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RADIONUCLIDE INVENTORY CALCULATIONS FOR STRUCTURAL MATERIALS OF SMALL MODULAR REACTORS (SMR)

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Abstract

In support of the Pan-Canadian Small Modular Reactor (SMR) roadmap activities, radionuclide inventory calculations for structural materials of various SMR types are performed. The calculations provide essential data for improving the understanding of reactor safety, radioactive material management, and environmental impacts. The Monte Carlo code SERPENT is used to model five major types of SMR, based on the latest published design data, to obtain neutron flux distributions in space and energy as functions of operating schedule, and the SCALE/ORIGEN code is then used for time-dependent neutron activation and decay calculations. Radionuclide inventories have been comprehensively estimated for principal structures of five SMRs: integral pressurized water reactor (iPWR), molten salt reactor (MSR), high-temperature gas-cooled reactor (HTGR), lead-cooled fast reactor (LFR), and heat pipe based reactors. The primary contributing reactor components, the dominating radionuclides to the total activity, and the radioactivity evolution with time after reactor shutdown, are discussed.

Keywords: neutron activation, SMR, radionuclide inventory, SERPENT, ORIGEN

1. Introduction

As outlined in CNL's ten year plan [1], it is our goal to become recognized globally as a leader in small modular reactor (SMR) prototype testing and S&T support, including hosting a demonstration reactor on a CNL site by 2026. The work described in this paper estimates radionuclide inventories for various types of SMRs for the purpose of providing essential data for improving the understanding of reactor safety, radioactive waste management, and environmental impacts in support of the Pan-Canadian SMR Roadmap Activities. This includes neutron activation waste for the major reactor components that will be decommissioned and disposed of according to applicable waste management processes. Fuel assemblies to be removed and disposed separately are not included in this study, and may be assessed in future work.

Neutron activation inventory was calculated for five selected SMR designs from five major types of SMRs [2], which include:

- The NuScale Power Module™ (NPM) [3], a passive, small modular integral pressurized-water reactor (iPWR), developed by NuScale Power Inc. USA. It utilizes established light-water reactor technology, features a factory-fabricated compact self-contained module with natural circulation coolant flow, which is targeted to be used for electricity production and non-electrical process heat applications.

- The integral molten salt reactor (IMSR®) [4], a small modular molten salt fuelled reactor designed by Terrestrial Energy Inc. Canada. It features a replaceable completely sealed IMSR® core-unit including integrated pumps, heat exchangers and shutdown rods, which is designed for a short service lifetime and semi-automatic manufacture. The design provides the highest levels of inherent safety and a walk-away safe nuclear power plant.

- eVinci™ micro reactor [5], a semi-autonomous, very small modular reactor (vSMR) that utilizes proven heat pipe technology, designed by Westinghouse Electric Company LLC USA. Its small size allows it to be fully factory-built, fuelled and assembled. eVinci™ is targeted for generation of clean, safe and cost-competitive heat and electricity for remote communities, mines and military installations.

- The Micro-Modular Reactor (MMR) [6], a compact concept of high temperature gas cooled reactor (HTGR) designed by the Ultra-Safe Nuclear Corp. (USNC), USA, with all modules of power reactor, power conversion and power generation (PRM, PCM and PGM) integrated within a single unit. The MMR will be easily transportable and retrievable, and suitable for electricity generation and process heat applications in remote areas.

- SEALER (Swedish Advanced Lead Reactor) [7], a very small lead-cooled fast reactor (LFR) developed by LeadCold Reactors, Sweden. It features completely passive safety and is targeted for electricity and heat generation for remote communities such as arctic mining industry that lacks emergency response options.

These SMRs are all at various stages of design. The physical dimensions, materials and overall design features used in this study were interpreted from referenced open literature publications. They may not be exact copies of the actual designs and may be subject to changes.

2. Overall Methodology

The estimation of radionuclide inventories generated from neutron activations of SMR structural materials comprises the following two parts:

- neutron transport calculations using a Monte Carlo neutron transport code with burnup capability to obtain neutron flux distributions in space and energy based on the operating schedule of the SMR; and

- neutron activation calculations for SMR major components using the neutron flux data from transport calculations, followed by decay calculations for extended decay periods.

The neutron transport calculations were performed using SERPENT [8], a multi-purpose three-dimensional continuous-energy Monte Carlo particle transport code, developed at VTT Technical Research Centre of Finland, Ltd. SERPENT was selected for this study due to its ability to perform burnup calculations and provide higher accuracy results within reasonable computing resource demands as compared to other Monte Carlo codes. In this study, SERPENT was set to output group neutron spectra aligned with one of the SCALE/ORIGEN [9] neutron group structures for direct input into COUPLE, a module of SCALE, for library creation. Neutron fluxes for SMR components of interest were calculated in burnup steps from the beginning of life cycle (BOL) until the end of life cycle (EOL). The five SMR designs simulated in the study differ from each other significantly; therefore, they were modelled individually with their specific structure and operational features, which are discussed in the next section.

The neutron activation calculations were performed using SCALE/ORIGEN 2.2 [9]. ORIGEN calculates time-dependent concentrations, activities and radiation source terms for a large number of isotopes simultaneously generated or depleted by transmutation, fission, activation, and radioactive decay. In this study, COUPLE generates cross-sections directly using SERPENT-calculated spectra and fluxes corresponding to each burnup step.

3. Calculations and Discussions

3.1 NuScale Power Module™ (NPM)

The NuScale NPM [2] [3] is a self-contained passive nuclear steam supply system comprised of a reactor core, a pressurizer, and two steam generators integrated within the reactor pressure vessel and housed in a compact steel containment vessel (Figure 1). The core configuration consists of 37 fuel assemblies and 16 control rod assemblies (CRAs). The fuel assembly used in NPM is NuFuel HTP2™, a 17×17 square lattice with 264 fuel rods, 24 guide tubes, and one instrument tube in the centre. The fuel is made of UO_2 with Gd_2O_3 as a burnable absorber homogeneously mixed within the pellet in select locations, encapsulated in a seamless M5® fuel rod cladding. The uranium enrichment of individual fuel assemblies varies depending on the position of the fuel assembly in the core. Reactivity compensation and control are achieved through changing boron concentrations in coolant and maneuvering B_4C and Ag-In-Cd (AIC) control and shutdown rods. An NPM is modelled to have a thermal power of 160 MW, a two-year, three-batch refuelling cycle and a 60-year life time.

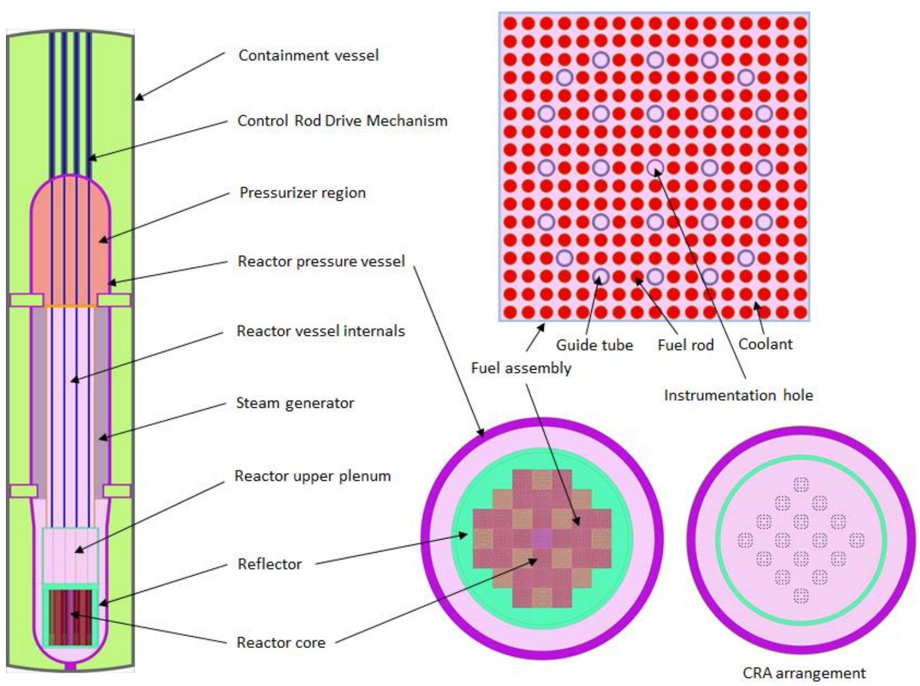

Figure 1. NPM™ reference equilibrium core and SERPENT full-reactor model

SERPENT burnup calculations were first performed for the core loaded with fresh fuel to obtain once-burnt and twice-burnt fuel compositions. A reference equilibrium core (REC) (Figure 1) was then created with regulating CRAs fully inserted in the core as would be the configuration for the beginning of cycle (BOC). Burnup calculations were performed in several burnup steps starting from BOC until the end of cycle (EOC) when CRAs would be fully withdrawn from the core. At each burnup step, burnable fuel material compositions were input into the next burnup step, and CRAs were partially raised at each burnup step. Boron concentrations in coolant were automatically adjusted using SERPENT feature "set iter nuc" in each burnup step to maintain core criticality.

Neutron spectra and fluxes calculated by SERPENT were input into COUPLE and ORIGEN for neutron activation calculations for 700 full-power days followed by a 20-day shutdown period for refuelling (a two-year refuel cycle). The refuel cycle is repeated 30 times in sequence to simulate 60 years of operation, followed by a decay period of 100 years.

Selected results presented in Table 1 indicate that:

- The total activity decreases by two orders of magnitude over a period of 100 years. The reflector and core barrel contribute about 90% of the total activity. CRAs and the RPV are the other two major contributors, representing about 10% of the total.

- After the short-lived isotopes decay away in the first year, Fe-55 and Ni-63 (with half-lives of 2.8 years and 101 years, respectively), produced from activation of the major steel composition Fe-54 and Ni-62, dominate the activity, representing 77% and 16% of the total activity, respectively, and continue to dominate for 10 years after shutdown. After most Fe-55 decays away in the first ten years, Ni-63 becomes the dominant radionuclide.

Table 1 Major Contributors to Total Activity for NPM

Component		**Decay time (year)**					
		0	**1**	**5**	**10**	**50**	**100**
	Total Activity (Ci)	**3.58E+06**	**4.59E+05**	**2.04E+05**	**1.08E+05**	**5.30E+04**	**3.79E+04**
	Mass (ton)	**Fraction of total (%)**					
Reflector	17.085	67	60	59	60	61	61
Core barrel	13.117	26	26	28	29	31	31
Control rod assembly	20.214	6	12	12	10	8	8
Reactor pressure vessel	116.618	1	2	1	1	<1	<1
Nuclide		**Fraction of total (%)**					
Fe-55		13	77	63	34	<1	<1
Ni-63		2	16	35	63	98	98
Co-60		<1	2	2	2	<1	<1
Ni-59		<1	<1	<1	1	1	1
C-14		<1	<1	<1	<1	1	1

3.2 Terrestrial Energy Integral Molten Salt Reactor (IMSR®400)

The IMSR®400 [2] [4] is a 400 MW thermal power small modular molten salt fuelled reactor (Figure 2). It features a completely sealed reactor vessel with integrated pumps, heat exchangers and shutdown rods all mounted inside a single vessel – the IMSR® core-unit. The reactor core consists of a graphite moderator with fuel channels arranged in triangular pitches. The fuel contains low-enriched uranium fluoride diluted with sodium fluoride and potassium fluoride, which constitutes both fuel and primary coolant. The fuel-coolant mixture is pumped between a graphite-moderated core, and then through the integral heat exchangers to transfer heat to the secondary loop. Reactivity is compensated through online fuel addition. Reactivity control is achieved through the intrinsic properties of the fuel salt combined with a strongly negative temperature coefficient of reactivity. The sealed core-unit is replaced at the end of its seven-year life cycle.

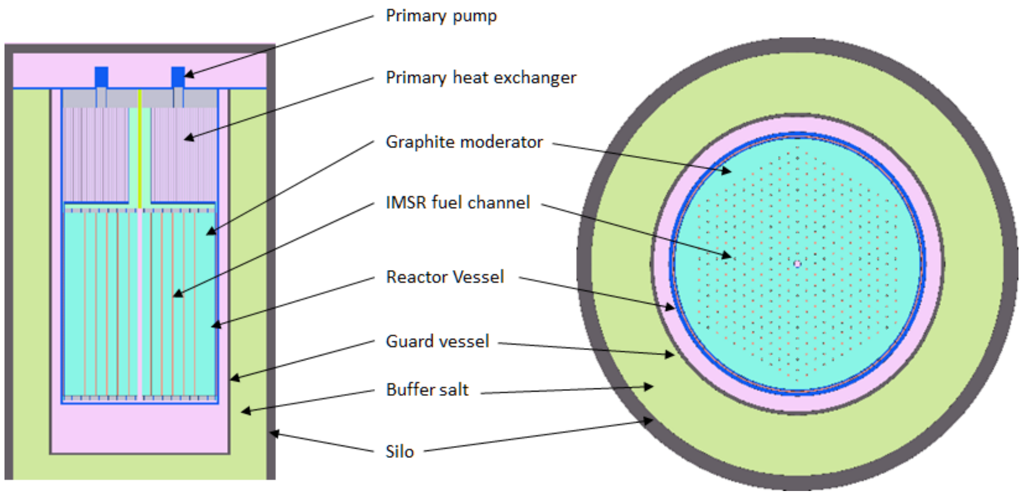

Figure 2. SERPENT model of IMSR®400

SERPENT burnup calculations were performed starting from a core loaded with fresh fuel at designed nominal temperature for a short period of time until poison level reaches equilibrium and the core is still close to critical, and this is used as a "snap shot" of an equilibrium core in this study. Neutron spectra and fluxes were taken from this equilibrium core and input into COUPLE and ORIGEN for neutron activation calculations for seven full-power years, followed by a decay period of 100 years.

Selected results presented in Table 2 indicate that:

- The total activity decreases by two orders of magnitude over a period of 50 years after shutdown, with a further 50% reduction in the next 50 years. The core structure and reactor vessel contribute about 90% of the total activity and continue to increase their contributions to 99% after 50 years.

- Fe-55 dominates the activity after the short-lived isotopes decay away in the first year, representing 94% of the total for over 10 years. Then, Ni-63 takes over as the dominant radionuclide after Fe-55 decays away. After about 50 years, Ni-63 and Ni-59 constitute more than 99% of the total activity.

Table 2 Major Contributors to Total Activity for IMSR®400

Component		Decay time (year)					
		0	1	5	10	50	100
	Total Activity (Ci)	7.48E+06	1.93E+06	7.47E+05	2.65E+05	5.45E+04	3.89E+04
	Mass (ton)	Fraction of total (%)					
Core structure	16.577	72	71	71	72	81	81
Reactor vessel	21.840	20	17	17	17	18	18
Guard vessel	43.654	4	11	10	8	<1	<1
Heat exchanger	10.016	3	1	2	3	1	1
Nuclide		Fraction of total (%)					
Fe-55		31	94	88	70	<1	<1
Ni-63		1	4	10	27	99	98
Co-60		<1	1	2	3	<1	<1
Ni-59		<1	<1	<1	<1	1	2

3.3 Westinghouse eVinci® Micro Reactor

The SERPENT model of an eVinci® reactor (Figure 3) [2][5] includes a gamma shield, heat exchangers, neutron shield, reactor vessel, reflector with embedded control drums, and a monolithic core, all housed in a steel outer wall. The core consists of uranium oxide fuel rod channels, metal hydride moderator rod channels, and high-temperature, double-ended sodium filled heat pipes arranged in hexagonal pitches in the solid monolith. The heat pipes are filled with liquid sodium that is effective at moving heat over long-distances with minimal temperature drop, eliminating the need for reactor coolant pumps and other auxiliary systems related to primary reactor cooling. An eVinci® reactor with a 10 MW thermal power and a 10-year life time is modelled in SERPENT.

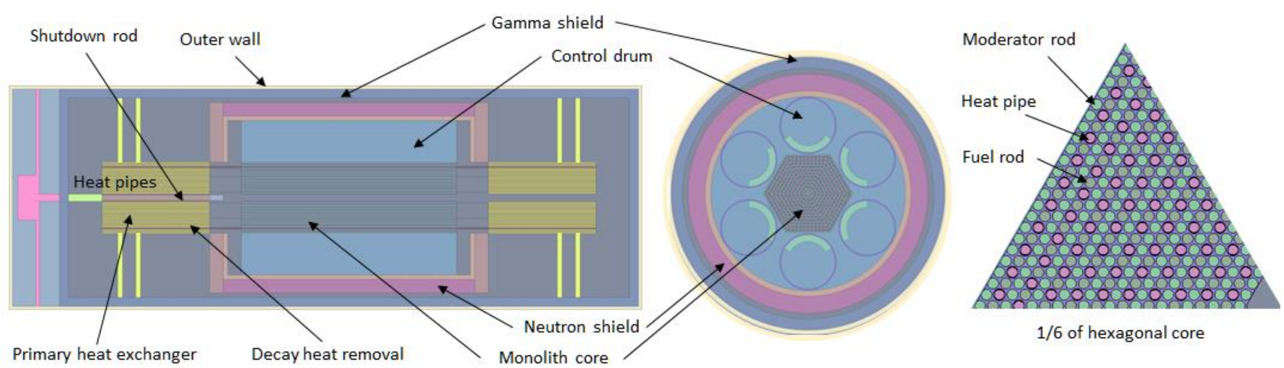

Figure 3. SERPENT model of eVinci®

The beginning of cycle (BOC) is the core with fresh fuel and control drum absorbers facing the core. SERPENT burnup calculations were performed in several burnup steps starting from BOC until the end of cycle (EOC) with control drum absorbers facing away from the core. At each burnup step, burnable fuel material compositions were input into the next burnup step, and control drum absorbers were rotated away gradually from the core to maintain core criticality. Neutron spectra and fluxes were input into COUPLE and ORIGEN for neutron activation calculations for 3600 full-power days followed by a decay period of 100 years.

Selected results presented in Table 3 indicate that:

- The total activity decreases by two orders of magnitude after 10 years of decay, with a further order of magnitude decrease for the period from 50 to 100 years after shutdown. The largest contributor to the total activity is the monolith core, with 62% to 86% contributions at the end of year 10 and 100, respectively. Contributions from heat exchanger and reflector together vary from 18% to 35% for the first 10 years and decrease to be below 10% after 50 years. After 50 years decay, the contributions from heat pipes exceed that from reflector and heat exchangers with contribution increasing from 6 to 9%.

- Fe-55, Co-60, Ni-63 and H-3 are the dominant radionuclides after the short-lived isotopes decay away in the first year representing over 98% of the total activity for the first 10 years. After Fe-55 ($T_{1/2}$=2.737 y) and Co-60 ($T_{1/2}$=5.26 y) decays away in about 10 years, Ni-63 dominates the activity. At the same time, Nb-94 ($T_{1/2}$=2.03E+04 y), produced from activation of Nb-93, starts to be noticeable and continue to become more significant as H-3 ($T_{1/2}$=12.3 y) decays away after 50 years.

Table 3 Major Contributors to Total Activity for eVinci®

Component		**Decay time (year)**					
		0	**1**	**5**	**10**	**50**	**100**
	Total Activity (Ci)	**1.42E+06**	**1.21E+05**	**4.95E+04**	**2.10E+04**	**3.94E+03**	**2.72E+03**
	Mass (ton)	**Fraction of total (%)**					
Heat pipe assembly (heat pipe, wick and liquid sodium)	0.818	36	<1	<1	1	6	9
Monolithic core	4.463	29	80	71	62	84	86
Reflector	12.545	23	2	4	8	4	<1
Heat exchanger	0.513	12	16	22	27	5	4
Nuclide		**Fraction of total (%)**					
Fe-55		8	71	63	42	<1	<1
Co-60		1	15	21	26	1	<1
Ni-63		<1	<1	9	21	87	89
H-3		<1	<1	5	9	5	<1
Nb-94		<1	<1	<1	1	6	9
Ni-59		<1	<1	<1	<1	1	1

3.4 Ultra-Safe MMR

The USNC MMR [2] [6] is an inert gas cooled reactor using fully-ceramic-microencapsulated (FCM) fuel. An MMR consists of three modules (Figure 4) - power reactor module (PRM), power conversion module (PCM) and power generation module (PGM). The PRM houses the reactor core and the reflectors, including reactivity control systems. The monolithic core is ceramic, with FCM fuel compacts and helium gas coolant arranged in hexagonal lattices with a pitch of 0.17 cm. The TRISO fuel particles are randomly distributed within a SiC matrix, doped with Er_2O_3 burnable poisons, for an exact 3,650 particles in each compact which are modelled using the "pbed" feature in SERPENT. Reactor control is provided by 12 control drums of BeO partially filled with B_4C absorber, located in the side reflector and eight in-core control rods, containing B_4C absorber. The MMR is modelled to have 10 MW thermal power and a 12-year life-time.

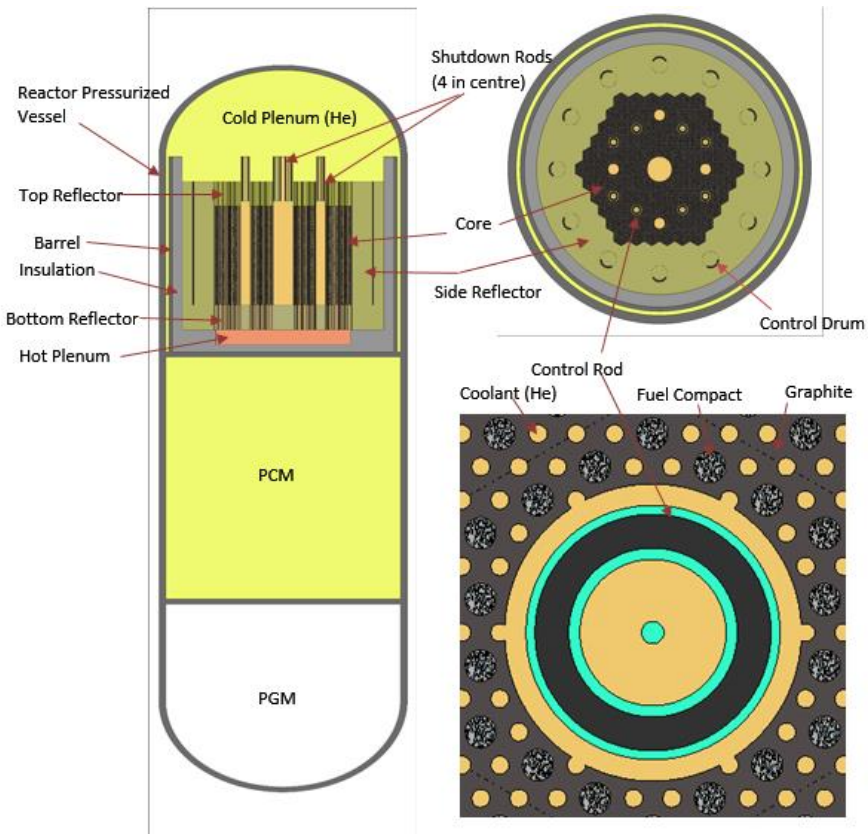

Figure 4. SERPENT Full-Reactor Model of USNC MMR

The SERPENT burnup calculations were first performed to provide isotopic compositions for the fuel and poison-doped matrix at a number of operation time steps. Then, at each time step, the fuel and matrix compositions from the previous step were taken, and the control rod positions were adjusted to maintain core criticality. The neutron spectra and fluxes were input into COUPLE and ORIGEN for neutron activation calculations for 4,000 full-power days followed by a decay period of 100 years.

Selected results given in Table 4 indicate that:

- The total activity decreases by two orders of magnitude over 100 years. The reflectors contribute the most, 75%-92%, to the total activity during the first 50 years. But it is the control drums and control/shutdown rods that prevail with more than 75% of the total activity after 100 years.

- Tritium (H-3, $T_{1/2}$=12.3 y) almost makes up the entire activity of the reflector waste; it would decrease by a factor of 16 after 50 years and, in terms of radioactivity, trail behind Ni-63 ($T_{1/2}$=101.2 y), which is 44 times smaller initially but 3.6 times greater than tritium after 100 years.

Table 4 Major Contributors to Total Activity for MMR

Component		Decay time (year)				
		1	**5**	**10**	**50**	**100**
	Total activity (Ci)	**5.05E+04**	**3.85E+04**	**2.85E+04**	**3.72E+03**	**8.14E+02**
	Mass (tons)	**Fraction of Total (%)**				
Reflectors	39.774	86	90	92	75	22
Control Drums	2.844	11	7	6	22	72
Control rods	0.436	2	1	1	2	4
Shutdown Rods	0.436	<1	<1	<1	1	2
Coolant	0.100	1	1	1	1	<1
Nuclide		**Fraction of Total (%)**				
H-3		88	92	94	76	21
C-14		0.03	0.04	0.1	0.4	2
Fe-55		9	4	2	<1	<1
Ni-63		2	3	4	23	76

3.5 LeadCold SEALER

The SEALER design [2] [7] has the smallest possible core that can achieve criticality in a fast spectrum using 19.75% enriched UO_2 fuel and lead coolant. The core consists of 19 fuel assemblies, 12 control assemblies, 6 shutdown assemblies, 24 reflector assemblies and 24 shield assemblies, all arranged in a hexagonal lattice. Each type of assembly, in turn, consists of a number of pins (*i.e.*, 91 fuel, 19 control, 7 shutdown, 37 reflector, and 19 shield pins) enclosed in a hexagon duct. Each type of pin also contains a stack of type-specific material pellets in a specific metal sheath. The SEALER is modelled to have 8 MW thermal power and a 27-year life-time (Figure 5).

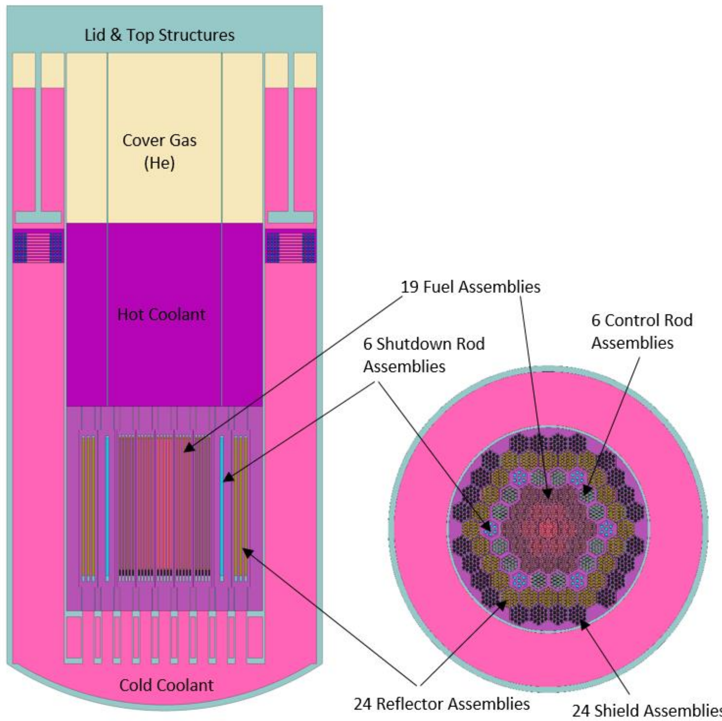

Figure 5. SERPENT full-reactor model of SEALER

First, SERPENT burnup calculations were performed to provide isotopic compositions for the fuel and any burnable materials at a number of operation time steps. Then, at each time step, the fuel and burnable-material compositions from the previous step were input into the next burnup step, and the control rod positions were adjusted to maintain criticality. The neutron spectra and fluxes were input into COUPLE and ORIGEN for neutron activation calculations for 9,000 full-power days followed by a decay period of 100 years.

Selected results given in Table 5 indicate that:

- The total activity of the SEALER waste decreases with decay time, by two orders of magnitude over 100 years. The core assemblies containing boron absorbers contribute half or more, 48%-80%, activity to the total waste during the first 50 years. But it is the steel structures (*i.e.*, barrel, vessel, and core support) which contribute more than 80% of the total activity after 100 years.
- Tritium (H-3), produced from activations of B-10 on fast neutrons, almost makes up the entire activity of the boron-bearing assemblies, contributing 37-70%, to the total SEALER waste activity during the first 50 years after shutdown. Fe-55, produced from activation of Fe-54, is the main contributor of activity during the first few years in all structural materials of SEALER containing iron. However, as Fe-55 has a relatively short half-life, 2.737 years, it is replaced by Ni-63 after 10 years of decay, and it remains to contribute over 79% activity to the total after 100 years.

Table 5 Major Contributors to Total Activity for SEALER

Component		Decay time (years)				
		1	5	10	50	100
	Total activity (Ci)	1.81E+04	9.13E+03	5.28E+03	6.08E+02	1.56E+02
	Mass (tons)	Fraction of Total Activity (%)				
Control Assemblies	1.326	32	42	50	44	10
Shutdown Assemblies	0.333	1	2	2	2	<1
Reflector Assemblies	2.234	35	20	10	0	1
Shield Assemblies	1.302	15	22	28	25	6
Barrel Sections	4.517	4	3	2	6	17
Foot & Diagrid plates	4.929	9	7	5	17	48
Vessel Sections	7.319	5	4	2	6	17
Nuclide		**Fraction of Total Activity (%)**				
H-3		37	59	77	70	16
C-14		0.001	0.002	0.004	0.04	0.1
Fe-55		53	38	19	<1	<1
Ni-63		1	3	4	29	79
Zr-95		2	-	-	-	-
Nb-95		5	-	-	-	-

4. Summary

The neutron activation inventory (Table 6) is determined by many factors such as neutron spectrum and flux level (fission power), fuel and structural material and mass, geometry and operation scheme and schedule, etc.

For the SMRs studied in this paper except USNC MMR, most reactor structures are made of steel-based materials, which usually have iron and nickel as major elements. In general, Fe-55, produced from neutron activation of Fe-54, is the dominating contributor to the total activity for the first 10 years or so after the short-lived isotopes decay away in the first year. Due to its short half-life of 2.737 years, Fe-55 hardly remains after 10 years. Ni-63 ($T_{1/2}$=101 years), produced from neutron activation of Ni-62, then becomes the dominating contributor to the total activity afterwards. For the USNC MMR, the massive BeO reflector produces a significant amount of H-3 ($T_{1/2}$=12.32 years) that dominates the activity for 50 years before Ni-63 becomes dominant.

Table 6 Summary of total neutron activation waste activity for selected SMRs

Small Modular Reactor	Mass (ton)	Thermal Power (MW)	Life Time (year)	Total Activity (MCi)					
				0	1	5	10	50	100
NPM	547	160	60	3.58	0.459	0.204	0.108	0.053	0.0379
IMSR	166	400	7	7.48	1.93	0.747	0.265	0.0545	0.0389
eVinci	119	8	10	1.42	0.121	0.0495	0.021	0.00394	0.00272
MMR	93	10	12	0.662	0.0505	0.0385	0.0285	0.00372	0.00081
SEALER	268	8	27	0.22	0.0181	0.00913	0.00528	0.00060	0.00015

Accuracies associated with estimates of SMR waste inventories may be improved through further studies and analyses. Increased confidence in waste inventory estimates will benefit future studies related to SMR safety, radioactive waste management, cost estimation and environmental impact assessments. Specific improvements achievable in the short term are as described below.

- Simulation uncertainties for structures further from the core may be reduced by using additional neutron histories and by employing special modelling techniques.
- The scope of SMR waste inventory assessments may be broadened to include fuel depletion and inventory estimates.
- Neutronic simulations of molten salt fuel flowing in IMSRs may be improved by including two fuel effects not previously modelled: fuel flow and reactivity-temperature sensitivity.

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