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Joseph A. Angelo Jr. & Roy G. Post

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NUCLEAR FUEL CYCLE AND THE ISOTOPIC COMPOSITIONS

RADIOACTIVE WASTE

KEYWORDS: *after-heat, spent fuel elements, isotope ratio, radioactive waste processing, HTGR-type reactors, PWR-type reactors*

JOSEPH A. ANGELO, Jr. *Florida Institute of Technology, Melborrne, Florida 32901*

ROY G. POST *University of Arizona, Department of Nuclear Engineering Tucson, Arizona 85721*

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The heat generated by spent fuel elements and typical processing waste from both a 1000 MW(e) reference design pressurized water reactor (PWR) and 1160 MW(e) reference design high temperature gas-cooled reactor (HTGR) were calculated for times up to 1000 years. To compensate for differences in exposure, the heat generated was expressed in terms of watts of heat generated per megawatt day of exposure. Examination of both tabular data and graphical presentations of these normalized heat generation data indicates noticeable differences in the contribution of different isotopes for each system. As anticipated, the heat generation for each fuel was greatly influenced by the transmuted isotopes ^{233}Pa and ^{238}Pu for the HTGR with ^{137}Cs and ^{90}Sr for the PWR. Data provide quantitative detailed information on the thermal power output of typical processing waste for both reactor systems for the first millennium of cooling.

Composition of spent fuel and nuclear fuel reprocessing waste is an important quantity for optimizing the fuel processing, the partitioning of the nuclei, and the selection of waste management techniques. Most of the earlier work has been well served by broader treatments of the radiation levels and heat generation rates as functions of time. More detailed data have been needed to understand and evaluate the potential for partitioning nuclides and to predict the thermal history of the waste. The concept for deep burial of nuclear waste in nuclear excavated cavities reported by Cohen et al.[1] raised a number of interesting possibilities. The immediate programmatical interest was to perform more sophisticated heat transfer calculations, and a primary data requirement was the heat generation data in great detail for different nuclear waste materials. A program was established at the University of Arizona to calculate these quantities using existing codes to the maximum extent possible.[2] Some of these data are of interest to many fuel reprocessing and waste management systems, so the following work has been presented to provide information of general interest concerning most of these processes and systems. The present nuclear energy generation is dominated by light-water reactors, but high temperature gas-cooled reactors are expected to assume an increasingly important role over the next 10 to 20 years. The first major effort for control or management of nuclear waste will be directed to the by-products from these operations. For these reasons we have elected to present data for typical thermal reactors.

Projections of the installed nuclear electrical capacity for the United States, in Fig. 1, clearly show magnitude and importance of these thermal reactors.[3] There are a number of designs of light-water reactors and a wide range of sizes. A 1000 MW(e) reference design pressurized water reactor[4] (PWR) was chosen as typical for this study. The recent investigations[5] have concluded that the high level waste from boiling water reactors will closely approximate those from a PWR with equal exposure. The design and performance characteristics for this reference PWR are shown in Table I.

The high temperature gas-cooled reactor (HTGR) chosen for this study is a reference design based on studies at Oak Ridge National Laboratory,[5] and the design performance characteristics of this reference system are given in Table II.

To provide the detail and accuracy necessary for design, calculations were made on the changes in isotope concentration due to the production of isotopes by fissioning, neutronic transmutation, and as daughters of other radioisotopes. The code also must take into consideration the decrease in isotopes due to neutron absorption and radioactive decay. In the treatment of the waste from fuel reprocessing, certain other assumptions are made regarding the chemical processing of various isotopes. Some remain completely unchanged, some remain in part, and some are totally removed. The ORIGEN isotope generation and depletion code[6] was augmented by the RADEC heat generation code, developed at the University of Arizona,[2] to provide data on the contribution of fission product, actinide, and cladding nuclides to the heat generation as well as that of various specific isotopes. These contributions were calculated for decay times up to 10 years for the spent fuel and extended out to 1000 years for the processing wastes from the fuel. The heat generation characteristics of the spent fuel for the first 10 years are considered to be of primary interest to fuel reprocessing and waste management schemes.

The data are presented in two forms beginning with Table III. This table presents the total thermal power output for PWR spent fuel normalized to exposure. Table IV presents the same data for the HTGR. Tables V and VI present data

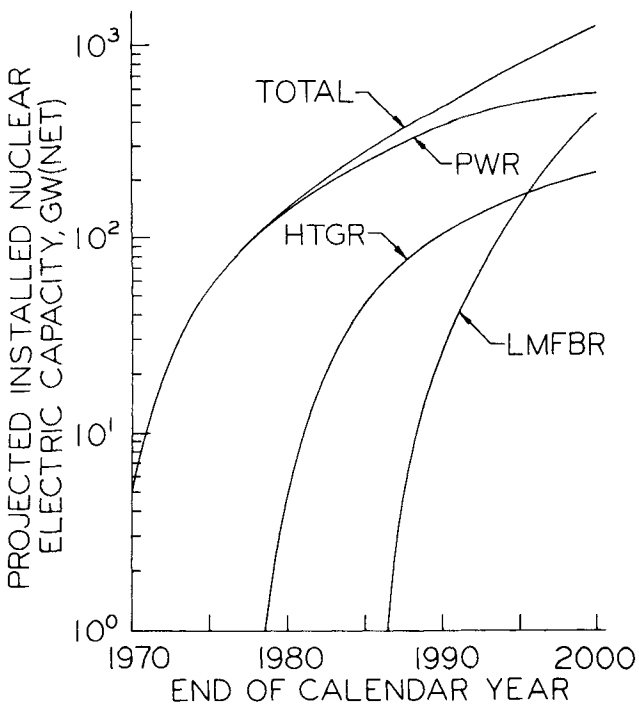

Fig. 1. Projected U.S. nuclear electric economy.

TABLE I

PWR Design and Performance Characteristics

Power	3083 MW(th), 1000 MW(e)
Fuel form	oxide pellets
Enrichment (^{235}U)	3.3%
Plutonium recycle	no
Average specific power (full charge, full power)	37.5 MW/t
Average burnup	33 000 MWd/t
Refueling interval (at 80% capacity)	~365 days
Refueling fraction	$\frac{1}{3}$

TABLE II

HTGR Design and Performance Characteristics

Power	3000 MW(th) 1160 MW(e)
Fuel form	coated particles in hexagonal graphite blocks
Fuel	Th/^{235}U (93% enriched)/ ^{233}U (recycle)
^{233}U recycle	yes (assumed to begin at third reload)
Plutonium recycle	no
Average specific power (full charge, full power)	80.7 MW/t
Average burnup	94 271 MWd/t
Refueling interval (at 80% capacity)	365 days
Refueling fraction	$\frac{1}{4}$

TABLE III

PWR Spent Fuel Total Thermal Power Output
(Burnup = 33 000 MWd/MT)

Time After Discharge (day)	Total Afterheat	
	(W/T)	(W/MWd)
0	1.595 *E* + 06	4.834 *E* + 01
10	7.868 *E* + 04	2.384 *E* + 00
30	4.913 *E* + 04	1.489 *E* + 00
60	3.509 *E* + 04	1.063 *E* + 00
90	2.841 *E* + 04	8.609 *E* - 01
120	2.417 *E* + 04	7.324 *E* - 01
150	2.105 *E* + 04	6.378 *E* - 01
270	1.392 *E* + 04	4.219 *E* - 01
365	1.108 *E* + 04	3.356 *E* - 01
1096	3.799 *E* + 03	1.151 *E* - 01
3650	1.290 *E* + 03	3.909 *E* - 02

TABLE IV

HTGR Spent Fuel Total Thermal Power Output
(Burnup = 94 271 MWd/MT)

Time After Discharge (day)	Total Afterheat	
	(W/MT)	(W/MWd)
0	3.688 *E* + 06	3.912 *E* + 01
10	2.148 *E* + 05	2.279 *E* + 00
30	1.342 *E* + 05	1.424 *E* + 00
60	8.700 *E* + 04	9.229 *E* - 01
90	6.513 *E* + 04	6.909 *E* - 01
120	5.232 *E* + 04	5.550 *E* - 01
150	4.392 *E* + 04	4.659 *E* - 01
270	2.765 *E* + 04	2.933 *E* - 01
365	2.208 *E* + 04	2.342 *E* - 01
1096	9.285 *E* + 03	9.849 *E* - 02
3650	4.057 *E* + 03	4.304 *E* - 02

TABLE V

Fractional Contribution to Total PWR
Spent Fuel Afterheat

Time After Discharge (day)	Percent of Total Afterheat		
	Cladding (%)	Actinides (%)	Fission Products (%)
0	0.04	4.68	95.28
10	0.63	3.80	95.57
30	0.83	2.69	96.48
60	0.91	3.33	95.76
90	0.92	3.70	95.38
120	0.89	3.94	95.17
150	0.87	4.11	95.02
270	0.85	4.35	94.80
365	0.90	4.30	94.80
1096	1.81	6.05	92.14
3650	2.09	17.29	80.62

on the relative contribution of cladding, actinides, and fission products to the total heat generation for periods up to 10 years. Considerations of the heat generation data in Table V show that ~1% of the total heat, generated up to 1000 days after discharge, is due to the activated cladding nuclides, ^{58}Co, ^{95}Zr-Nb, and ^{60}Co. The cladding nuclides contribute a negligible fraction of the total heat generation at discharge, but their contribution rises to about 2% of the total at the end of the first decade. Actinide nuclides from PWR spent fuel exhibit a similar pattern, contributing some 3 or 4% for the first year of decay and then becoming more significant by the end of the first 10 years of cooling.

The HTGR spent fuel has a significantly different behavior. There is no significant contribution to the heat evolution of spent HTGR fuel from activated nuclides in the fuel coating or structural materials, but the actinide nuclides in this fuel contribute a larger fraction of the total heat between 10 and 150 days after discharge than they do in the PWR. The primary contributor is ^{233}Pa. Its contribution to the heat generation begins at almost 6% at discharge, rising to 24% in 10 days, dropping to 3% in 270 days, and finally reaching 17% again after 10 years. Contributions from various isotopes to the total heat generation of spent fuel from PWRs, as they vary with time up to 10 years, are presented in Table VII. Similar data for HTGR spent fuel are contained in Table VIII. Although the behavior is not unexpected from consideration of their nuclear properties, the exact relative importance of the various isotopes is such a complex function of the secular equilibria and other factors that a detailed calculation is necessary to define quantitatively the exact relationships among the various isotopes.

TABLE VI

Fractional Contributions to Total
HTGR Spent Fuel Afterheat

Time After Discharge (day)	Percent of Total Afterheat		
	Cladding[a]	Actinides (%)	Fission Products (%)
0	0	5.90	94.10
10	0	23.65	76.35
30	0	22.50	77.50
60	0	16.67	83.33
90	0	11.10	88.90
120	0	7.30	92.70
150	0	5.06	94.94
270	0	3.08	96.92
365	0	3.53	96.47
1096	0	7.92	92.08
3650	0	17.18	82.82

[a]For example, negligible thermal significance when compared to fission product or actinide contribution.

The convenient basis of comparison is simply the relative contribution, expressed as percent, for each isotope or closely coupled isotopic decay chain through the first year of decay. The total heat generated by waste from processing PWR and HTGR fuels is shown in Fig. 2. The major differences in the nuclear chemistry of the PWR and HTGR fuel cycles, the ^{232}Th-^{233}U in the HTGR and ^{238}U-^{239}Pu for the PWR, lead to significant differences in chemical composition of the radionuclides that survive over long periods of time in the two reactor systems. The fission-produced

TABLE VII

Thermally Significant Isotopes in PWR Spent Fuel
(Contribution to heat generation, %)

Isotopes	Time After Discharge (day)									
	10	30	60	90	120	150	270	365	1096	3650
$^{140}Ba/^{140}La$	25.32	13.79	3.81	0.93						
$^{95}Zr/^{95m}Nb/^{95}Nb$	16.62	23.24	25.74	24.52	21.85	18.80	8.43	3.92		
$^{144}Ce/^{144}Pr$	11.82	18.02	23.44	26.89	29.41	31.36	35.42	35.27	17.29	
$^{106}Ru/^{106}Rh$	7.17	11.08	14.65	17.10	18.97	20.59	24.85	26.05	19.11	
$^{103}Ru/^{103}Rh$	4.90	5.54	4.59	3.35	2.33	1.58				
^{91}Y	4.07	5.13	5.04	4.37	3.62	2.92	1.07	0.44		
^{134}Cs	3.29	5.17	7.04	8.45	9.68	10.78	14.58	16.79	24.90	6.90
^{89}Sr	2.87	3.52	3.31	2.74	2.16	1.66	0.51			
^{141}Ce	2.82	2.95	2.17	1.41	0.87	0.53				
^{239}Np	1.68									
^{242}Cm	1.50	2.22	2.72	2.96	3.06	3.10	2.81	2.36		
$^{147}Nd/^{147}Pm$	1.37	0.71								
$^{137}Cs/^{137m}Ba$		1.16	1.63	2.00	2.35	2.69	4.04	5.05	14.08	35.27
$^{90}Sr/^{90}Y$		1.13	1.59	1.96	2.30	2.63	3.94	4.92	13.65	33.84
^{238}Pu			0.26	0.33	0.39	0.44	0.68	0.85	2.49	6.94
^{60}Co						0.45	0.65	0.79	1.77	2.07
^{244}Cm							0.59	0.74	1.99	4.48
^{154}Eu							0.40	0.50	1.33	2.88
^{241}Am										3.63
^{240}Pu										1.15
^{239}Pu										0.78

isotopes, starting with ^{95}Zr (Fig. 3) and ^{140}Ba (Fig. 4), have approximately the same contribution to the heat generation rate for both reactor systems. However, as shown in Fig. 5, the radionuclides $^{106}Ru/^{106}Rh$ have a much greater contribution in spent PWR fuel than in HTGR fuel. On the other hand, Figs. 6 and 7 show that the radionuclides ^{134}Cs and $^{144}Ce/^{144}Pr$ have greater contributions in the HTGR. The larger amounts of ^{235}U-^{236}U in HTGR results in the ^{238}Pu having a much greater contribution to the total heat generation in the HTGR than in the PWR fuel, as shown in Fig. 8. Of course, ^{233}Pa is unique to HTGR spent fuel, and PWR fuel has a significant heat generation from the ^{242}Cm, as shown in Fig. 9. There are other differences in the fuel which can be seen by studying the tables.

Regarding the behavior of the heat generation in the waste left after processing each of the fuels, we can make a meaningful comparison by using the two reference reactors and assuming that each of the fuels is processed one year after discharge. Although there are a number of fuel reprocessing techniques, the comparisons can be made with the following assumptions which should be met by almost all those processes. All the tritium, krypton, xenon, 99.9% of the bromine and iodine, and 99.5% of the uranium and plutonium are removed. Thus, the typical waste from the PWR would contain all the fission products, all the actinide nuclides except 0.5% of the uranium and plutonium originally present in the spent PWR fuel. With the same assumptions for the HTGR, the waste would contain again all the fission products except those noted and all the actinide nuclides except 0.5% of uranium and thorium originally present in the spent fuel. The heat generation from these two systems is shown in Fig. 10. The typical PWR

TABLE VIII
Thermally Significant Isotopes in HTGR Spent Fuel
(Contribution to heat generation, %)

Isotopes	Time After Discharge (day)									
	10	30	60	90	120	150	270	365	1096	3650
^{233}Pa	22.53	21.76	15.36	9.81	5.71	3.19	0.24			
$^{140}Ba/^{140}La$	18.97	10.22	3.12	0.82	23.83	21.29	10.01	4.64		
$^{95}Zr/^{95m}Nb/^{95}Nb$	14.33	20.02	24.47	25.23						
$^{144}Ce/^{144}Pr$	9.99	15.24	21.84	27.15	31.29	34.75	41.19	40.85	16.32	
^{91}Y	4.70	5.97	6.46	6.06	5.29	4.44	1.71	0.70		
^{89}Sr	4.30	5.27	5.45	4.88	4.07	3.26	1.05	0.37		
^{134}Cs	3.86	6.07	9.10	11.84	14.34	16.60	23.58	27.04	32.74	7.03
^{141}Ce	2.73	2.85	2.31	1.63	1.07	0.67				
^{143}Pr	2.12	1.24								
$^{103}Ru/^{103m}Rh$	1.78	2.01	1.83	1.45	1.07	0.75				
$^{90}Sr/^{90}Y$	0.98	1.56	2.40	3.21	3.98	4.74	7.49	9.24	21.00	40.42
$^{106}Ru/^{106}Rh$	0.91	1.41	2.06	2.60	3.06	3.44	4.38	4.53	2.71	
$^{137}Cs/^{137m}Ba$	0.75	1.21	1.86	2.49	3.08	3.67	5.75	7.20	16.37	31.80
^{238}Pu	0.29	0.46	0.72	0.96	1.19	1.42	2.24	2.80	6.57	14.22
^{154}Eu								0.49	1.06	1.80
^{244}Cm										0.98

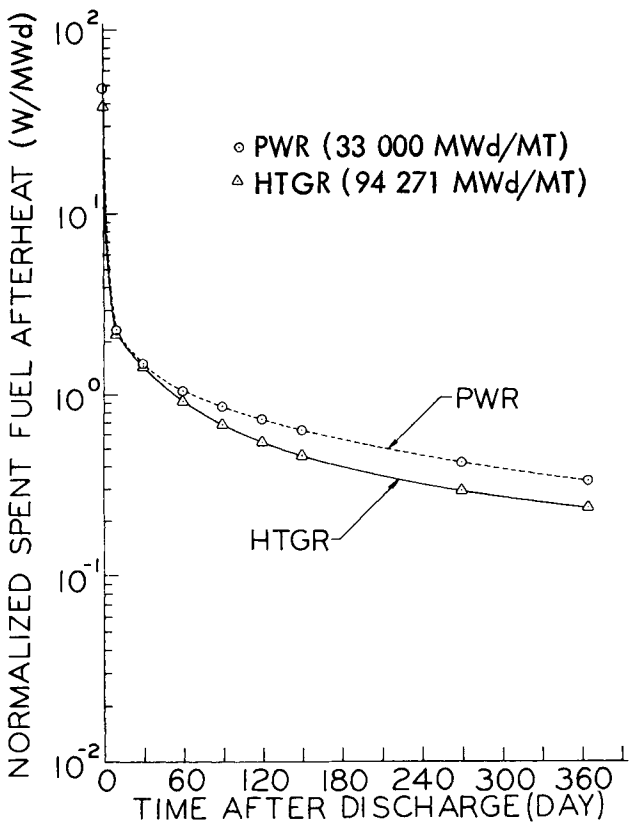

Fig. 2. Normalized heat generation by PWR and HTGR spent fuel.

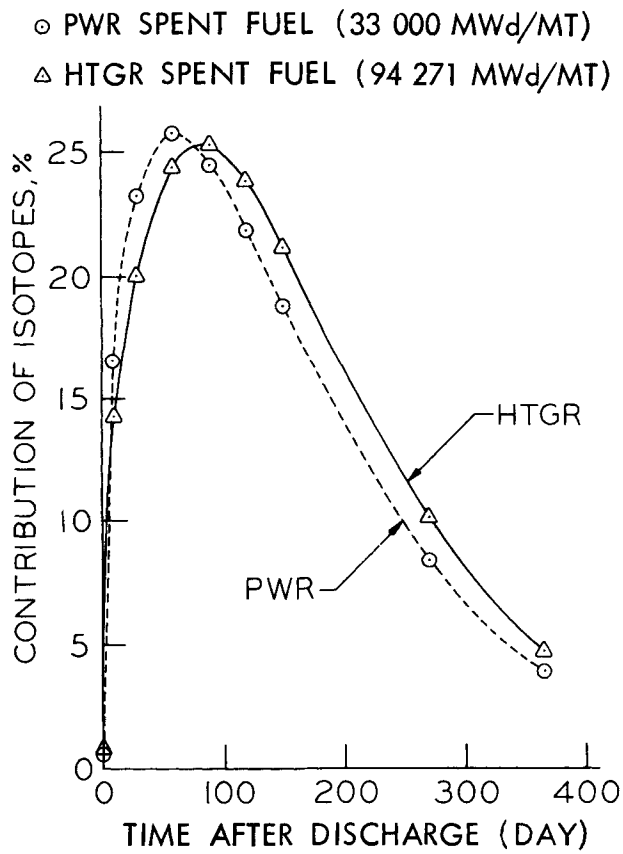

Fig. 3. Relative heat generation of $^{95}Zr/^{95m}Nb/^{95}Nb$.

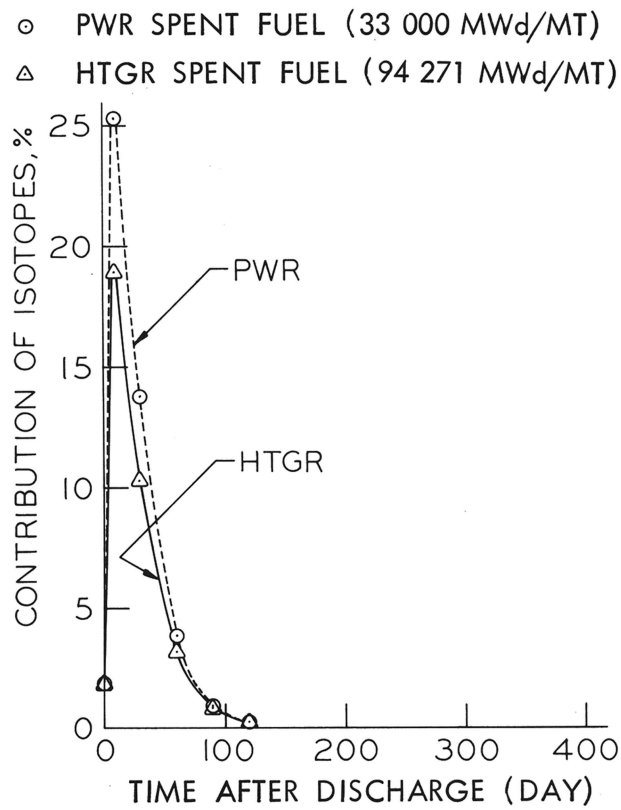

Fig. 4. Relative heat generation of $^{140}Ba/^{140}La$.

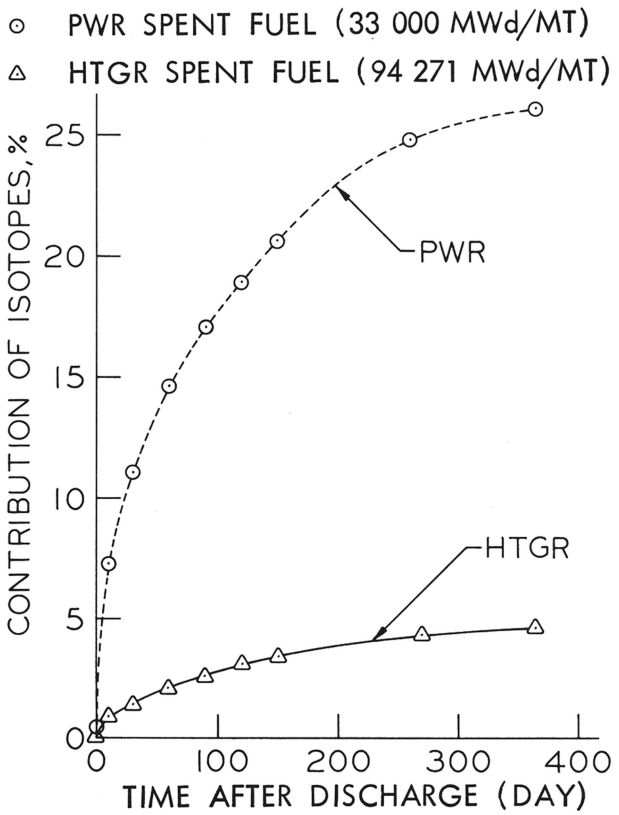

Fig. 5. Relative heat generation of $^{106}Ru/^{106}Rh$.

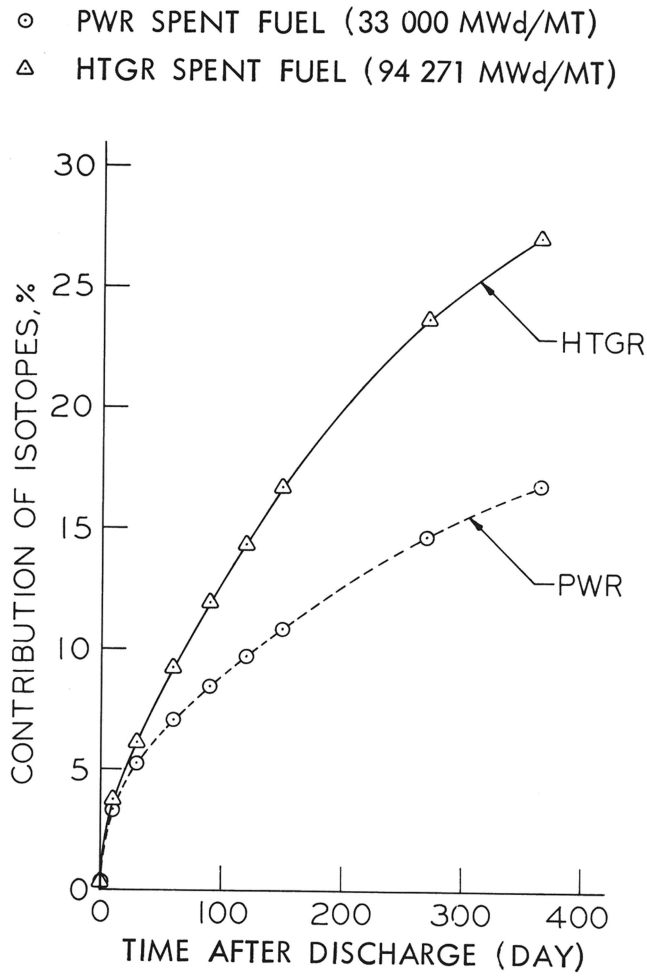

Fig. 6. Relative heat generation of ^{134}Cs.

waste generates the greatest amount of heat initially, but this amount is soon exceeded by that generated by the HTGR because of the increasing importance of the ^{238}Pu. This will persist until the ^{238}Pu is reduced and the heat generation of the longer life isotopes results in the wastes from the PWR generating more heat than that from the HTGR.

In summary, several comparisons have been made of the heat generated by spent fuel and processing waste from reference design PWR and HTGR by calculation using the ORIGEN and RADEC computer codes. The first year after discharge, the PWR heat generation rate exceeds that of the HTGR, and, in addition, the actinide nuclides, such as ^{233}Pa and ^{238}Pu, have a far greater significance in HTGR spent fuel than in PWR spent fuel. Finally, the HTGR waste generates heats which exceed those of the PWR waste from ~5 years to some 500 years of cooling. The numerous tables generated in the course of those calculations are too voluminous to permit publica-

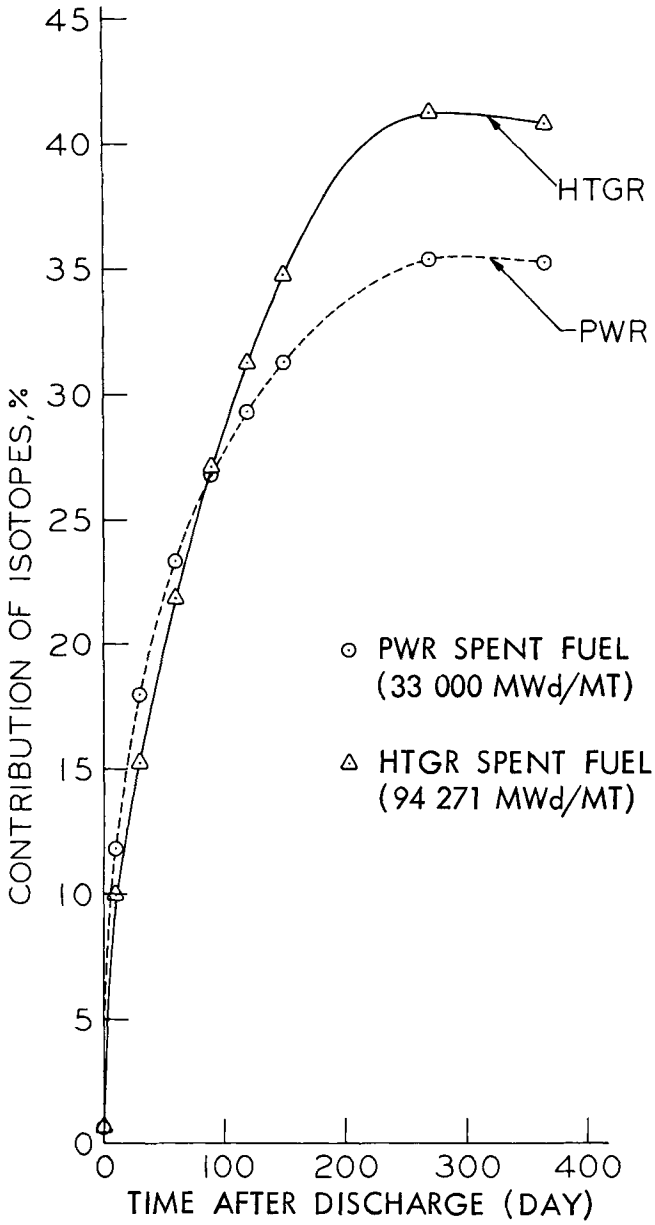

Fig. 7. Relative heat generation of $^{144}Ce/^{144}Pr$.

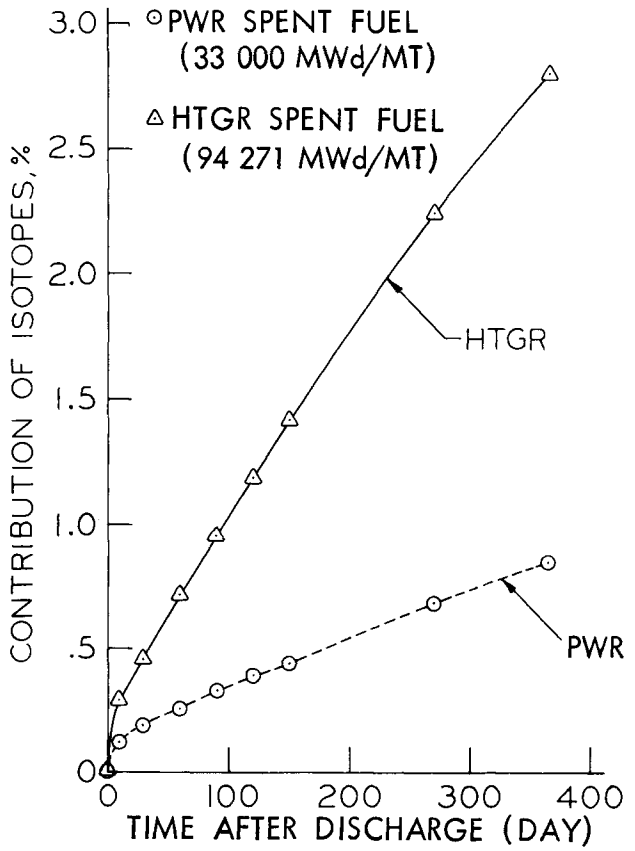

Fig. 8. Relative heat generation of ^{238}Pu.

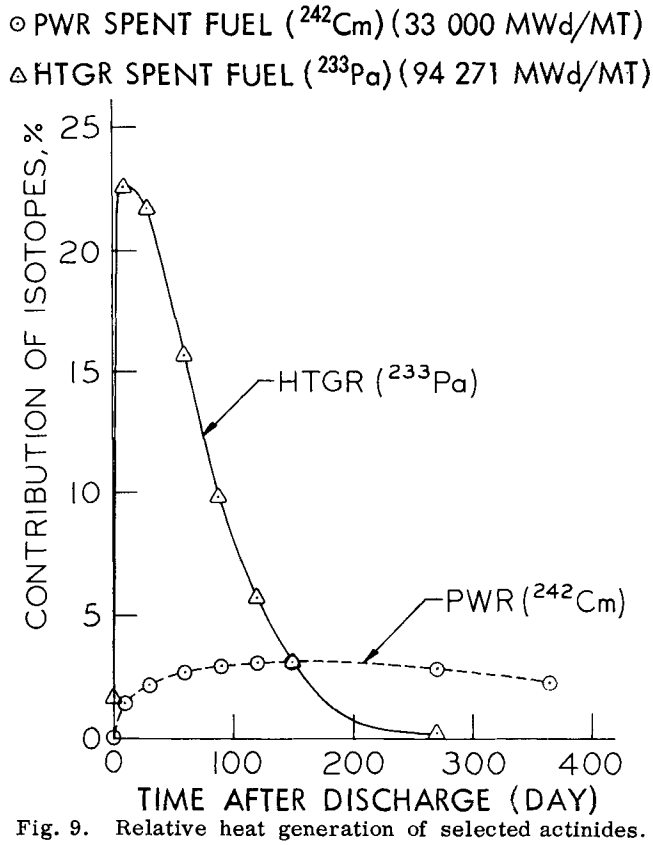

Fig. 9. Relative heat generation of selected actinides.

tion in an archival journal. The details are available in an Engineering Experiment Station Document being published by the College of Engineering of the University of Arizona.

The availability of the details of the heat generation rates should prove useful in the calculation of other quantities needed for the optimization of cooling times and heat requirements for the various fuel processing strategies. In addition, some information which may enable assessment of the economics of partitioning may also be provided.

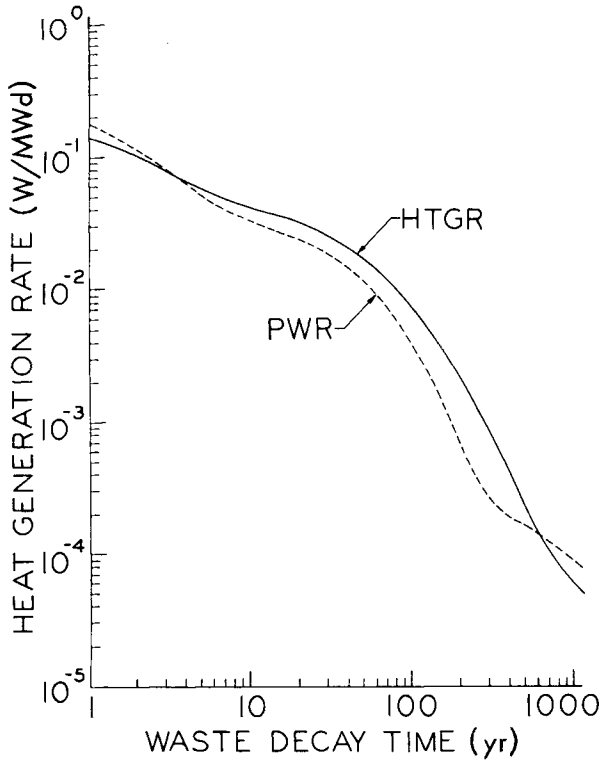

Fig. 10. Normalized heat generation of typical wastes from PWR and HTGR spent fuel processing.

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