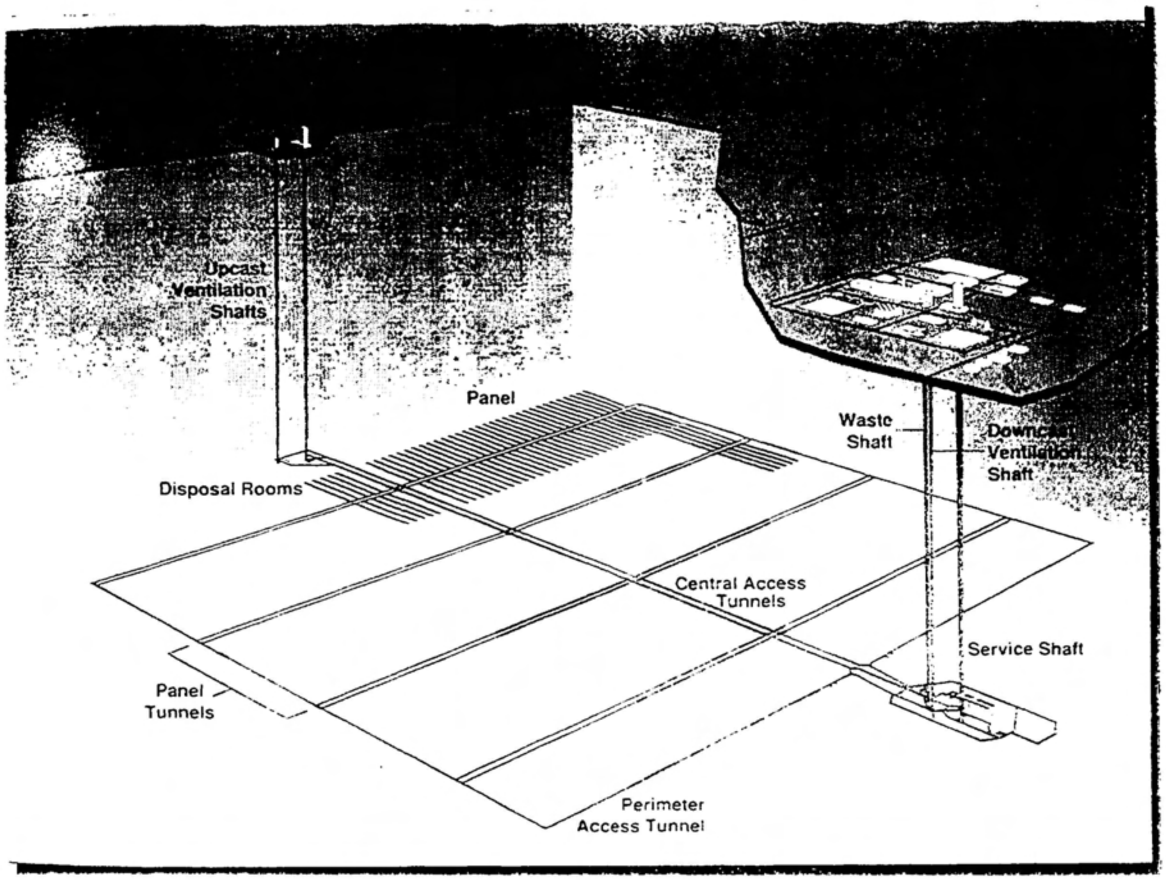

Figure 1: Deep Underground High Level Radioactive Waste Disposal [3].

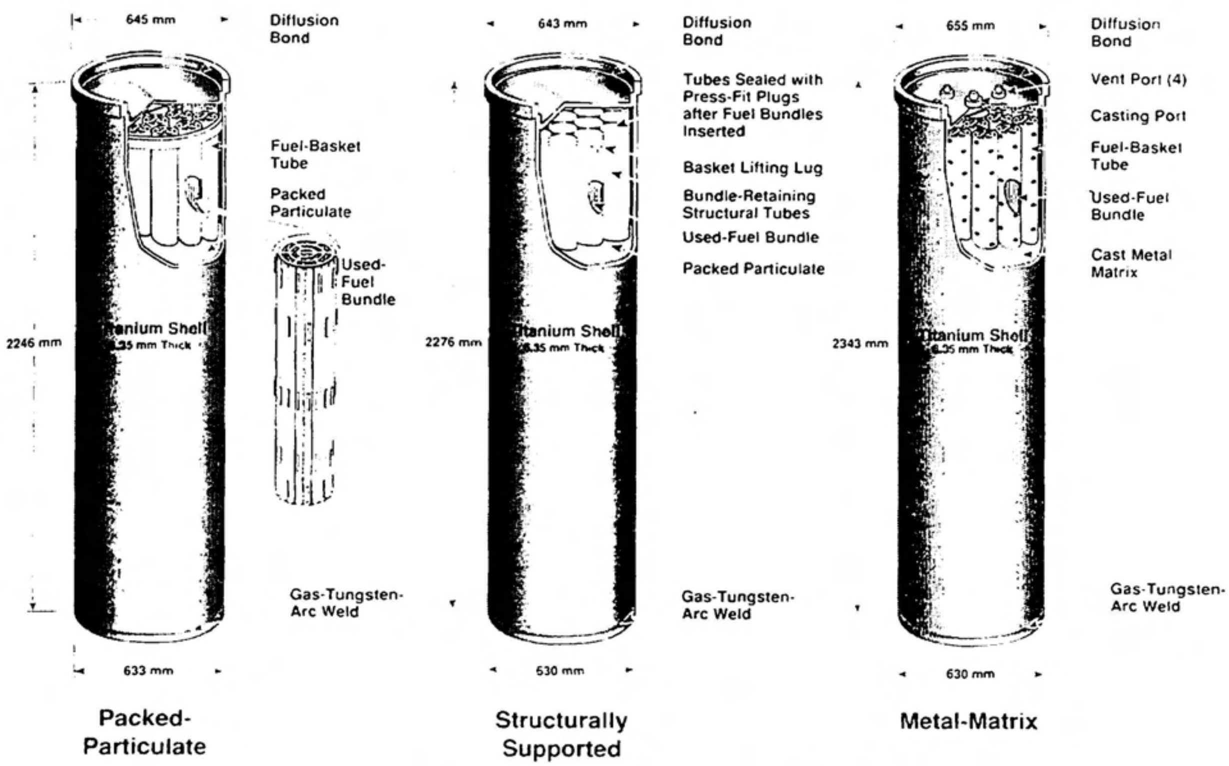

Figure 2: Spent Fuel Storage Container [3].

quite high and it was included in this study, like the PS, because it was readily available and for knowing more on its behaviour under radiations.

Epoxies have been investigated at RMC-CMR since several years [22-24] and they have displayed good resistance to radiations, even to the point of actual strengthening when subjected to moderate doses of neutrons and gammas. These high polymer compounds are thermosetting resins that cure when heat is applied (often resulting from chemical reactions from the hardening agent) and form highly cross-linked, insoluble and infusible matrix resins. They display very high chemical stability and resistance to corrosive agents in addition to outstanding adhesive and mechanical strength. There is a wide variety of epoxies available (several taylor-made) for specific applications, and using a wide range of curing agents such that the setting time varies from a few minutes to 24 hours or more. The Devcon 10210 epoxy chosen for this work is a general purpose variety with a 24-hour curing time and is often used in the fabrication of composite materials.

PEEK (Polyetheretherketone) is a semicrystalline material with superior cheracteristics such as resistance to high temperatures, radiations and corrosion [25]. It displays excellent mechanical properties such as providing high wear resistance and may well be used in applications in high temperature environments (about 300°C). As demonstrated in another work [26,27], its resistance to radiations is much higher than for most of the polymers. The glass transition temperature is 143°C, which does not represent a problem for application in the container fabrication. The tensile strength of PEEK exceeds that of most of the other engineering plastics and, with reinforcement, tensile strengths over 100 MPa can be obtained and maintained at temperature above 300°C.

Boron is selected as the reinforcement fibre mostly because of its excellent machanical and chemical properties at elevated temperatures [28]. The high absorption cross section for boron for thermal neutrons (759 b) is also a desirable asset of this material, although the intensity of the thermal neutron flux at the position of the container wall would be very small making the neutron absorption in these fibres negligible indeed. It is rather the excellent properties of resistance to corrosion and outstanding compressive strength, plus the ever increasing uses of this material in the fabrication of composites which justify this choice.

In addition to evaluating glass beads suggested in the AECL proposal for the container filler material, this work also looks at thorium dioxide (ThO_2) for this filler. This choice is based on the excellent physical and chemical properties of this ceramic, such as excellent resistance to corrosion by air and water and very high melting point (3050°C) [29]. Although mildly radioactive itself, thorium-232 has a very long half-life (1.39×10^{10} years), longer than the two natural uranium isotopes and its presence even in more concentrated form wouldn't add much to the natural radioactivity background. In addition, ^{232}Th is mainly an alpha and beta emitter, with low penetration power, and it displays a moderate thermal neutron absorption cross section of 7.4 b sufficient to shield the container wall from most of the residual neutrons emitted by the decaying fuel bundles. The most interesting property is the gamma and X-ray absorption properties of ThO_2 which could be comparable to those of lead, since the atomic number of ^{232}Th (90) is higher than that of lead (82). The density of 10.03 g cm^{-3} of ThO_2 (after sintering) is comparable to that of lead (11.3 g cm^{-3}), but the mass attenuation coefficients (μ/ρ) at 1 MeV are 5.293 $cm^2\,g^{-1}$ and 5.105 $cm^2\,g^{-1}$ for ThO_2 and Pb, respectively, as given by the code Microshield™ [30]. In addition, the cost of ThO_2 is expected to be quite low considering that thorium is three to five times more abundant than uranium in the Earth's crust, is very often associated with uranium in the same ores and is discarded among the mine tailings at uranium mines concentrators.

HEAT TRANSFER

Within the vault at a nominal depth of 1000 m, the temperature of the plutonic rock is approximately 17°C [3]. However, the decay of the radioisotopes within the spent fuel produces a significant amount of heat and, as the result of heat transfer through the filler material, the walls of the container may be subjected to temperatures quite higher than that of the ambient granitic rock. Basic heat transfer calculations were carried out for all four composites, based on a TK SOLVER [31] program, and gave the following temperatures at the surface of the container wall: 61.5°C for the PS, 49°C for the PMMA, and 49-50°C for both the PEEK-based and the Devcon epoxy-based composites. For the polystyrene-based composite, the temperature constraint of 100°C imposed in the AECL proposal for the disposal vault would be exceeded, thus eliminating this material *de facto*. As for the PMMA-based composite, the eventuality that the disposal vault temperature may approach 100°C leads to the elimination of this material as a suitable candidate since the glass transition temperature of PMMA is only 105°C and this could lead to structural failure of the material.

As for the other two materials, the use of either one is acceptable since the disposal vault temperature wouldn't exceed the 100°C constraint, nor the glass transition temperature would be approached since, for both materials, it is well above 100°C.

MECHANICAL STRENGTH

The mechanical stress applied to the container is evaluated on the basis of a 13 MPa constant external pressure as assessed in the AECL proposal [3], and the calculations [32] were based on determining the minimal wall thickness that would resist to such external pressures in the case of an empty container. Of course, in an actual application, the container would be filled with not only the spent fuel bundles, but also the filler material and possibly partition walls, all of which would provide resisting forces on the container walls counteracting the effects of the external pressure. Two boron fibre/polymer mixtures (50% and 70% mass) were considered for this study and the container wall thickness was varied from 16.99 to 25.97 mm for the four polymers considered. Additional calculations indicated that, for these wall thicknesses, these materials could easily withstand the stresses from the container payloads, their strengths being, in most cases, of two orders of magnitude greater than the required strengths.

DEGRADATION PROCESSES OTHER THAN RADIATIONS

Geochemical conditions have both positive and negative effects on the waste container. The presence of minerals with high sorption capacity for contaminants provides favourable conditions for the retention of contaminants and impedes their transport within the groundwater pathways. The type of groundwater found at the site of the burial vaults is rich in dissolved minerals as a result of extended percolation through the fractures within the plutonic rock for thousands of years [3]. While the polymeric materials considered for the container fabrication all display high resistance to corrosion, the salinity of the groundwater may in some cases play a role and, in the case of some epoxies, the polymer matrix may become brittle as a result of water absorption due to the presence of hydrophilic hydroxyl and ether groups in cures epoxies. This problem may however be alleviated by modifying the chemical structure of some of the reactants.

Some concern may be given to the degradation of polymers by microbian activity since the temperature would be higher than 17°C in the first decades of storage. However, at the depths considered for the disposal of spent nuclear fuel, the lack of nutrients within the environment coupled to a small ambient radioactivity would not favour the proliferation of microorganisms leading to the conclusion that biodegradation would not represent a significant process.

DETERMINATION OF RADIATION DOSE ON THE CONTAINER WALL

An important part of the research investigated the resistance to radiations of the composite materials under consideration. Resistance to radiations was one of the main reasons high polymer composites were not retained at first for the fabrication of the container, but new polymers have recently appeared on the market that are indeed much resistant to radiations. In a first part, dose calculations were carried out using the computer program Microshield™ [30] in order to determine the maximum dose rate on the wall of the container from the 72 spent fuel bundles which were assumed to have been placed in the container after a 10-year cool-down period in the storage pool at the nuclear generating station following discharge from the nuclear reactor. Figure 3 shows the geometry used by Microshield™ to carry out the calculations with T1 and L being the radius of the inner cylinder containing the fuel bundles and the length of the container, respectively, T2 is the thickness of the filling material, T3 the thickness of an air gap, T4 and T5 are the thicknesses of the fibre and polymeric materials. Since the dose point is at mid-plane, the vertical position Y is zero.

For these calculations, the most important radionuclides in terms of activities were included in the source term of the model. The CANDU fuel bundle investigated in this work is the standard 37-element Bruce "A" reactor bundle which has been exposed to a thermal neutron flux of 1.26×10^{14} neutrons cm^{-2} s^{-1} during 228.72 days, thus having accumulated a burnup of 685 GJ kg^{-1} initial U (7928 MW-days $tonne^{-1}$ initial U). The activities of these radionuclides were taken from an AECL publication [33] and actinides were retained for the calculations if their activities after 10-year decay was larger than 10^{-5} Ci kg^{-1}, whereas the fission products retained were those for which the activity was more than 10^{-3} Ci kg^{-1}. Table I lists the radioisotopes retained for this study. The Microshield™ model of the

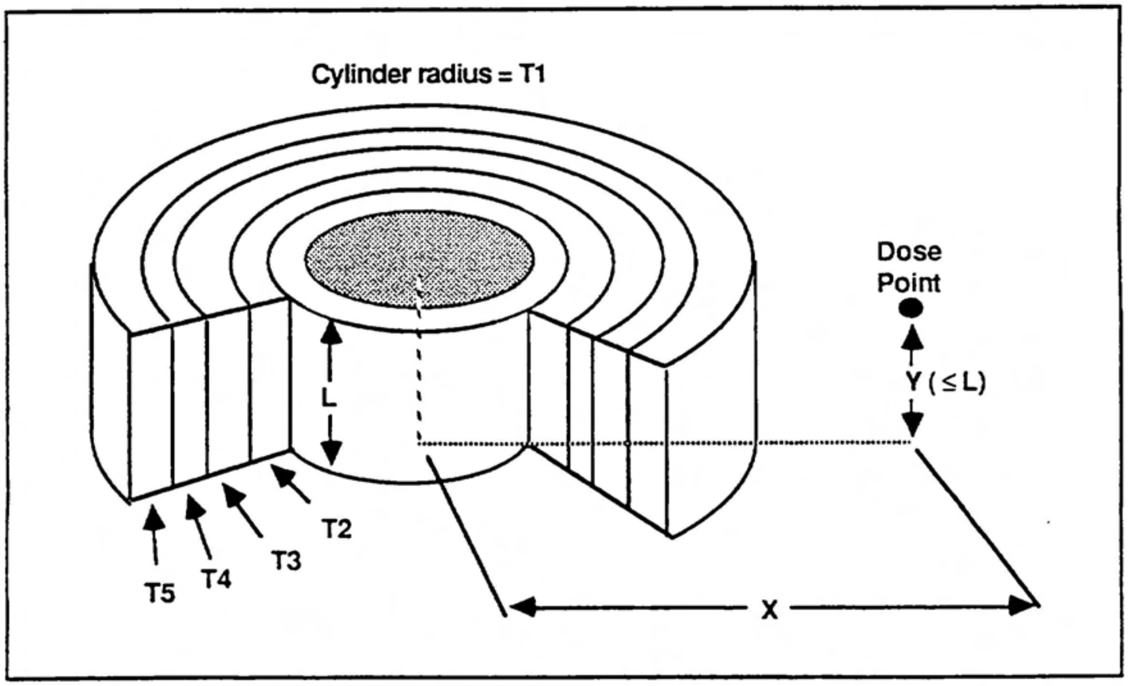

Figure 3: Geometry of the Container Used by Microshield™

container included two different buffer materials: glass beads as proposed by AECL, and thorium dioxide (ThO_2). Typical dose rates of 11 Gy h^{-1} in the container wall were calculated with glass beads as filling material, and of about 0.05 Gy h^{-1} when thorium dioxide serves as filling material. Calculations without any filling material were also carried out and yielded dose rates at the container wall of about 19 Gy h^{-1}. Table II presents the detailed results of the Microshield™ calculations. The results indicate that the ThO_2 filler absorbs most of the gamma radiations leaving the walls of the container well shielded. Many of the results in the last column of Table II are negative, indicating that, for the materials of the container wall, the Compton effect is more important than the photoelectric and the pair production effects, giving an important built-up of photons (as calculated by Microshield™ using the conservative Taylor option). In reality, the dose absorbed within the container walls for these materials would be quite small. In this work, Microshield™ was used with the other build-up calculations options (GP and no build-up effect (B=1)), and the Taylor's method option yielded results comparable to those of the GP method, although slightly conservative. Finally, since thorium is a naturally radioactive isotope itself, an additional calculation was made to estimate its contribution to the radiation dose sustained by the container wall: this was found to be very negligible indeed.

RESISTANCE OF MATERIALS TO RADIATIONS

The research then focused on determining how the composite materials were capable to sustain such doses without failing. Samples of the four types of materials were irradiated in the pool of the SLOWPOKE-2 nuclear reactor at RMC-CMR for various durations ranging from 8 hours to more than 80 hours . Various tests were carried out on irradiated dog bone-shaped samples and the results were compared with those for unirradiated samples [22-24. 26-27]. The dose rate at the irradiation site has been determined at 2400$\pm$960 Gy h^{-1} at half reactor power [33]. The doses accumulated within the samples during their irradiation in the reactor pool (about 3.2×10^5 Gy for the PEEK) correspond to a 3.2 year storage time in the underground vault with glass bead filling material, and to 698 years with thorium dioxide filler, if the initial source strength remains constant. When accounting for the decay of the fission products and the actinides, this corresponds to storage times of 3.3 years and infinity for the glass beads and thorium dioxide fillers, respectively.

TABLE I : ACTIVITY WITHIN CONTAINER AFTER 10 YEARS OF STORAGE[1]

ACTIVES *	SPECIFIC ACTIVITY	ACTIVITY	FISSION PRODUCTS **	SPECIFIC ACTIVITY	ACTIVITY
Nuclide	Ci (kg U)$^{-1}$	Ci per container	Nuclide	Ci (kg U)$^{-1}$	Ci per container
Cm 244	0.0121	16.6	H 3	0.0799	109
Cm 243	1.41×10^{-4}	0.193	Kr 85	1.23	1.68×10^{3}
Cm 242	1.8×10^{-4}	0.246	Sr 90	13.9	1.90×10^{3}
Am 243	5.24×10^{-4}	0.717	Y 90	13.9	1.90×10^{3}
Am 242	2.17×10^{-4}	0.297	Tc 99	0.00347	4.75
Am 242m	2.17×10^{-4}	0.298	Ru 106	0.226	309
Am 241	0.299	409	Rh 106	0.226	309
Pu 242	2.28×10^{-4}	0.325	Cd 113m	0.00459	6.29
Pu 241	14.5	1.98×10^{4}	Sb 125	0.241	330
Pu 240	0.235	321	Te 125m	0.0588	80.4
Pu 239	0.172	235	I 131	~0	~0
Pu 238	0.0822	112	Cs 134	0.525	718
Np 239	5.24×10^{-4}	0.717	Cs 137	20.7	2.83×10^{4}
Np 237	2.74×10^{-5}	0.0375	Ba 137m	19.6	2.68
U 238	3.31×10^{-4}	0.453	Ce 144	0.0772	106
U 237	3.62	0.495	Pr 144	0.0772	106
U 236	4.97×10^{-5}	0.0680	Pm 147	5.45	7.46×10^{3}
U 234	2.69×10^{-4}	0.368	Sm 151	0.0607	83.0
Pa 234m	3.31×10^{-4}	0.453	Eu 154	0.503	688
Th 234	3.31×10^{-4}	0.453	Eu 155	0.213	291
TOTAL	15.3	2.09×10^{4}	TOTAL	77.1	1.05×10^{5}

TOTAL ACTIVITY IN CONTAINER DUE TO ACTINIDES AND FISSION PRODUCTS: 1.26×10^{5} Ci

*: Actinides were included only if their activity was greater than 10^{-5} Ci (kg U)$^{-1}$.

**: Fission products were included only if their activity was greater than 10^{-3} Ci (kg U)$^{-1}$.

TABLE II : Microshield™-CALCULATED DOSES IN CONTAINER WALLS

CONTAINER MATERIAL	FILLER	DOSE RATE INSIDE WALL Gy h^{-1}	DOSE RATE OUTSIDE WALL Gy h^{-1}	ABSORBED DOSE IN SHELL Gy h^{-1}
TITANIUM	NONE	14.5	15.2	-0.66
TITANIUM	GLASS BEADS	11.2	7.14	4.02
TITANIUM	ThO_2	0.0517	0.0573	-0.00565
COPPER	NONE	14.5	4.98	9.50
COPPER	GLASS BEADS	11.2	1.83	9.34
COPPER	ThO_2	0.0517	0.014	0.0379
PS-50% BORON	NONE	14.5	18.6	-4.65
PS-50% BORON	GLASS BEADS	11.2	10.9	-0.180
PS-50% BORON	ThO_2	0.0517	0.0917	-0.0400
PS-70% BORON	NONE	14.5	19.0	-4.52
PS-70% BORON	GLASS BEADS	11.2	11.2	-0.06
PS-70% BORON	ThO_2	0.0517	0.0944	-0.427
PMMA-50% BORON	NONE	14.5	18.6	-4.14
PMMA-50% BORON	GLASS BEADS	11.2	10.4	0.72
PMMA-50% BORON	ThO_2	0.0517	0.0872	-0.0355
PMMA-70% BORON	NONE	14.5	19.1	-4.6
PMMA-70% BORON	GLASS BEADS	11.2	11.1	0.72
PMMA-70% BORON	ThO_2	0.0517	0.0931	-0.0414
EPOXY-50% BORON	NONE	14.5	19.1	-4.58
EPOXY-50% BORON	GLASS BEADS	11.2	10.5	0.67
EPOXY-50% BORON	ThO_2	0.0517	0.0871	-0.0354
EPOXY-70% BORON	NONE	14.5	19.3	-4.77
EPOXY-70% BORON	GLASS BEADS	11.2	11.0	0.13
EPOXY-70% BORON	ThO_2	0.0517	0.0924	-0.0407
PEEK-50% BORON	NONE	14.5	19.0	-4.46
PEEK-50% BORON	GLASS BEADS	11.2	11.0	0.16
PEEK-50% BORON	ThO_2	0.0517	0.0923	-0.0406
PEEK-70% BORON	NONE	14.5	19.1	-4.65
PEEK-70% BORON	GLASS BEADS	11.2	11.3	-0.18
PEEK-70% BORON	ThO_2	0.0517	0.0957	-0.0440

For the Devcon #10210 epoxy, the effects of irradiation indicated an increase of the tensile strength from 34±7 MPa to 44±7 MPa after 8 hours of irradiation. Previous work [22-24, 26,27] shows that the strengthening effect due to radiation promoted crosslinking becomes eventually overcome by chain scissions as exposure progresses, resulting in a significant decrease of the tensile strength past 120 hours exposure. Several samples of PEEK of two different grades were also irradiated at half reactor power for up to 83.6 hours. Even after long exposure times, little change was observed in the tensile strength other than, in some cases, a modest increase. For the 450P grade PEEK, the tensile strength was measured at 103 MPa when unirradiated and a similar value was obtained after a dose of 3.2×10^5 Gy (83.6 hours irradiation). Unirradiated 150P grade PEEK yielded a 108 MPa tensile strength while a higher value of 109 MPa was obtained after a 83.6 hour irradiation. It is important to note here that all samples were irradiated in contact with the pool water providing an oxygenated environment which emphasizes the chain scission process and decreases the polymers' resistance to radiations.

DETERMINATION OF PRESSURES

The pressure calculations were based on conservative assumptions such as considering that the container would have to support by itself the external pressures to which it would be subjected in the deep underground storage vault, without consideration for resistance offered by the filling material or other structural materials located inside the container. The average container inside diameter (0.645 m) and the length (2.246 m) for the containers proposed by the AECL were used, and the characteristic external pressure was that of 13 MPa determined in the AECL proposal [3]. Using the respective strengths of the materials considered for the fabrication of the container, the minimum thickness needed for the basic right cylinder to withstand the pressure effects of the plutonic rock were calculated by solving the usual axial and hoop stress equations [32] based on the thin-walled vessel assumption and an internal constant pressure pushing out on the container. In a second step, a reversal of external pressure and compressive properties was executed, using the Poisson's ratio for the materials. In a third step, the total strength of the composite (polymer + boron fibres) was determined as a simple volume-weighted average of the strength of the fibre and that of the polymer. The fourth part of the pressure calculations took in consideration the load applied to the container during the transportation of the container to the vault, as the container itself must be strong enough to withstand the weight of the 72 fuel bundles and the filler material. This weight is 1880 kg when thorium dioxide is used as filler. Table III below presents the results of these calculations.

TABLE III: STRENGTH OF COMPOSITE MATERIALS AND THICKNESS OF CONTAINER WALL

COMPOSITE MATERIAL (% by mass)	TENSILE STRENGTH (MPa)*	REQUIRED TENSILE STRENGTH (MPa)	COMPRESSIVE STRENGTH (MPa)	WALL THICKNESS OF CONTAINER (mm)
PS with 50% boron	41.9	0.337	161	26
PS with 70% boron	49.1	0.425	207	20
PMMA with 50% boron	51.5	0.372	178	24
PMMA with 70% boron	57.0	0.477	226	19
EPOXY with 50% boron	52.0	0.432	205	20
EPOXY with 70% boron	57.6	0.523	247	17
PEEK with 50% boron	88.1	0.406	193	22
PEEK with 70% boron	82.6	0.506	239	18

* : The tensile test data for PS and PMMA are based on the minimum tensile strengths measured using the 28.5 hour irradiation data. Those for the Devcon 10210 epoxy and the PEEK are based on the 8- and 80-hour irradiation in the SLOWPOKE-2 reactor pool.

COST ANALYSIS

It was possible to carry out only a crude cost analysis since the prices of some of the materials could be only roughly estimated and the exact fabrication process for the container wall remains to be designed along with some details such as the engineering of the lid cover and the structural elements used to position the fuel bundles. One of the best kept secrets is the value of the thorium dioxide, but considering that it is part of the uranium mine tailings and that the uses for thorium are very few, its real value must be much lower than that of natural uranium dioxide. In fact, the mine tailings may well have a negative value if one accounts for the costs incurred in disposing these materials in an environmentally acceptable fashion. As an upper limit cost, this analysis uses the cost of nuclear purity UO_2, which itself varies by more than 50% due to the demand-offer on the spot market. A value of \$30/kg ThO_2 is therefore assumed for the filling material, vice \$3.40/kg for the glass beads. The prices for PEEK and boron fibre have been estimated at respectively \$110/kg and \$275/kg. The costs of the other materials are as reported in Table IV below.

These results show that, even if an extreme assumption was made equating the value of ThO_2 , a non-fissile material with little demand, to that of natural UO_2 , a fissile material highly sought to produce 19% of the electricity in Canada and up to 75% in France, the cost of a container containing ThO_2 as the filling material would not be even three times higher than the cost of the same container containing glass beads as proposed by AECL. When one compares the cost of the polymer-based container with that of a titanium alloy container, the ratio is about 1.8 for the PEEK and 2.13 for the epoxy. Since the price of PEEK is likely to decrease as more applications are found for this material, and considering that the cost of ThO_2 used in this study is much inflated, it is expected that the cost of the container as proposed in this work may well be comparable to that of AECL's proposal.

TABLE IV: COST ANALYSIS

COMPOSITE MATERIAL (% by mass)	VOLUME OF POLYMER (cm^3)	VOLUME OF BORON FIBRE (cm^3)	CONTAINER WALL MATERIAL COST \$	CONTAINER COST GLASS BEADS FILLER \$	CONTAINER COST THORIUM DIOXIDE FILLER \$
PS with 50% boron	96150	38130	26500	28400	99200
PS with 70% boron	55760	48270	33550	35300	106000
PMMA with 50% boron	124510	13520	10200	12100	82900
PMMA with 70% boron	49080	52320	36300	38300	109000
EPOXY with 50% boron	52560	52140	42000	44000	114700
EPOXY with 70% boron	30260	56460	42300	44300	115000
PEEK with 50% boron	74080	37490	36400	38300	109000
PEEK with 70% boron	41010	48530	39200	41200	109000

Density of materials ($g\ cm^{-3}$): PS:1.07 ; PMMA: 1.19 ; Epoxy : 2.11 ; PEEK : 1.30 ; Boron : 2.50.
Costs of materials (\$ kg^{-1}): PS : 2.74 ; PMMA: 6.07 ; Epoxy: 55.74 ; PEEK : 110.00 ; Boron : 275.00.
Glass beads : 3.40 ; Thorium dioxide : 30.00.
Volume of filler material : 245700 cm^3.

Cost of Titanium Container : with glass filler: \$20800. ; with ThO_2 filler : \$90900.

DISCUSSION

The analysis of various polymers for use in the fabrication of a suitable container for the long-term deep underground storage of high level radioactive waste is based on the proposal by Atomic Energy of Canada Limited and used many of the data presented in the relevant publications. The activity data presented by the AECL [33] used only three significant digits, indicating that their accuracy was on the average 0.5%. The Microshield™ calculations are fairly straightforward and based on built-in data such as the mass attenuation coefficients with a 0.1% accuracy or better. However, the build-up calculations may produce results with as much as 20% variation depending on the build-up factor option selected. A conservative estimation of the accuracy of the dose calculation by Microshield™ must then retain this 20% error on these results.

The results obtained from the irradiation of the polymers in the pool of the SLOWPOKE-2 reactor are reported with accuracies of 5% or better. The reactor power is actually maintained within 0.2% by the reactor automatic control system, which is a remarkable achievement. However, the accurate determination of the dose received represents a challenging problem since, even if the thermal neutron flux, the fast neutron flux and the gamma flux have been determined within 10% accuracy or better at the irradiation site. The dose-to-flux conversion factor remains an elusive target as long as the neutron flux spectrum and accurate quality factors cannot be determined with sufficient accuracy. Therefore, a 40% error bar is used in a conservative fashion for the dose determination from the irradiation durations.

The heat transfer and the mechanical strength calculations are straightforward and all the parameters are known with good accuracy, permitting to assess the error bars on these results within 1-2% at the most. It remains that the confidence in the results obtained in this work is sufficient for a good analysis of the potential of using high polymers in the fabrication of the containers for the storage of spent CANDU nuclear fuel bundles. Provided sufficient radiation shielding is provided by the filler material used within the container, both epoxy and PEEK may be used along with reinforcing boron fibre, with PEEK neatly the best of the two materials. This may be stated with assurance since both materials were irradiated in the SLOWPOKE-2 reactor pool in contact with water and subsequently investigated with a battery of tests to assess their behaviour and resistance to radiations. It was determined in previous work [22-24, 26,27] that when high polymers are irradiated in an oxygenated environment such as water, their mechanical strength deteriorates significantly more than when irradiated in an inert environment. Therefore, our work represents a conservative approach to the investigation of these polymers as its results would apply for a worst case scenario in which the composite container wall would be in contact with underground water when irradiated. One could easily imagine protective coatings as part of the container wall fabrication which would need to keep the composite material wall dry for 50 to 100 years only, after which the radiation level would decrease to level too low to cause significant damage to the container wall, even if the material becomes in contact with water.

An important point remains here to be discussed: that is the density of the ThO_2. In the Microshield™ model, the density of sintered thorium dioxide was used (10.03 g cm^{-3}). Assuming that sintering of thorium dioxide pellets could be compared to that of uranium dioxide pellets, it is known that the sintering operation increases the density of the compacted UO_2 pellet by about 25% so that sintering ThO_2 would have a comparable effect. As filler for the spent fuel container, the thorium dioxide would be used as a compacted powder since sintering would be hardly practical and costly, therefore, the density of the thorium dioxide powder would rather be of the order of 8 g cm^{-3}. The dose rates within the container would then be a bit higher than those calculated here (by a factor of approximately two), but still very much acceptable.

CONCLUSIONS AND RECOMMENDATIONS

These results represent the first phase of a more extensive research aimed at evaluating the possibilities of using high polymer composite materials for the fabrication of the high level radioactive waste storage container. There are clear indications that both PEEK and Devcon #10210 epoxy are promising candidates for this kind of application, especially when the design of the storage container provides radiation shielding materials to protect the container wall in the form of thorium dioxide. Further tests are needed on these materials to ascertain their resistance and their properties under radiation. In particular, the resistance to corrosion, which was only considered up to now from literature survey and found to be comparable if not superior to that of the metals considered in the AECL proposal, needs to be confirmed in laboratory testing which would involve ageing techniques. In particular, more knowledge is needed on water ingression within some composites. A crude cost analysis was carried out revealing that the high cost of PEEK (about $110/kg) and that of the boron fibres ($275/kg) may represent a significant drawback, but it is expected that increased production to meet a larger demand for this product may well drive the prices down within a few years, making the economics indeed favourable to the point of making the cost of producing a polymer (PEEK)-based container with thorium dioxide filler comparable to the cost of a similar container based on titanium alloy for the shell and using glass beads for the filling material.

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