

MOLTEN SALT REACTOR BENCHMARK PROBLEM TO CONSTRAIN PLUTONIUM*

N. HIRAKAWA, A.I. ABOANBER
Tohoku University, Sendai

K. MITACHI
Toyohashi University of Technology, Toyohashi

T. MISAWA
Kyoto University

Japan

Abstract. A molten salt reactor (MSR) based on ^{233}U -Th cycle has a positive feature from the standpoint of neutron economy, and it has been studied in Japan to utilize the MSR for the incineration of minor actinides (MA) produced in light water reactors (LWR). Therefore, Japan has proposed to study a MSR in the frame of the IAEA co-ordinated research project on the "Potential of thorium-based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicities". It is important to study the basic character of the reactor with simple model especially for the purpose of comparison with other type of reactors. This report presents the results of the benchmark calculation and the effect of fuel volume ratio for the burnup characteristics.

1. INTRODUCTION

It is considered that a molten salt reactor (MSR) based on ^{233}U -Th cycle is one of the best reactor system from the standpoint of neutron economy, and it has been studied in Japan to utilize the MSR for the incineration of minor actinides (MA) produced in light water reactors (LWR) [1,2,3]. Therefore, Japan has proposed to study a MSR in the IAEA research co-ordinated meeting on the "Potential of Thorium -based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicities" held in Vienna in October 1996. However, in the case of MSR, at least in principle, it is possible to make continuous fuel loading or extraction of poison material (such as Xe or ^{233}Pa), it is important to study the basic character of the reactor with simple model especially for the purpose of comparison with other type of reactors.

In this respect, a benchmark problem is prepared under the following considerations:

- (1) To make comparison with the first stage of benchmark problem of IAEA LWR lattice [4], a simple two region cell of graphite moderator and fuel, in which the fuel salt flows in the central circular hole opened at the center of graphite hexagonal column. This is basically the lattice design of MSR proposed by K. Furukawa [5].
- (2) Fuel salt is the mixture of LiF-BeP₂-ThF₄(-PuF₃ for the initial loading and plutonium concentration must not exceed 1 mol %.
- (3) Fuel salt does not flow.
- (4) To make the comparison with the High Temperature Gas Cooled Reactor which is proposed for the second stage of benchmark [6], the reactor power of 200 MW(th) is assumed, although it does not directly appear in the present calculation.

* 1997 meeting.

- (5) Plutonium vector is same as the IAEA LWR benchmark.
From the past experience, the following parameters are settled.
- (6) Fuel salt volume ratio is 0.1, and the face to face distance of the moderator graphite hexagon is 40.0cm. The graphite density is 1.84 g/cm³.
- (7) The power density of the fuel salt is 100 W/cm³ and the average temperature is 900°K.

According to the preliminary calculations, it was found that the k-inf is not sensitive to the plutonium concentration, and finally, the following fuel salt composition was determined for the initial loading: LiF-BeF₂-ThF₄-PuF₃= 72-16-11.8-0.2 mol %. The required quantities for the calculation is same as the IAEA LWR benchmark. That is,

- (1) k-inf at burnup of 20, 40, 60 MW·d/kg of Heavy Metal;
- (2) nuclide densities (n/cm³) from Th through Cm at above burnup points;
- (3) the total neutron flux;
- (4) average one group cross sections for capture, fission and (n,2n) reactions at a fuel burnup of 0 and 60 MW·d/kgHM;
- (5) average energy per fission.

2. RESULT OF BENCHMARK CALCULATION

Figure 1 shows the definition of the problem.

Case 1 Normal Case

Specification of a cell (infinite lattice): two zone composed of hexagonal graphite column with circular channel for flowing salt at the center (Fig .1). Side length of the hexagonal is 23.1 cm and the radius of the central zone is 6.64 cm (fuel volume ratio is 1).

(Fuel: LiF-BeF₂-ThF₄-PuF₃=72 : 16 : 11.8 : 0.2 mol% , Graphite : 1.840 g/cm³)
Temperature : 900°K Average Power: 100 W/cm³ for fuel salt) (constant)

Initial nuclide density (n/cm ³)		
Fuel salt		Graphite salt
Th-232	3.778E-03	C-12 9.226E-02
Pu-238	6.359E-07	
Pu-239	3.906E-05	
Pu-240	1.513E-05	
Pu-241	5.018E-06	
Pu-242	3.177E-06	
Li-7	2.260E-02	
Be-9	5.037E-03	
F-19	4.978E-02	

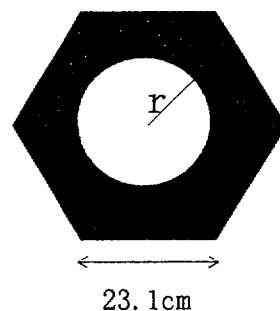


Figure 1. Specification of MSR Benchmark problem for plutonium burning for different cases.

For this problem, three institutions, Tohoku University (A), Nagoya University (B) and Toyohashi University of Technology (C), participated. The methods are: SWAT [7] in A, SRAC-95 [8] in B and SRAC [9]+ ORIGEN-2 [10] in C, respectively. SWAT is a burnup code based on SRAC and ORIGEN-2 connected by sub-codes for the preparations of cross sections for the burnup calculation and appropriate input for both main codes. SRAC-95

includes a burnup code called COREBN which is based on the analytical solution of the burnup equation. At present, 29 actinide nuclides from ^{230}Th to ^{246}Cm can be treated. As for the fission products (FP), 26 nuclides and 4 pseudo FPs are included. The method used in C is same as A, but the input preparation at each burnup step is carried out manually. The cross section library based on JENDL-3.2 is used in all the calculations, however, A uses data based on ENDF/B4 for Li, Be and F.

Table I, II, III show the change in k -inf., the total neutron flux and the average energy per fission with burnup, respectively. The change in k -inf. is also shown in Fig. 2. Table IV, V, VI show the values of (total plutonium/total initial plutonium), (fissile plutonium/total plutonium) and (minor actinide/total initial plutonium), with burnup, respectively, where the minor actinide means Np, Am and Cm. Table VII shows $(^{233}\text{Pa} + ^{233}\text{U})/(\text{total initial plutonium})$ with burnup which is the indication of the quantity of ^{233}U when the fuel is taken out at that burnup point. Table VIII is the summary of one group cross section at 0 and 60 MW·d/kg.

Since all the calculations are based on similar methods and cross sections, the tendency is same. As shown in Fig. 3, at first, plutonium decreases very rapidly and k -inf. also decreases very sharply. Actually most of ^{239}Pu is consumed by 20 MW·d/kg and at this point most of the fissile plutonium (~97%) is ^{241}Pu . Since the reactor power is kept constant, the flux increases and this promotes the conversion to ^{233}U from Th. Since η of ^{233}U is larger than for those of plutonium isotopes, k -inf. turns to increase and it reaches equilibrium at around 40 MW·d/kg of burnup. Total plutonium decreases steadily toward 60 MW·d/kg of burnup. The isotopic composition also changes and at 60 MW·d/kg of burnup, ^{242}Pu occupies about 96% in total plutonium.

Burnup (MW·d/kg)	A	B	C
0	1.14850	1.14165	1.13229
10	0.80344	0.74987	0.74777
20	0.77196	0.80440	0.77989
30	0.85391	0.84888	0.82595
40	0.86573	0.85542	0.83474
50	0.86369	0.85117	0.83377
60	0.85877	0.84502	0.83007

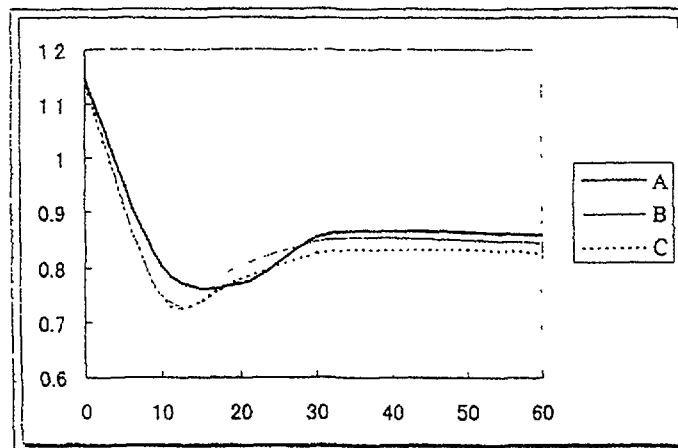


Figure 2. Change of k -inf vs. H. M. burnup (Case1)

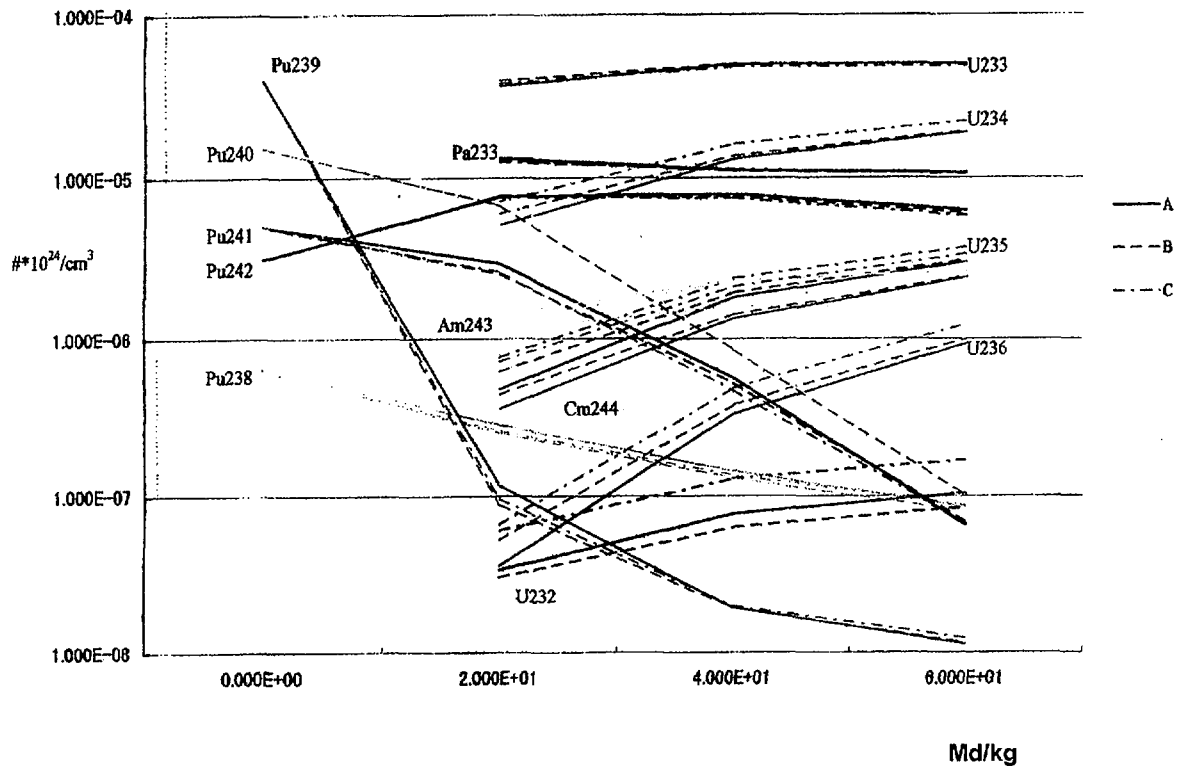


Figure 3. Change in nuclide densities with burnup.

Table I. Change in k-inf. vs. H.M. burnup.

Burnup MW·d/kg	0	10	20	30	40	50	60
A	1.14850	0.80344	0.77196	0.85391	0.86573	0.86369	0.85887
B	1.14165	0.74987	0.80440	0.84888	0.85542	0.85117	0.84502
C	1.13229	0.74777	0.779890	0.82595	0.83474	0.83377	0.83007

Table II. Total Neutron Flux vs. H.M. burnup (n/cm^2s).

Burnup(MW·d/kg)	0	20	40	60
A	3.510(E+14)	5.560(E+14)	5.030(E+14)	5.020(E+14)
B	3.229(E+14)	5.335(E+14)	4.903(E+14)	4.909(E+14)
C	3.27(E+14)	5.33(E+14)	4.88(E+14)	4.74(E+14)

Table III. Average Energy per Fission vs. H.M. burnup (MeV).

Burnup(MW·d/kg)	0	20	40	60
A	210.6	202.1	201.1	201.1
B	211.8	202.4	200.8	200.6
C	211.3	202.4	200.6	200

Table IV (Total Pu/Total Initial Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	1	0.3014	0.1513	0.1022
B	1	0.2776	0.1464	0.1022
C	1	0.280	0.144	0.0993

Table V. (Pu-fiss./Total Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	0.6994	0.1635	0.0611	0.0120
B	0.6994	0.1520	0.0566	0.0108
C	0.6994	0.1564	0.0544	0.01

Table VI. (Minor Actinides/Initial Total Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	0	0.0269	0.0603	0.0853
B	0	0.0318	0.0632	0.0868
C	0	0.0321	0.0644	0.0857

Table VII. ($^{233}\text{Pa} + ^{233}\text{U}$ /Total Initial Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	0	1.117	1.362	1.375
B	0	1.406	1.369	1.375
C	0	1.107	1.299	1.303

Table VIII-1. Cross sections at burnup = 0 MW·d/kg (barn).

Institution	A		B		C	
Nuclide	σ -f	σ -c	σ -f	σ -c	σ -f	σ -c
Th-232	0.0139	1.494	0.0137	1.544	0.014	1.636
Pa-233	0.0689	28.42	0.0676	28.53	0.068	31.44
U-233	84.37	10.14	88.09	10.49	94.40	11.26
U-234	0.3137	28.33	0.3085	28.92	0.306	31.23
U-235	70.63	14.44	74.61	15.04	79.70	16.23
U-236	0.2455	10.08	0.2427	10.02	0.252	11.39
U-238	0.0560	7.566	0.0551	7.477	0.056	8.263
Np-237	0.3420	48.97	0.3365	49.15	0.334	53.40
Np-239	0.363	17.91	0.3616	16.62	0.355	18.42
Pu-238	2.819	50.70	2.904	54.11	3.028	57.67
Pu-239	201.4	116.2	205.4	117.5	217.9	124.5
Pu-240	0.3781	118.2	0.3727	120.2	0.369	127.8
Pu-241	201.2	70.33	209.1	72.48	222.8	77.15
Pu-242	0.2687	31.43	0.2643	31.78	0.262	34.04
Am-241	1.384	188.2	1.347	181.5	1.430	196.2
Am-242m	1136	223.7	1206	237.7	1286	253.4
Am-243	0.3050	60.25	0.3017	60.86	328.8	69.0
Cm-242	1.415	5.124	1.416	4.863	1.469	65.38
Cm-243	121.4	19.39	125.3	20.25	134.5	21.67
Cm-244	0.6454	18.90	0.6432	18.56	0.667	21.78

Table VIII-2. Cross sections at burnup = 60 MW·d/kg (barn).

Institution		A		B		C	
Nuclide	σ -f	σ -c	σ -f	σ -c	σ -f	σ -c	
Th-232	0.0110	1.869	0.0109	1.916	0.011	1.994	
Pa-233	0.0547	21.75	0.0545	21.67	0.054	23.82	
U-233	115.3	12.65	118.1	12.93	124.2	13.68	
U-234	0.2460	25.21	0.2451	25.92	0.241	27.53	
U-235	106.4	20.34	109.5	20.76	114.4	21.83	
U-236	0.1836	7.076	0.1821	6.989	0.193	7.936	
U-238	0.0443	5.774	0.0440	5.691	0.044	6.467	
Np-237	0.2739	58.94	0.2729	58.86	0.269	60.81	
Np-23	0.2575	17.24	0.2925	15.99	0.284	17.40	
Pu-238	3.387	7.433	3.481	77.76	3.581	80.90	
Pu-239	375.9	252.2	360.8	213.4	379.2	224.7	
Pu-240	0.3246	216.9	0.3229	212.5	0.319	228.5	
Pu-241	335.7	121.8	335.9	120.0	352.3	126.0	
Pu-242	0.2173	21.18	0.2131	21.59	0.209	24.02	
Am-241	2.198	329.6	2.018	298.8	2.119	316.4	
Am-242m	1800	355.7	1855	366.9	1940	383.7	
Am-243	0.2464	47.40	0.2454	47.09	0.249	52.05	
Cm-242	1.62	5.582	1.589	5.238	1.634	5.618	
Cm-243	155.6	25.68	157.6	26.35	166.7	27.71	
Cm-244	0.5933	15.27	0.5968	14.90	0.609	16.58	

On the other hand, minor actinides increase almost linearly with burnup. Looking at the minor actinides in detail, those of more than 95% are ^{243}Am and ^{244}Cm , which have relatively small absorption cross sections, though the ratio somewhat decreases with burnup. Although there are some discrepancies in the range around 20 MW·d/kg of burnup, the results at 60 MW·d/kg of burnup almost agree.

3. EFFECT OF FUEL VOLUME RATIO

It is interesting to see the effect of the fuel volume ratio since the neutron spectrum and the effective cross sections are mainly determined by the moderator volume ratio. Therefore, in addition to the above benchmark calculation (case 1), cases with $V_p/V = 0.05$ (case 2) and $V_p/V = 0.20$ (case 3) were studied with SWAT system. The volume ratio was changed by varying the radius of the molten salt region at the center. It is 4.697 cm for case 2 and 9.395cm for case 3, respectively. The initial Pu-mol% in the fuel salt was determined so that the initial k-inf. to be the same as in case 1, and it was 0.16% for case 2. However, for case 3, we adopted 0.6 mol% of plutonium since this value is recommended as the upper limit of plutonium concentration. The initial nuclide densities of fuel salt are given in Table IX.

Table IX. Initial nuclide densities for fuel salt.

Case	Pu (mol%)	Th-232	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
case 1	0.2	3.778(-3)	6.359(-7)	3.906(-5)	1.513(-5)	5.013(-6)	3.177(-6)
case 2	0.16	3.791(-3)	5.087(-7)	3.125(-5)	1.210(-5)	4.014(-6)	2.542(-6)
case 3	0.6	3.650(-3)	1.908(-6)	1.172(-4)	4.539(-5)	1.505(-5)	9.53

Those of Li-7, Be-9, F-19 are same as nominal case.

The power density is kept to be $100\text{W}/\text{cm}^3$ for the fuel salt. Thus, the volume of the reactor core should be doubled for case 2 and should be halved for case 3 from case 1, although this effect does not taken into account for the present study.

Figure 4 shows the change in k -inf. with burnup. It is interesting that k -inf. shows similar character for case 1 and case 2, but for case 3 the curve is quite different.

Burnup (MW·d/kg)	A	B	C
0	1.14850	1.14165	1.13229
10	0.80344	0.74987	0.74777
20	0.77196	0.80440	0.77989
30	0.85391	0.84888	0.82595
40	0.86573	0.85542	0.83474
50	0.86369	0.85117	0.83377
60	0.85877	0.84502	0.83007

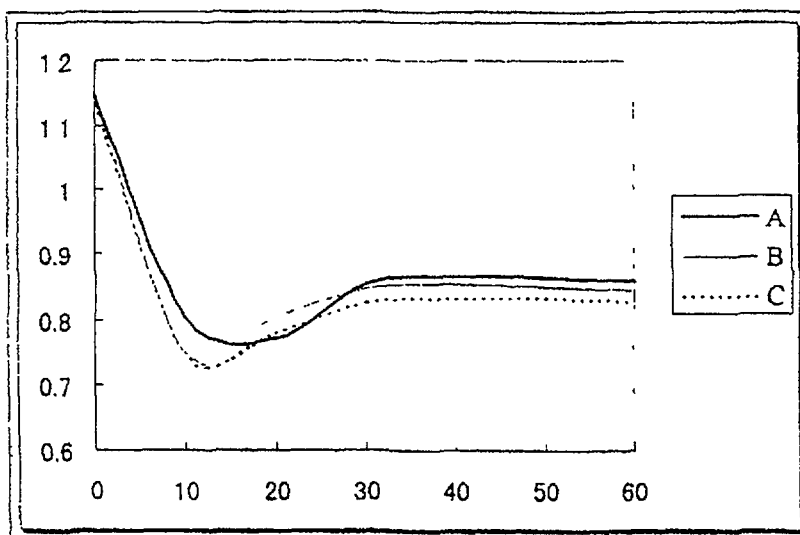


Figure 4. : Change of k -inf. vs. H. M. burnup.

This fact can be explained from the difference in the effective cross sections. For instance, the one-group effective fission cross sections at 0 MW·d/kg of burnup for case 1, 2, 3 are 225b, 303b and 107b, respectively. Due to the large fission cross sections for case 1 and case 2, ^{239}Pu is consumed very rapidly and the reduction of ^{239}Pu concentration increases the effective fission cross section even more since the neutron spectrum becomes softer with the decrease of materials with large cross sections. The effective fission cross section of ^{239}Pu becomes 379b at 20 MW·d/kg of burnup for case 1, which is 1.7 times as large as at 0 MW·d/kg of burnup and it reaches 376b at 60 MW·d/kg of burnup. Thus most of fissile plutonium isotopes are destroyed by 20 MW·d/kg of burnup. In contrast, for case 3, the effective fission cross section of plutonium at 0 MW·d/kg is relatively small and the decrease of plutonium concentration is slow, therefore, the behavior is somewhat similar to the IAEA benchmark case for LWR lattice, which gives the values of Pu-total/Pu initial = 0.16, Pu-fissile/Pu total = 0.18 and Minor actinide/Initial Pu = 0.0687 at 60 MW·d/kg of burnup. Table X and XI presents the results of the total neutron flux, the average energy per fission.

Table X. Total neutron flux vs. HM-burnup (n/cm²s) with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	3.510(E+14)	5.560(E+14)	5.030(E+14)	5.020(E+14)
case 2	2.630(E+14)	4.340(E+14)	4.030(E+14)	4.050(E+14)
case 3	4.360(E+14)	5.110(E+14)	5.870(E+14)	6.180(E+14)

Table XI. Average energy per fission vs. burnup (MeV) with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	210.6	202.1	201.1	201.1
case 2	210.6	201.6	201.2	201.6
case 3	211.3	207.9	203.2	202.8

For case 1 and 2, the total neutron flux initially increases with burn up because of the decrease in fissile plutonium concentration. the flux shows a peak around 20 MW·d/kg of burnup due to the increase of ²³³U nuclide density. For case 3, the flux increases monotonically to compensate the decrease in the fissile plutonium nuclide density. The average energy per fission clearly shows the contribution of main fissioning nuclide at that time.

From Table XII to XV, changes of (total plutonium/total initial plutonium), (fissile plutonium/total plutonium), (minor actinides/total initial plutonium) and (²³³Pa + ²³³U)/(total initial plutonium) for different fuel salt volume ratio with burnup are presented, respectively.

Table XII (Total Pu/Total Initial Pu) vs. HM-burnup with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	1	0.3014	0.1513	0.1022
case 2	1	0.2379	0.1302	0.0957
case 3	1	0.6530	0.3714	0.2070

Table XIII. (Pu-fiss./Total Pu) vs. HM-burnup with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	0.6994	0.1635	0.0611	0.0120
case 2	0.6994	0.1447	0.0543	0.0139
case 3	0.6994	0.4994	0.2738	0.1738

Table XIV. (Minor actinides/initial total Pu) vs. HM-burnup with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	0	0.0269	0.0603	0.0853
case 2	0	0.0125	0.0330	0.0507
case 3	0	0.0170	0.0370	0.0587

Table XV. ([Pa-233 + U-233]/Total Initial Pu) vs. H.M.Burnup with volume ratio change.

Burnup(MW·d/kg)	0	20	40	60
case 1	0	1.117	1.362	1.375
case 2	0	1.406	1.633	1.625
case 3	0	0.2611	0.4560	0.5372

It is seen that (total plutonium/total initial plutonium) and (fissile plutonium/total plutonium) are smallest for case 1, though the (minor actinide/total initial plutonium) is highest for the case. From these observations, the choice of $V_p/V = 0.10$ for the benchmark calculation seems to be appropriate for the present purpose.

4. SUMMARY

For the first step to investigate a molten salt reactor for the purpose "to Constrain Plutonium and to Reduce Long-term Toxicities", a benchmark problem was constructed. Three institutions participated to solve the problem. Since their methods are basically the same, the final results are similar although some discrepancies exist in the course of burnup. The effect of fuel volume ratio which affects the neutron spectrum was also investigated. It turns out that the volume ratio of $V_p/V = 0.1$ selected for the benchmark is suitable to destroy plutonium effectively. Since the decrease of k -inf. is very rapid due to the destruction of fissile plutonium, the means should be taken to keep the change in k -inf. below certain range by the addition of plutonium salt properly. The strategy for this addition will be the task for next stage.

ACKNOWLEDGEMENT

K. Mitachi wishes to thank to Atsushi Namekata of Toyohashi University of Technology for his assistance in the calculation.

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