High Polymer-Based Composite Containers for The Disposal/Storage of High, Intermediate, And Low Level Radioactive Waste

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Spent nuclear fuel bundles account for the bulk of Canada's high level radioactive waste (HLW). Presently approximately 1.5 million bundles of this waste is accumulating in interim storage according to the Atomic Energy of Canada Limited (AECL)[1,2]. The question is: what to do with it? In 1957 scientists first proposed to bury waste deep in the geologically stable rock formations of the Canadian Shield. After many years of research, this option is now realised to be technically feasible, economically justifiable, and safe to do so. Since the spent fuel must be safely housed for thousands of years (far longer than any single human civilization has survived) the disposal management system must not depend on institutional control to maintain safety. AECL therefore, proposes the indefinite, non-retrievable, storage in deep underground vaults within the plutonic granite rock of the Canadian Shield.

Prior to implementation of AECL's proposed disposal concept[1,2], bundles would spend six to ten years in wet storage at the reactor site to dissipate most of their heat and allow the radioactive emissions from over 200 isotopes to decay to a safer level. The proposal is based on a strategy of multiple containment barriers to ensure that leakage of radioactive material will not enter and contaminate the biosphere. The proposed ASTM Grade-2 Titanium (first choice) or oxygen-free copper (second choice), container would hold up to 72 spent CANDU bundles packed in glass beads. Each container would then be packed in a clay buffer in separate holes 500 to 1000 m deep in the plutonic rock and back filled with concrete and rock. Disposal in this manner would ensure security and safety indefinitely. One of the greatest challenges prior to implementation of such a project is to gain political and public acceptance. This process may be eased by the assurance of leak-tight disposal containers. Preliminary research conducted at Royal Military College (RMC)[3] shows that containers made from high polymers/fibre composites may provide a better containment vessel than titanium.

Once put in use, these containers would be subjected to environmental aggression over a period extending thousands of years. They absolutely have to be designed with a guarantee that failure is all but impossible until long after such time as the radiation emitted by their cargo has decreased to a safe level. Design criteria must therefore consider mechanical strength, as well as resistance to radiological and chemical effects. The greatest concern of course is that if the containers fail, ground water may come into contact with the spent fuel and that radioactive contaminants may be leached out into the water table. The most likely cause of container failure is corrosion, in the case of a metal container since ground water often carries corrosive impurities. Titanium is resistant to corrosion and this is why it was AECL's first choice. Due to the long time periods involved, greater confidence in a leak-tight capability would be achieved if corrosion were not an issue at all. This is where composites may appear as a possible solution to the problem of corrosion, since they display excellent chemical resistance, even to strong acids, in addition to being comparable to metals with respect to mechanical strength. They are also lighter (less dense), often cheaper, and some of them show promising resistance to radiation. These material properties make composites good candidates for the replacement of titanium.

The preliminary work done at RMC has been conducted with the AECL design criteria in mind and has shown that the use of polymeric-based composites is justified for a number of properties: mechanical, radiation resistance, thermodynamics, and chemical resistance.

In terms of ultimate strength, a composite with a high polymer matrix, such as epoxy or poly(ether ether ketone) (PEEK), with a strong fibre reinforcement, such as carbon or boron fibre, can exhibit properties which rival and in some cases better than those seen in metals. An analysis has been conducted on the AECL proposed container design, using an external pressure of 13 MPa to simulate hydrostatic pressure in the disposal vault and found that the minimum container thickness is 20 mm with an epoxy / 50% boron fibre composite. However, in order to accommodate for safety factors, the thickness should be between 2 and 3 times higher than this. It is also especially important to account for the inherent viscoelastic properties of polymers. The containers will be exposed to pressure and heat for an extended period of time. The epoxy-based composite is very resistant to creep because of the crosslinking which takes place in the polymer upon curing. PEEK has also been considered as a matrix material in the composite with 50% boron fibre, and has been found that the minimum thickness of 22 mm is sufficient to withstand the 13 MPa hydrostatic pressure. Table I shows some tested strengths of composite materials and their corresponding wall thickness.

Radiation resistance of polymers is well documented, and research at RMC has shown that both epoxy and PEEK are very resistant, even increasing in strength due to an increase in crosslink density. If the radiation from spent fuel becomes a concern, then thorium dioxide is proposed to be used in place of the AECL proposed glass beads: THO_2 has been shown to not only have excellent physical and chemical properties, but also has excellent gamma and x-ray absorption properties. Table II shows Microshield™ [4] calculations at the container walls for various materials.

For various reasons, the container is designed such that the temperature of the container wall must be below 373 K. For this to occur, proper heat transfer mechanisms must exist to dissipate the heat produce by the fuel. Heat transfer simulations, both from a scaled down mockup and by numerical and analytic calculations, have been conducted with the results indicating that at the outer surface of the container wall would not exceed 365°±4 K. Table III contains the results of the analytic heat transfer solution.

Because the containers in the disposal vault will be exposed to ground water and other chemicals that are dissolved in the water, chemical resistance is an important property. Corrosion will not be a problem as it is in metals, however interfacial deterioration between the fibres and polymer, as well as hydratation of the polymer, are indeed concerns. Both polymers proposed in this paper are chemically resistant to many chemicals and have been shown to be able to withstand the adverse environments in the vault.

Research previously done at RMC justifies further work especially in the application of composites. Recommendations for further work include:

a. investigation of additional high polymers (Epoxies, PEEK, etc.),
b. investigation of composites with graphite and glass fibres and various mat patterns,
c. determination of chemical resistance of composites (Ageing procedures),
d. further study of $Th0_2$ as filler,
e. application to intermediate and low level radioactive waste disposal,
f. processing and fabrication of container, and
g. costs.

Most of the research done to date involves designs focussed on the spent fuel disposal container. Although this is a complex problem with many engineering considerations, the safe disposal/storage of the low level and intermediate level radioactive wastes (LLW, ILW) is of great interest as well, due to the sheer quantities involved. Application of composites to these lower level radioactive waste containers holds great potential and is the subject of a separate research project at RMC. The main differences in design criteria between the HLW and ILW/LLW containers is that the spent fuel container must withstand higher external stresses, heat, and radiation effects compared to the other container. The use of PEEK in the design of a better HLW container is the subject of a second research project.

The design criteria for the HLW container have already been determined by AECL, however, this is not the case for the ILW/LLW container(s). Design criteria can only be established once a thorough knowledge of the contaminants is known, such as the form, activity, and isotopic content. The next step will then be to acquire relevant materials based on properties that will hopefully meet design requirements. A series of testing will then follow including the use of the SLOWPOKE-2 Nuclear Reactor to irradiate the material samples. Samples will then undergo mechanical testing to determine the feasibility of composite materials for use in radioactive waste containers.

REFERENCES

1. "Summary of the Environmental Impact Statement on the Concept for Disposal of Canada's Nuclear Fuel Waste", AECL-10721, 1994.

2. "Environmental Impact Statement on the Concept for Disposal of Canada's Nuclear Fuel Waste", AECL-10711, 1994.

3. H.W. Bonin and V.T. Bui, "High Polymer-Based Composites for Spent Nuclear Fuel Disposal Containers", Royal Military College of Canada, Kingston, Ont., 1999. (Conf. Proc. Vol 1, Sixth International Conference on CANDU Fuel, 1999 September 26-30, Niagara Falls Canada)

4. Grove Engineering Inc, Microshield^{TM}: Version 3 (1988)

TABLE I: 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.

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

TABLE III: HEAT TRANSFER DATA (Analytical Solution)

(Ten-year storage after discharge from reactor core)

FUELLED REGION	PACKING MATERIAL	CONTAINER WALL MATERIAL	TEMPERATURE AT THE CENTRE OF THE FUELLED REGION	TEMPERATURE AT THE INSIDE SURFACE OF THE CONTAINER WALL	TEMPERATURE AT THE OUTSIDE SURFACE OF THE CONTAINER WALL
UO_2 +GLASS BEADS	GLASS BEADS	TITANIUM	311±3 K	296.3±.2 K	296.3±.2 K
UO_2 +GLASS BEADS	GLASS BEADS	COPPER	311±3 K	296.3±.2 K	296.3±.2K
UO_2 +GLASS BEADS	GLASS BEADS	POLYMER	315±8 K	300±7 K	297.4±.3 K
UO_2 +GLASS BEADS	ThO_2	TITANIUM	300.3±.5 K	296.3±.2 K	296.3±.2 K
UO_2 +GLASS BEADS	ThO_2	COPPER	300.3±.5 K	296.3±.2 K	296.3±.2 K
UO_2 +GLASS BEADS	ThO_2	POLYMER	304±16 K	300±6 K	297.4±.3 K

SIMULATION PARAMETERS

Volumetric heat production $Q = 8.44 \pm .01 \times 10^2$ W m^{-3} (Corresponds to 4.4 kW per bundle, after 10 years storage after discharge from the reactor core).

Fuelled region radius a = 0.23175 m
Outer radius of packing material region b = 0.31015 m
Outer radius of container wall c = 0.3165 m

Height of inside cavity of container H = 2.246 m

Heat transfer coefficient by conduction for fuelled region $k_1 = 3.49 \pm 0.35$ W m^{-1} K^{-1}
Heat transfer coefficient by conduction for packing region (glass beads) $k_2 = 0.58 \pm 0.1$ W m^{-1} K^{-1} Heat transfer coefficient by conduction for packing region (ThO_2) $k_2 = 8.37 \pm 1.0$ W m^{-1} K^{-1}
Heat transfer coefficient by conduction for container wall (Titanium) $k_3 = 22.0 \pm 1.0$ W m^{-1} K^{-1} Heat transfer coefficient by conduction for container wall (Copper) $k_3 = 399.0 \pm 10.0$ W m^{-1} K^{-1} Heat transfer coefficient by conduction for container wall (Polymer) $k_3 = 0.16 \pm 0.1$ W m^{-1} K^{-1}

Heat transfer coefficient by convection for container wall (Titanium and Copper) $h = 22.7 \pm 1.0$ W m^{-2} K^{-1}
Heat transfer coefficient by convection for container wall (Polymer) $h = 16.9 \pm 1.0$ W m^{-2} K^{-1}

Temperature of ambient air $T_{air} = 293.16$ K