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Developing a Method to Quantitatively Screen Advanced Reactor Radionuclides Using a Heat Pipe Reactor Inventory

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Outline

- Introduction and Background
- Screening Methodology
- Important Findings
- Conclusions, Limitations and Opportunities for Future Work

WASH-1400 identifies a subset of radionuclides to be included in a consequence analysis

- Section 3.3 and 8.2.1 of Appendix VI
- This methodology includes consideration of:
  - Radionuclide half-life
  - Emitted radiation type and energy
  - Inventory
  - Release fraction
  - Elemental chemistry

Alpert et al. (1986) updated the list of radionuclides identified in WASH-1400

- Estimation of release fractions was subject of considerable uncertainty at the time
- Alpert et al. developed a method to consider relative importance of individual elements to reactor accident consequences assuming equal release fractions
- Ultimately resulted in a list of 60 radionuclides
- SOARCA updated this list even further
  - Explicitly includes short lived decay progeny
  - 71 radionuclides in total identified for LWR consequence analysis

Table 1.

60 Nuclides Used for Offsite Consequence Assessment in MACCS

Element	Nuclides
Cobalt	58 60
Krypton	85 85m 87 88
Rubidium	86
Strontium	89 90 91 92
Yttrium	90 91 92 93
Zirconium	95 97
Niobium	95
Molybdenum	99
Technetium	99m
Ruthenium	103 105 106
Rhodium	105
Antimony	127 129
Tellurium	127 127m 129 129m 131m 132
Iodine	131 132 133 134 135
Xenon	133 135
Cesium	134 136 137
Barium	139 140
Lanthanum	140 141 142
Cerium	141 143 144
Praseodymium	143
Neodymium	147
Neptunium	239
Plutonium	238 239 240 241
Americium	241
Curium	242 244

* The 60 nuclides are the same as those examined in WASH-1400 [1] and in the CRAC2 computer code [9] except for the addition of six nuclides: Sr-92, Y-92, Y-93, Ba-139, La-141, and La-142.

Alpert et al., 1986

71 radionuclides were identified as important for LWRs based on inventory, half-life and potential biological hazard of radionuclides expected to be present in a large LWR

Chemical Group	Isotope	$T_{1/2}$
Noble Gas	Kr-85	10.72 yr
	Kr-85m	4.48 hr
	Kr-87	76.3 min
	Kr-88	2.84 hr
	Xe-133	5.25 d
	Xe-135	9.09 hr
	Xe-135m	15.3 min
Alkali Metals	Rb-86	18.7 d
	Rb-88	17.8 min
	Cs-134	2.062 yr
	Cs-136	13.1 d
	Cs-137	30.0 yr
Alkaline Earths	Sr-89	50.5 d
	Sr-90	29.1 yr
	Sr-91	9.5 hr
	Sr-92	2.71 hr
	Ba-137m	2.55 min
	Ba-139	82.7 min
	Ba-140	12.74 d
Halogens	I-131	8.04 d
	I-132	2.30 hr
	I-133	20.8 hr
	I-134	52.6 min
	I-135	6.61 hr

Chemical Group	Isotope	$T_{1/2}$
Chalcogens	Te-127	9.35 hr
	Te-127m	109 d
	Te-129	69.6 min
	Te-129m	33.6 d
	Te-131	25.0 min
	Te-131m	30.0 hr
	Te-132	78.2 hr
Platinoids	Ru-103	39.3 d
	Ru-105	4.44 hr
	Ru-106	368.2 d
	Rh-103m	56.1 min
	Rh-105	35.4 hr
	Rh-106	29.9 sec
Early Transition Elements	Co-58	70.8 d
	Co-60	5.271 yr
	Nb-95	35.1 d
Early Transition Elements	Nb-97	72.1 min
	Nb-97m	1.0 min
	Mo-99	66.0 hr
	Tc-99m	6.02 hr
Tetravalents	Zr-95	64.0 d
	Zr-97	16.9 hr
	Ce-141	32.5 d
	Ce-143	33.0 hr
	Ce-144	284.3 d
	Np-239	2.35 d
	Pu-238	87.74 yr
	Pu-239	2.41E4 yr
	Pu-240	6.54E3 yr
	Pu-241	14.4 yr

Chemical Group	Isotope	$T_{1/2}$
Trivalents	Y-90	64.0 d
	Y-91	58.5 d
	Y-91m	49.7 min
	Y-92	3.54 hr
	Y-93	10.1 hr
	La-140	40.3 hr
	La-141	3.9 hr
	La-142	92.5 min
	Pr-143	13.56 d
	Pr-144	17.3 min
	Pr-144m	7.2 min
	Nd-147	11.0 d
	Am-241	432.2 y
	Cm-242	162.8 d
	Cm-244	18.11 yr
Cadmium Group	Sb-127	3.85 d
	Sb-129	4.32 hr

Source: State of the Art Reactor Consequence Analysis (SOARCA)

Advancing reactor technology has motivated an investigation into developing a similar subset of radionuclides relevant to advanced non-LWRs

- High-temperature gas reactors (HTGR)
- Fluoride-salt-cooled high-temperature reactor (FHR)
- Molten-salt reactors (MSR)
- Sodium fast reactor (SFR)
- Liquid metal fast reactors (LMR)

The NRC Vision and Strategy Document (Vol. 3) outlines a radionuclide screening effort

- Calls for the identification of a subset of radionuclides to be included in MACCS calculations for non-LWRs
- Radionuclide selection should be based upon factors such as:
  - Core inventory
  - Nature of radioactivity
  - Specific organ effects
- Expand upon previous qualitative efforts to screen advanced reactor radionuclides in *Preliminary Radioisotope Screening for Off-site Consequence Assessment of Advaned Non-LWR Systems* (Andrews et al., 2021)
  - Developed a preliminary, qualitative list of radionuclides for these reactors (57 radionuclides)
  - Identified knowledge gaps that still exist regarding reactor chemistry and system behavior

57 radionuclides identified in preliminary qualitative screening assessment

Chemical Group	Isotope	$T_{1/2}$	Reactor Type
New Proposed Group	H-3	12.3 y	HTGR, FHR, MSR, SFR
	C-14	5,730 y	HTGR, FHR
Alkali Metals	Na-22	2.6 y	SFR
	Na-24	15 h	SFR
Alkaline Earths	Ra-224	3.66 d	MSR
Noble Gas	Ar-41	110 m	SFR
	Kr-83m	1.83 hr	MSR
	Xe-131m	11.9 d	MSR
	Xe-133m	2.2 d	MSR
Early Transition Elements	Cr-51	27.7 d	SFR
	Mn-54	312.3 d	SFR
	Fe-59	44.5 d	SFR
	Nb-93m	16.13 yr	MSR
	Ta-182	114.4 d	SFR
Cadmium Group	As-77	38.5 hr	MSR
	Cd-113m	14.1 yr	MSR
	Cd-115m	44.5 d	MSR
	Sb-125	2.8 y	HTGR, FHR
	Sb-126	12.3 d	MSR
	Sb-128	9.01 hr	MSR
Chalcogens	Se-81	18.4 m	MSR
	Se-81m	57.3 m	MSR
	Se-83	22.3 m	MSR
	Te-125m	57.5 d	MSR
	Te-133m	55.4 min	MSR
	Te-134	41.8 min	MSR

Chemical Group	Isotope	$T_{1/2}$	Reactor Type
Halogens	Br-83	2.4 hr	MSR
	Br-84	31.8 min	MSR
Platinoids	Pd-109	13.7 h	MSR
	Pd-112	21.0 hr	MSR
Tin Group	Ag-110m	250 d	HTGR, FHR
	Ag-111	7.45 d	MSR
	Sn-117m	13.7 d	MSR
	Sn-119m	293 d	MSR
	Sn-121m	43.9 yr	MSR
	Sn-123	129 d	MSR
Trivalents	Pr-146	24.2 hr	MSR
	Pm-147	2.6 y	HTGR, FHR, MSR
	Pm-148m	41.3 d	MSR
	Pm-149	53.1 hr	MSR
	Pm-151	28.4 hr	MSR
	Sm-151	88.8 y	HTGR, FHR, MSR
	Sm-153	46.3 hr	MSR
	Eu-154	8.6 y	HTGR, FHR, MSR
	Eu-155	4.8 y	HTGR, FHR, MSR
	Eu-156	15.2 d	MSR
	Eu-157	15.2 hr	MSR
	Cm-243	29 y	MSR, LMR
	Cm-245	8,500 y	HTGR, FHR, MSR, LMR
	Cm-246	4700 y	MSR, LMR
	Am-242m	150 y	MSR, LMR
	Am-243	7400 y	MSR, LMR

Chemical Group	Isotope	$T_{1/2}$	Reactor Type
Tetravalents	Th-228	1.91 y	MSR
	Pa-233	27.0 d	MSR, MSR, LMR
	Pu-242	373,300 y	HTGR, FHR, MSR, LMR
Uranium Group	U-232	68.9 y	MSR
	U-237	6.75 d	MSR, LMR

Source: Andrews et al., 2021. *Preliminary Radioisotope Screening for Off-site Consequence Assessment of Advanced Non-LWR Systems*, SAND2021-11703

Advanced reactor research is still underway, but some preliminary information does exist

- Information on half-life and potential biological hazard for 825 radionuclides in MACCS is available
- Still need reliable information for advanced reactors:
  - Inventories
  - Transport pathways
  - Chemistries
- Some preliminary inventories are available
  - INL Heat Pipe Reactor Design
  - See Walker et al. (2022) *SCALE Modeling of the Fast-Spectrum Heat Pipe Reactor*
- Available inventories allow us to illustrate a method that can be applied to identify a list of radionuclides for any advanced reactor technology, provided that a quantitative inventory is available
  - Method analogous to Alpert et al. to estimate relative importance assuming equal release fraction
  - Identify "most important" contributors based on relative importance
  - Consider doses to multiple organs, scaled to that of I-131 (early phase) and Cs-137 (long-term phase)

MACCS 4.1 was used to assess the relative importance of advanced reactor radionuclide suite

- **Step 1**: calculate an activity-normalized dose of combined list of 57 preliminary radionuclides and heat pipe reactor suite
  - Screen heat pipe reactor by eliminating radionuclides with short half-lives (<1 hour) and low contribution to the total inventory (<0.0001%)
    - > 1200 radionuclides reduced to 108
  - EARLY and CHRONC doses from a unit 1-Ci release were modeled for each of these radionuclides and normalized to equivalent releases of I-131 and Cs-137, respectively
  - In this manner, a relative biological hazard list was developed
- **Step 2**: illustrate using a heat pipe reactor inventory to scale these "hazard rankings" by the inventory (relative to I-131 or Cs-137 as appropriate)
  - Identify radionuclides that, if released in sufficient quantities, may be important to early or long -term dose
- **Step 3**: additionally screen this list by eliminating radionuclides with effective and organ doses less than 1% of those of I-131 (early phase) and Cs-137 (late phase doses)

The MACCS assumptions used for this analysis mirrored those in previous studies

- L-ICRP60ED dosimetric quantity used as surrogate for potential latent health effects from both EARLY and CHRONC phase doses. A-RED MARR and A-LUNG used for surrogates of early health effects from early phase doses
- Constant, “typical” weather conditions – D stability, 4 m/s windspeed, no rain
- Release occurs outside of the growing season
- Doses from elements/isotopes include the effect of radioactive decay and in-growth or decay progeny during transport.
- Nonbuoyant release from a single plume at 40 m elevation. Uniform 1-hour release immediately after accident initiation
- 0.002 m/sec dry dep. velocity, radiation protection factors of 0.75, 0.22, and 0.46 for cloudshine, groundshine, and inhalation and skin, respectively
- Uniform population distribution of approximate CONUS average
- No emergency protective actions, exposure duration of 7 days for EARLY, CHRONC duration of 1 year, no intermediate phase

Analysis suggests that 69 heat pipe reactor radionuclides may be of importance if released in sufficient quantities

- 48 of these radionuclides are already considered for LWR analyses
- 21 new radionuclides listed here
- Note: decay progeny not listed here

Isotope	EARLY Relative ICRP60 Effective Dose	EARLY Relative Red Marrow Dose	EARLY Relative Lung Dose	CHRONC Relative ICRP60 Effective Dose	CHRONC Relative Red Marrow Dose	CHRONC Relative Lung Dose
Ag-111			0.04			
Ag-112			0.01			
Cd-115			0.03			
Eu-155			0.017			
Eu-156		0.031	0.063			
Nb-95m			0.06			
Nd-149		0.014	0.22			
Pd-109			0.09			
Pm-147	0.34		1.57			0.03
Pm-148m			0.015			
Pm-149	0.035		0.97			
Pm-151	0.012	0.043	0.396			
Pr-145	0.03		1.58			
Sb-125		0.02	0.017	0.04	0.04	0.04
Sm-153			0.173			
Sn-121			0.011			
Sn-125			0.0301			
Sn-127			0.028			
Te-125m			0.0107			
U-234	0.40		0.663			0.03
U-237	0.035	0.041	0.663			

Summary and Limitations

- A method for the identification of radionuclides of potential for advanced reactors – based on half-life, biological hazard, and relative abundance in a core – is provided and illustrated using a radiological inventory developed for a heat pipe reactor
  - Provide a traceable and transparent basis for selecting radionuclides for inclusion in advanced reactor consequence analysis
- Radionuclides were progressively screened, first based on half-life and relative inventory, and then further based upon relative biological hazard to develop a list of 21 new radionuclides to consider for the heat pipe reactor
- In theory, this method can be applied to any advanced reactor inventory as they become available
- Some radionuclides did not have dose coefficients in MACCS
- Complexities of H-3 and C-14 are generally unaccounted for in MACCS
- Food ingestion ignored
- Normalizing to doses of volatile isotopes of iodine and cesium means that large releases not associated with high elemental volatility may need to be reassessed
- Other thresholds for half-life, relative abundance, or relative biological hazard may be used

Thank you!

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References

United States Nuclear Regulatory Commission (US NRC) 2019. NRC Non-Light Water Reactor (Non-LWR) Vision and Strategy: Volume 3: Computer Code Development Plans for Severe Accident Progression, Source Term, and Consequence Analysis.

WASH-1400 Reactor Safety Study: An Assessment of Accident Risks in U.S. Commercial Nuclear Power Plants. NUREG-75/014.

Alpert, D.J., Chanin, D.I., Ritchie, L.T. 1986. Relative Importance of Individual Elements to Reactor Accident Consequences Assuming Equal Release Fractions. NUREG/CR-4467, SAND85-2575.

Chang, R., Schaperow, J., Ghosh, T., Barr, J., Tinkler, C., Stutzke, M. November 2012. State-of-the-Art Reactor Consequence Analysis (SOARCA) Report. NUREG-1935

Andrews, N.C., Higgins, M., Taconi, A., Leute, J. 2021. Preliminary Radioisotope Screening for Off-site Consequence Assessment of Advanced Non-LWR Systems, SAND2021-11703

2022 SCALE Non-LWR Models for NRC Volume 3. https://code.ornl.gov/scale/analysis/non-lwr-models-vol3

Walker, E., Skutnik, S., Wieselquist, W., Shaw, A., Bostelmann, F., 2022. SCALE Modeling of the Fast-Spectrum Heat Pipe Reactor. ORNL/TM-2021/2021

Nosek, A.J., Bixler, N. 2021. MACCS Theory Manual. SAND2021-11535

Naegeli, R.E. 2003. A MACCS2 Single Nuclide Downwind Dose Database for Sandia National Laboratories, Technical Area V. SAND2003-0883

Leute, J., Walton, F., Mitchell, R., Eubanks, L. 2021. MACCS (MELCOR Accident Consequence Code System) User Guide – Version 4.0. SAND2021-8998