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CONSISTENCY EVALUATION OF ACCOUNTABILITY DATA FOR A CHEMICAL PROCESSING PLANT

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ABSTRACT

The purpose of this report is to evaluate accountability data collected during the normal dissolution of irradiated fuel at a chemical reprocessing plant. Results important to safeguarding nuclear materials are presented.

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CONSISTENCY EVALUATION OF ACCOUNTABILITY
DATA FOR A CHEMICAL PROCESSING PLANT

D.E. Christensen
R.A. Schneider

Accountability data collected at a chemical reprocessing plant are of themselves a valuable safeguards tool. The basic principle supporting this conclusion is based on the following observation:

- a. Accountability data are actually nuclear fuel burnup data and are taken by the same analytical procedures used in any burnup experiment.
- b. Burnup data are inherently consistent.

By being consistent, it is meant that a burnup parameter traces a continuous, distinct path characteristic of a given reactor and its fuel loading as a function of exposure. It should be understood also that a burnup parameter can

be a part of the burnup data in the form it was obtained or a quantity derived from the data. The inherent consistency of burnup data is a built-in property provided by nature. A look at the transmutation equations,

$$\frac{dN^i}{dt} = -\lambda^i N^i - \hat{\sigma}_a^i N^i \phi + \hat{\sigma}_c^{i-1} N^{i-1} \phi \quad (1)$$

suggests that there is an internal order to burnup data. The equations relate concentration change of a fuel isotope with time to loss by decay, loss by neutron absorption, and gain by neutron capture in a preceding isotope. Loss by absorption and gain by capture in a preceding isotope are the main processes that determine the isotopic concentration change of the uranium isotopes of mass 235, 236 and 238 and of the plutonium isotopes of mass 239, 240 and 242. Loss by decay becomes important when considering ^{241}Pu and ^{238}Pu isotopic concentrations. The cross section quantities, $\hat{\sigma}$, are flux and volume averaged values. Absorption, $\hat{\sigma}_a$, capture, $\hat{\sigma}_c$, and fission, $\hat{\sigma}_f$, of an isotope are related by

$$\hat{\sigma}_a = \hat{\sigma}_c + \hat{\sigma}_f \quad (2)$$

and values of the cross sections are dependent on the neutron energy spectrum of a reactor. They have a unique set of average values for a given reactor situation since each reactor is different due to its type, fuel enrichment, fuel spacing, control material, coolant, etc. For a given reactor and fuel type, the interrelations between plutonium production and

- the fission and transmutation of uranium isotopes,
- the fission and transmutation of plutonium isotopes, and
- the formation of transuranium and transplutonium nuclides are consistent

and informative. Our intent, then, is to examine the plant accountability data made available and investigate their use for

safeguards application by checking the consistency of the data. Information will be learned that will indicate the reliability of the data and how data consistency is to be applied as a safeguards tool.

THE DATA - MEASUREMENT PROCEDURES⁽¹⁾

The data was made available from measurements made to quantitatively determine the input of irradiated fuel to the reprocessing plant. For each batch of dissolved fuel, measurements of the liquids level, density, and temperature were made to determine the volume of solution. After adequate mixing, eight to ten sample vials were filled from each dissolver batch and the contents of at least two sample vials were analyzed by the plant laboratory for uranium and plutonium concentration and for isotopic compositions. A detailed description of all the measurement procedures can be found in Reference 2. The analytical data were transmitted to the accountability representative for determination of input uranium and plutonium concentrations for each batch. The data received by the evaluators were the concentrations and isotopic weight percents for a total of 85 input dissolution batches.

The dissolution batches represent fuel from two reactors, one a pressurized water reactor (PWR) and the other, a boiling water reactor (BWR). Three groups of fuel from each reactor were processed. The dissolution batches for each group are summarized in Table I. It is not known how many elements were dissolved in each batch for BWR cores but the number of elements dissolved in a batch varied from two to eight elements for PWR cores.⁽³⁾ The data received are tabulated in Table II in data sets representing each group as outlined in Table I. The zero sample of each set shown in Table II is the initial fuel composition and since these values were not known accurately for all cores, the ^{234}U , ^{236}U , and ^{238}U are estimates in some cases (all the uranium initial values are estimates for BWR group 3). The accountability data are tabulated first in Table II and from

TABLE I

SUMMARY OF FUEL DISSOLVED

<u>Fuel From</u>	<u>Fuel Group Designation In Table 1</u>	<u>Number of Dissolution Batches</u>	<u>Initial ^{235}U Enrichment Weight Percent</u>	<u>Plotting Symbol</u>
Yankee Core 1	PWR1	16	3.4	•
Yankee Core 2,3	PWR2	14	3.4	○
Yankee Core 4	PWR3	11	4.1	×
				Batches containing fuel of different enrichments. ▲
Dresden Core 1	BWR1	17	1.5	◻
Dresden Core 2	BWR2	18	1.5	△
Dresden Core 3	BWR3	9	2.3 (?)	◊

TABLE II
ACCOUNTABILITY DATA FROM THE DISSOLUTION OF REACTOR FUELS AND BURNUP PARAMETERS DERIVED FROM THE DATA.

PWR Group 1

BATCH No.	URANIUM WEIGHT PERCENTS				GRAMS U	PLUTONIUM WEIGHT PERCENTS					GRAMS Pu
	U234	U235	U236	U238		Pu238	Pu239	Pu240	Pu241	Pu242	
0	.020	3.401	.020	96.560	100.0	.000	100.000	.000	.000	.000	.0
1	.018	2.511	.201	97.270	2166000.0	.251	82.787	11.859	4.572	.531	11309.0
2	.017	2.542	.188	97.253	2158000.0	.221	83.553	11.465	4.283	.478	10380.0
3	.018	2.404	.227	97.352	1072000.0	.267	81.490	12.538	5.073	.632	6280.0
4	.017	2.431	.218	97.334	1070000.0	.249	82.070	12.249	4.849	.583	5947.0
5	.017	2.518	.193	97.273	1087000.0	.241	82.791	11.824	4.582	.562	5347.0
6	.018	2.502	.199	97.281	1101000.0	.237	82.555	11.970	4.675	.563	5650.0
7	.011	2.620	.181	97.187	689000.0	.206	84.337	11.049	3.981	.427	3071.0
8	.010	2.597	.184	97.209	1157000.0	.197	84.599	10.942	3.864	.397	5249.0
9	.018	2.754	.150	97.078	1052000.0	.139	87.097	9.585	2.929	.250	3762.0
10	.019	2.908	.121	96.952	1102000.0	.107	89.395	8.208	2.131	.159	3149.0
11	.019	2.950	.113	96.918	1084000.0	.092	90.190	7.734	1.858	.126	2742.0
12	.019	2.737	.157	97.088	1072000.0	.184	85.404	10.404	3.597	.411	4067.0
13	.018	2.538	.192	97.252	1075000.0	.208	83.385	11.552	4.355	.501	5197.0
14	.019	2.768	.149	97.064	1666000.0	.141	86.974	9.607	3.007	.272	5829.0
15	.018	2.541	.197	97.244	1558000.0	.201	83.946	11.361	4.141	.351	7650.0
16	.017	2.373	.231	97.378	944000.0	.252	81.189	12.687	5.196	.676	5654.0
TOTAL GRAMS AND AVERAGE WEIGHT PERCENTS											
GRAMS	3465.4	521769.1	36457.2	19491323.0	20053000.0	193.1	76608.8	10270.3	3789.5	421.4	91283.0
Wt%	.017	2.602	.182	97.199		.212	83.924	11.251	4.151	.462	

OTHER PARAMETERS DETERMINED FROM ABOVE DATA

	PLUTONIUM ATOM RATIOS				URANIUM ATOM RATIOS				Conversion Ratio ^a	
	238/239 ^a	240/239 ^a	241/239 ^a	242/239 ^a	U234/U238	U235/U238 ^a	U236/U238	Pu/U Ratio ^a		U235 Depl. ^a
0	.000000	.000000	.000000	.000000	.000211	.035672	.000209	.000000	.000000	.000000
1	.003045	.142649	.054767	.006334	.000188	.026145	.002084	.005221	.261688	.575170
2	.002656	.136646	.050835	.005650	.000178	.026472	.001950	.004810	.252573	.549490
3	.003290	.153217	.061735	.007659	.000188	.025010	.002352	.005858	.293149	.576072
4	.003047	.148628	.058592	.007015	.000176	.025295	.002259	.005558	.285210	.562032
5	.002923	.142221	.054884	.006704	.000178	.026217	.002001	.004919	.259630	.546725
6	.002883	.144389	.056158	.006735	.000188	.026048	.002063	.005132	.264334	.559952
7	.002453	.130463	.046611	.005000	.000115	.027303	.001878	.004457	.229638	.559836
8	.002338	.128800	.045294	.004634	.000105	.027057	.001909	.004537	.236401	.553645
9	.001603	.109590	.033350	.002835	.000189	.028732	.001558	.003576	.190238	.542514
10	.001202	.091434	.023640	.001757	.000199	.030378	.001259	.002858	.144957	.568416
11	.001024	.085394	.020430	.001380	.000199	.030827	.001176	.002530	.132608	.550371
12	.002164	.121313	.041767	.004753	.000199	.028552	.001631	.003794	.195237	.560471
13	.002505	.137960	.051793	.005934	.000188	.026431	.001991	.004834	.253749	.549714
14	.001628	.109997	.034286	.003089	.000199	.028882	.001548	.003499	.186122	.542534
15	.002404	.134772	.048919	.004129	.000188	.026464	.002043	.004910	.252867	.560071
16	.003117	.155613	.063467	.008223	.000178	.024681	.002392	.005989	.302264	.571307

TABLE II CONTINUED

PWR Group 2

BATCH No.	URANIUM WEIGHT PERCENTS				GRAMS U		PLUTONIUM WEIGHT PERCENTS					GRAMS Pu	
	U234	U235	U236	U238			Pu238	Pu239	Pu240	Pu241	Pu242		
0	.020	3.401	.020	96.560	100.0		.000	100.000	.000	.000	.000	.0	
17	.017	2.362	.225	97.396	1562000.0		.293	80.104	12.839	5.986	.779	9440.0	
19	.012	2.502	.202	97.285	1613000.0		.213	82.585	11.788	4.876	.538	8455.0	
21	.017	2.461	.205	97.318	1669000.0		.226	82.331	11.927	4.980	.536	9103.0	
22	.017	2.329	.231	97.424	1513000.0		.306	79.869	12.968	6.079	.779	9344.0	
23	.017	2.420	.214	97.348	1603000.0		.266	81.272	12.299	5.524	.640	9216.0	
24	.017	2.417	.213	97.352	1541000.0		.240	81.601	12.273	5.291	.595	8885.0	
25	.017	2.402	.215	97.365	1659000.0		.242	81.394	12.369	5.382	.613	9607.0	
26	.017	2.398	.220	97.374	1499000.0		.264	80.893	12.522	5.656	.666	8752.0	
27	.017	2.398	.218	97.366	1646000.0		.262	91.017	12.495	5.566	.660	9662.0	
28	.017	2.401	.214	97.348	1572000.0		.250	81.578	12.288	5.275	.608	9066.0	
29	.017	2.278	.241	97.464	1099000.0		.374	78.464	13.673	6.448	1.042	7203.0	
30	.016	2.111	.271	97.602	1068000.0		.372	76.576	14.438	7.451	1.163	8057.0	
31	.018	2.108	.270	97.605	1059000.0		.412	76.641	14.375	7.421	1.152	7831.0	
32	.018	2.618	.173	97.192	1093000.0		.158	85.080	10.601	3.830	.331	4978.0	
TOTAL GRAMS AND AVERAGE WEIGHT PERCENTS													
Grams	3363.5	480981.4	44512.1	19666817.0	20196000.0		332.0	96423.7	15137.1	6842.5	864.0	119599.0	
wtl.	.017	2.382	.220	97.380			.278	80.623	12.657	5.721	.722		

OTHER PARAMETERS DETERMINED FROM ABOVE DATA

	PLUTONIUM ATOM RATIOS				URANIUM ATOM RATIOS				Pu/U Ratio ^a	U235 Depletion ^a	Conversion Ratio ^a
	238/239 ^a	240/239 ^a	241/239 ^a	242/239 ^a	U234/U238	U235/U238 ^a	U236/U238				
0	.000000	.000000	.000000	.000000	.000211	.035672	.000209	.000000	.000000	.000000	.000000
17	.003673	.159610	.074106	.009604	.000178	.024562	.002330	.006044	.305498	.570385	
19	.002590	.142142	.058551	.006434	.000125	.026047	.002094	.005242	.264334	.571731	
21	.002757	.144262	.059985	.006429	.000178	.025612	.002124	.005454	.276389	.569002	
22	.003847	.161688	.075476	.009632	.000177	.024212	.002391	.006176	.315201	.565031	
23	.003287	.150700	.067404	.007777	.000178	.025177	.002217	.005749	.288445	.574604	
24	.002954	.149775	.064301	.007201	.000178	.025145	.002207	.005766	.289327	.574500	
25	.002986	.151330	.065573	.007438	.000178	.024986	.002227	.005791	.293737	.568457	
26	.003277	.154151	.069338	.008131	.000178	.024838	.002279	.005839	.297854	.565282	
27	.003248	.153583	.068130	.008045	.000178	.024944	.002258	.005870	.294913	.573822	
28	.003077	.150000	.064124	.007360	.000178	.024980	.002217	.005767	.294031	.565623	
29	.004787	.173631	.081494	.013115	.000177	.023672	.002494	.006554	.330197	.572269	
30	.004878	.187758	.096493	.014999	.000167	.021905	.002800	.007544	.379300	.573402	
31	.005398	.186780	.096023	.014844	.000188	.021874	.002790	.007395	.380182	.560992	
32	.001865	.124080	.044642	.003842	.000188	.027281	.001795	.004554	.230226	.570382	

TABLE II-CONTINUED

PWR Group 3

BATCH NO.	URANIUM WEIGHT PERCENTS				GRAMS U	PLUTONIUM WEIGHT PERCENTS					GRAMS PU
	U234	U235	U236	U238		Pu238	Pu239	Pu240	Pu241	Pu242	
0	.020	4.101	.040	95.840	100.0	.000	100.000	.000	.000	.000	.0
33	.019	2.675	.316	96.990	890000.0	.432	77.977	13.337	7.294	.960	6684.0
34	.019	2.721	.313	96.947	854000.0	.441	78.390	13.150	7.096	.924	6207.0
35	.018	2.462	.316	97.205	882000.0	.458	76.505	14.030	7.851	1.156	7007.0
36	.019	2.637	.333	97.011	880000.0	.475	77.408	13.549	7.538	1.029	6908.0
37	.019	2.726	.312	96.943	891000.0	.424	78.481	13.114	7.079	.901	6561.0
38	.018	2.490	.314	97.178	870000.0	.439	76.936	13.877	7.656	1.092	6802.0
39	.019	2.606	.338	97.037	877000.0	.484	76.787	13.820	7.811	1.097	7132.0
40	.019	2.706	.319	96.957	892000.0	.424	78.335	13.178	7.151	.912	6704.0
41	.019	2.643	.323	97.015	877000.0	.445	77.478	13.560	7.498	1.019	6802.0
42	.018	2.438	.326	97.218	896000.0	.460	76.297	14.132	7.932	1.171	7225.0
43	.017	2.261	.355	97.366	640000.0	.765	72.713	15.525	9.101	1.896	5764.0

TOTAL GRAMS AND AVERAGE WEIGHT PERCENTS

GRAMS	1756.0	244403.1	30550.1	9172301.7	9449000.0	549.4	56369.7	10138.4	5628.2	809.6	73796.0
wt. %	.019	2.587	.323	97.072		.473	77.063	13.738	7.627	1.097	

OTHER PARAMETERS DETERMINED FROM ABOVE DATA

PLUTONIUM ATOM RATIOS				URANIUM ATOM RATIO			Pu/U RATIO ^a	U235 DEPL. ^a	CONVERSION RATIO ^a
238/239 ^a	240/239 ^a	241/239 ^a	242/239 ^a	U234/U238	U235/U238 ^a	U236/U238			
0	.000000	.000000	.000000	.000212	.043337	.000421	.000000	.000000	.000000
33	.005563	.170324	.092763	.000199	.027933	.003286	.007510	.347720	.515522
34	.005649	.167051	.089769	.000199	.028426	.003256	.007268	.336503	.515542
35	.006012	.182621	.101767	.000188	.025652	.003278	.007944	.399659	.475266
36	.006162	.174333	.096570	.000199	.027530	.003462	.007850	.356986	.524665
37	.005425	.166400	.089450	.000199	.028479	.003246	.007364	.335284	.524028
38	.005730	.179618	.098684	.000188	.025951	.003259	.007818	.392831	.475842
39	.006330	.179227	.100877	.000199	.027199	.003513	.008132	.364545	.532095
40	.005435	.167524	.090528	.000199	.028266	.003318	.007516	.340161	.527113
41	.005768	.174237	.095971	.000199	.027592	.003358	.007756	.355523	.520603
42	.006054	.184451	.103098	.000188	.025398	.003382	.008064	.405511	.475430
43	.010565	.212620	.124123	.000178	.023519	.003677	.009006	.448671	.479838

TABLE II - Continued

BWR Group 1

BATCH NO.	URANIUM WEIGHT PERCENTS				GRAMS U	PLUTONIUM WEIGHT PERCENTS					GRAMS PU
	U234	U235	U236	U238		Pu238	Pu239	Pu240	Pu241	Pu242	
0	.013	1.500	.017	98.470	100.0	.000	100.000	.000	.000	.000	.0
1	.008	.931	.101	98.960	1402159.0	.152	77.915	16.886	4.332	.716	4674.0
2	.008	.940	.100	98.953	1992923.0	.144	78.351	16.601	4.230	.674	6606.0
3	.008	.937	.101	98.954	987882.0	.150	78.044	16.814	4.295	.693	3327.0
4	.008	.926	.103	98.963	971217.0	.151	77.740	17.009	4.369	.731	3348.0
5	.008	.934	.101	98.957	984830.0	.154	77.847	16.847	4.356	.718	3354.0
6	.008	.937	.101	98.953	2043163.0	.153	78.075	16.794	4.283	.696	7010.0
7	.009	.947	.099	98.944	2025279.0	.147	78.437	16.575	4.177	.664	6614.0
8	.008	.928	.107	98.956	977914.0	.159	77.571	17.143	4.387	.740	3348.0
9	.008	.941	.100	98.950	988138.0	.154	78.136	16.757	4.259	.695	3306.0
10	.008	.950	.097	98.945	1012095.0	.157	78.721	16.388	4.100	.634	3316.0
11	.008	.942	.098	98.952	2017602.0	.143	78.650	16.441	4.120	.646	6560.0
12	.008	.952	.096	98.944	1005255.0	.141	79.967	15.488	3.849	.556	3215.0
13	.008	.942	.101	98.949	989252.0	.159	78.078	16.788	4.276	.698	3293.0
14	.008	.947	.097	98.948	435550.0	.156	78.527	16.491	4.167	.660	1461.0
15	.008	.937	.101	98.954	978981.0	.149	78.144	16.771	4.245	.691	3302.0
16	.008	.928	.102	98.962	1003429.0	.154	77.758	16.979	4.371	.739	3384.0
17	.008	.994	.091	98.907	469463.0	.158	78.883	16.240	4.042	.677	1366.0

TOTAL GRAMS AND AVERAGE WEIGHT PERCENTS

GRAMS	1622.8	190718.5	20290.2	20072439.0	20285132.0	101.6	52817.3	11245.1	2856.3	461.0	67484.0
wt%	.008	.940	.100	98.951		.151	78.266	16.663	4.233	.683	

OTHER PARAMETERS DETERMINED FROM ABOVE DATA

PLUTONIUM ATOM RATIOS				URANIUM ATOM RATIOS			Pu/U RATIO ^a	U235 DEPL. ^a	CONVERSION RATIO ^a
238/239 ^a	240/239 ^a	241/239 ^a	242/239 ^a	U234/U238	U235/U238 ^a	U236/U238			
0	.000000	.000000	.000000	.000134	.015428	.000174	.000000	.000000	.000000
1	.001959	.215819	.055137	.000082	.009528	.001029	.003333	.379333	.580737
2	.001846	.210996	.053539	.000082	.009621	.001019	.003315	.373333	.586707
3	.001930	.214543	.054575	.000082	.009590	.001029	.003368	.375333	.592870
4	.001951	.217880	.055733	.000082	.009477	.001050	.003447	.382667	.595199
5	.001987	.215509	.055491	.000082	.009559	.001029	.003406	.377333	.596325
5	.001968	.214203	.054401	.000082	.009590	.001029	.003431	.375333	.603886
7	.001882	.210434	.052810	.000082	.009694	.001009	.003266	.368667	.585361
8	.002058	.220075	.056084	.000082	.009496	.001090	.003424	.381333	.593208
9	.001979	.213564	.054054	.000082	.009632	.001019	.003346	.372667	.593187
10	.002003	.207309	.051650	.000082	.009724	.000989	.003276	.366667	.590428
11	.001826	.208168	.051948	.000082	.009642	.000999	.003251	.372000	.577637
12	.001771	.192872	.047732	.000082	.009745	.000978	.003198	.365333	.578547
13	.002045	.214118	.054310	.000082	.009642	.001029	.003329	.372000	.591264
14	.001995	.209128	.052623	.000082	.009693	.000985	.003354	.368667	.601109
15	.001915	.213721	.053871	.000082	.009590	.001029	.003373	.375333	.593757
16	.001989	.217446	.055745	.000082	.009497	.001039	.003372	.381333	.584418
17	.002011	.205015	.050814	.000082	.010178	.000928	.002910	.337333	.570123

TABLE II CONTINUED

BWR Group 2

BATCH NO.	URANIUM WEIGHT PERCENTS				GRAMS U	PLUTONIUM WEIGHT PERCENTS					GRAMS PU
	U234	U235	U236	U238		Pu238	Pu239	Pu240	Pu241	Pu242	
0	.013	1.500	.017	98.470	100.0	.000	100.000	.000	.000	.000	.0
18	.007	.692	.141	99.160	812101.0	.408	67.821	22.279	7.459	2.033	3818.0
19	.007	.700	.140	99.153	987455.0	.372	73.352	18.501	6.155	1.621	4669.0
20	.007	.731	.134	99.127	1000767.0	.363	69.399	21.616	6.872	1.750	4518.0
21	.008	.781	.143	99.068	925880.0	.356	69.466	21.415	7.015	1.749	4239.0
22	.008	.735	.138	99.118	983057.0	.365	69.369	21.625	6.905	1.736	4411.0
23	.007	.721	.137	99.135	822331.0	.370	68.819	22.023	6.973	1.815	3763.0
24	.007	.733	.131	99.129	990108.0	.350	69.571	21.490	6.887	1.701	4357.0
25	.007	.734	.134	99.124	999539.0	.363	69.221	21.751	6.909	1.751	4467.0
26	.007	.745	.132	99.116	1942957.0	.341	69.876	21.346	6.758	1.679	8647.0
27	.007	.748	.129	99.117	2010243.0	.323	70.286	21.223	6.547	1.620	9109.0
28	.006	.752	.130	99.111	1018109.0	.320	70.526	21.155	6.409	1.589	4284.0
29	.007	.744	.132	99.116	992156.0	.321	70.675	20.983	6.422	1.599	4422.0
30	.007	.750	.129	99.114	1996092.0	.323	70.828	20.961	6.344	1.544	8629.0
31	.007	.748	.131	99.114	1937556.0	.325	70.159	21.171	6.650	1.695	8596.0
32	.007	.698	.140	99.154	948957.0	.394	68.288	22.251	7.128	1.940	4527.0
33	.007	.694	.140	99.159	1109187.0	.407	67.735	22.588	7.251	2.019	5295.0
34	.007	.722	.136	99.133	1034128.0	.384	68.830	21.906	7.002	1.878	4839.0
35	.007	.718	.135	99.140	7480.0	.370	68.865	21.891	6.998	1.877	34.0

TOTAL GRAMS AND AVERAGE WEIGHT PERCENT

GRAMS	1445.2	150681.9	27535.0	20338400.0	20518103.0	326.6	64632.9	19801.9	6261.8	1600.4	92624.0
WT%	.007	.734	.134	99.124		.353	69.780	21.379	6.760	1.728	

OTHER PARAMETERS DETERMINED FROM ABOVE DATA

	PLUTONIUM ATOM RATIOS				URANIUM ATOM RATIOS			Pu/U RATIO ^a	U235 DEPL. ^a	CONVERSION RATIO ^a
	238/239 ^a	240/239 ^a	241/239 ^a	242/239 ^a	U234/U238	U235/U238 ^a	U236/U238			
0	.000000	.000000	.000000	.000000	.000134	.015428	.000174	.000000	.000000	.000000
18	.006041	.327126	.109066	.029604	.000072	.007066	.001434	.004701	.538667	.576820
19	.005093	.251170	.083213	.021824	.000072	.007150	.001424	.004728	.533333	.585846
20	.005253	.310174	.098198	.024903	.000072	.007469	.001363	.004515	.512667	.581941
21	.005146	.306994	.100145	.024865	.000082	.007984	.001456	.004578	.479333	.630741
22	.005284	.310438	.098712	.024715	.000082	.007510	.001404	.004487	.510000	.581424
23	.005399	.318678	.100481	.02504	.000072	.007366	.001394	.004576	.519333	.582291
24	.005052	.307604	.098167	.024146	.000072	.007489	.001333	.004401	.511333	.568837
25	.005266	.312914	.098981	.024982	.000072	.007500	.001363	.004469	.510667	.578367
26	.004901	.304209	.095910	.023730	.000072	.007613	.001343	.004450	.503333	.584295
27	.004615	.300692	.092373	.022762	.000072	.007643	.001313	.004531	.501333	.597168
28	.004556	.298708	.090119	.022251	.000062	.007685	.001323	.004208	.498667	.557833
29	.004561	.295655	.090111	.022344	.000072	.007602	.001343	.004457	.504000	.584377
30	.004580	.294707	.088824	.021529	.000072	.007664	.001313	.004323	.500000	.571452
31	.004652	.300498	.093997	.023859	.000072	.007643	.001333	.004437	.501333	.584787
32	.005794	.324481	.103513	.028056	.000072	.007130	.001424	.004771	.534667	.589565
33	.006034	.332084	.106159	.029437	.000072	.007088	.001424	.004774	.537333	.587063
34	.005602	.316934	.100883	.026946	.000072	.007376	.001404	.004679	.518667	.596075
35	.005395	.316556	.100774	.026918	.000072	.007335	.001373	.004545	.521333	.576236

TABLE II CONTINUED

BWR Group 3

BATCH NO.	URANIUM WEIGHT PERCENTS				GRAMS U	PLUTONIUM WEIGHT PERCENTS					GRAMS PU
	U234	U235	U236	U238		Pu238	Pu239	Pu240	Pu241	Pu242	
0	.017	2.300	.023	97.660	100.0	.000	100.000	.000	.000	.000	.0
36	.012	1.686	.148	98.154	726004.0	.203	79.875	14.810	4.501	.611	2546.0
37	.013	1.736	.151	98.100	866797.0	.190	80.365	14.433	4.473	.538	3120.0
38	.013	1.737	.150	98.100	954645.0	.192	80.233	14.654	4.358	.563	3177.0
39	.013	1.722	.154	98.111	895695.0	.196	80.079	14.581	4.594	.550	3313.0
40	.013	1.789	.145	98.053	1798804.0	.172	81.288	13.901	4.164	.474	6013.0
41	.014	1.855	.131	98.000	1780458.0	.155	82.955	12.896	3.619	.375	5430.0
42	.013	1.779	.146	98.062	870815.0	.186	80.771	14.228	4.276	.539	2982.0
43	.013	1.735	.154	98.098	964390.0	.184	80.492	14.382	4.426	.516	3494.0
44	.013	1.730	.155	98.102	34070.0	.193	80.189	14.514	4.496	.607	123.0

TOTAL GRAMS AND AVERAGE WEIGHT PERCENTS											
GRAMS	1166.5	157315.5	12944.7	8720250.9	8891678.0	54.7	24454.9	4256.4	1279.4	152.2	30198.0
wt%	.013	1.769	.146	98.072		.181	80.982	14.096	4.237	.504	

OTHER PARAMETERS DETERMINED FROM ABOVE DATA

	PLUTONIUM ATOM RATIOS				URANIUM ATOM RATIOS			Pu/U RATIO ^a	U235 DEPL. ^a	CONVERSION RATIO ^a
	238/239 ^a	240/239 ^a	241/239 ^a	242/239 ^a	U234/U238	U235/U238 ^a	U236/U238			
0	.000000	.000000	.000000	.000000	.000177	.023852	.000238	.000000	.000000	.000000
36	.002552	.184641	.055882	.007554	.000124	.017397	.001521	.003507	.266957	.563745
37	.002374	.178844	.055196	.006611	.000135	.017923	.001552	.003599	.245217	.628969
38	.002403	.181881	.055865	.006930	.000135	.017933	.001542	.003328	.244783	.583179
39	.002458	.181323	.056891	.006783	.000135	.017776	.001583	.003699	.251304	.630649
40	.002125	.170296	.050799	.005759	.000135	.018479	.001491	.003343	.222174	.644467
41	.001876	.154809	.043263	.004464	.000145	.019171	.001348	.003050	.193478	.674708
42	.002313	.175417	.052500	.006590	.000135	.018374	.001502	.003424	.226522	.647482
43	.002296	.177930	.054530	.006331	.000135	.017913	.001583	.003623	.245652	.631922
44	.002417	.180242	.055601	.007476	.000135	.017860	.001593	.003610	.247826	.624277

^aThe use of 4, 5, or 6 digits is not intended to reflect accuracy but resulted from the format of the computer program. The numbers should be rounded off to 3 and 4 digits, depending on the parameter involved.

these data, the total grams and average weight percents of each isotope for each group were determined. A computer code was used to manipulate the data to reconstruct the basic mass spectrometry data, that is, the isotopic atom ratios corrected only for mass spectrometry bias. Other burnup parameters determined from the data and tabulated in Table II include:

- a. Pu/U ratios
- b. ^{235}U fractional depletion, where weight percents were used in the equation,

$$^{235}\text{U Fractional Depletion} = \frac{^{235}\text{U}_0 - ^{235}\text{U}}{^{235}\text{U}_0} \quad 3$$

(The quantity $^{235}\text{U}_0$ is the initial weight percent.)

- c. Conversion ratio, CR, which is defined as the plutonium measured to the ^{235}U consumed and is determined by

$$\text{CR} = \frac{\text{Pu measured (grams)}}{(\text{Initial Uranium}) w/f_0^{235} - (\text{U measured}) w/f^{235}} \quad 4$$

The quantity w/f_0^{235} is the initial ^{235}U weight fraction and the initial uranium (grams) is approximated by

$$\text{Initial Uranium (grams)} = \text{Pu measured (grams)} + \text{U Measured (grams)} \quad 5$$

This by no means exhausts the possible parameters to be determined from the data, but these parameters will satisfy our present needs.

CONSISTENCY OF DATA

Our approach to testing the consistency of the reprocessing plants input accountability data will be to plot the data beginning with isotopic atom ratios. Inconsistent atom ratios and the dissolution batch they represent will be identified first. The next step will be to determine which individual atom ratio is at fault and then to identify a single isotope or other data

that appear inconsistent. The consequence of data inconsistency will be discussed as we proceed.

A systematic check of the isotopic uranium and plutonium mass spectrometry data requires plotting at least three graphs. The uranium atom ratios for each of the 85 dissolution batches are shown in Figure 1, where $^{236}\text{U}/^{238}\text{U}$ atom ratio data are shown as a function for $^{235}\text{U}/^{238}\text{U}$ atom ratio data. It was indicated in Reference 3 that four dissolution batches of the PWR group 3 contained fuel of different initial enrichments. Although these data are shown, the consistency checks do not apply because fuel of differing initial enrichments were dissolved together. These data are excluded from the discussions that follow.

Data points representing the initial atom ratio values are also included in Figure 1 to illustrate that burnup paths are continuous functions. In this case, each burnup path is approximated by a straight line as drawn by inspection. Actually, the true function is not a straight line since the slope is equal to

$$\text{Slope} = \frac{\hat{\alpha}^{235}}{\hat{\alpha}^{235} + 1} - \frac{\hat{\sigma}_c^{236}}{\hat{\sigma}_a^{235}} \frac{N^{236}}{N^{235}} \quad (6)$$

where $\hat{\alpha}^{235}$ is the ratio of capture to fission in ^{235}U . However, a straight line approximation serves well to indicate the consistency of the uranium isotopic data. The burnup paths as traced by the uranium atom ratios are separate and distinct, representing the burnup of different reactors as well as the burnup of fuels of differing initial enrichments in the same reactor. Where data from a first core are available, the data of a second core of the same initial enrichment do not vary markedly from the straight line drawn by inspection through the data of the first core.

FIGURE 1
 $^{236}\text{U}/^{238}\text{U}$ Atom Ratio Data as a Function of $^{235}\text{U}/^{238}\text{U}$ Atom Ratio Data Illustrating
 the Consistency of Isotopic Uranium Mass Spectrometry Data

The consistency of the majority of the data makes it possible to identify inconsistent data. The error bars shown in Figure 1 are $\pm 3\%$ for each parameter and data from two dissolution batches lie outside these limits. These two dissolution batches are listed in Table III and are identified in Figure 1 by the numbers 1 and 2 (number 1 through 12 shown in Figure 1 correspond to numbers 1 through 12 in Table III and are used to indicate dissolution batches that are inconsistent in some part of their data). Whether the questionable atom ratios plot high or low is noted in Table III, column A. Any of the three isotopes involved, ^{235}U , ^{236}U , or ^{238}U , could be at fault since it is not known which ratio is inconsistent at this point. Of greatest concern is whether ^{238}U or ^{235}U is in error. If ^{238}U is at fault, it will have a large effect on the remaining uranium isotopes and some affect on the Pu/U ratio. If ^{235}U , a fissionable isotope, is at fault, it will affect the ^{235}U depletion, the conversion ratio, and the number of grams of ^{235}U , but not the total uranium.

The plutonium isotopic data are presented in Figure 2 where $^{242}\text{Pu}/^{239}\text{Pu}$ atom ratio data are shown as a function of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio data. The $^{241}\text{Pu}/^{239}\text{Pu}$ ratio is not presented since a decay correction must be applied to the ^{241}Pu concentrations in order for these data to be consistent with other Pu concentrations. The data in Figure 2 are plotted on semi-log coordinates because ^{242}Pu concentration changes rapidly and so the initial values are not included. The curves shown were drawn by inspection to represent the burnup paths whose functions are considerably more complex than for the uranium data. The burnup paths as traced by the plutonium atom ratio data are separate and distinct when representing the burnup of different reactors. However, a change in ^{235}U initial enrichment for the PWR group 3 did not result in a marked change in the plutonium isotopic ratios as did a ^{235}U initial enrichment change for the BWR group 3.

TABLE III

IDENTIFICATION OF DISSOLUTION BATCHES WHOSE DATA APPEAR INCONSISTENT

Identification			A. Figure 1		B. Figure 2		C. Figure 3		D. Figure 4		E. Figure 5		Comments	
Reactor & Group No.	Batch No.		$\frac{236}{238}\text{U}$	$\frac{235}{238}\text{U}$	$\frac{242}{239}\text{Pu}$	$\frac{240}{239}\text{Pu}$	$\frac{240}{239}\text{Pu}$	$\frac{235}{238}\text{U}$	$\frac{235}{\text{U}}$	Depl.	Conv.	$\frac{235}{\text{U}}$		Ratio
1	BWR 2	21	High	High				O.K.	High	High	Low	High	Low	The ^{238}U is low since a 5.3% decrease in both uranium ratios improves the data everywhere.
2	BWR 3	35	Low	Low				O.K.	Low	Low	High	Low	High	The ^{238}U is high by 2.9%, which makes only a -0.04% change in the ^{238}U weight percent.
3	PWR 1	12			High	Low		O.K.	O.K.	O.K.	O.K.	O.K.	O.K.	^{242}Pu is high since the batch data appear good everywhere except Figure 2.
4	PWR 1	15			Low	High		O.K.	O.K.	O.K.	O.K.	O.K.	O.K.	^{242}Pu is low since the batch data appear good everywhere except Figure 2.
5	PWR 2	32			Low	High		O.K.	O.K.	O.K.	O.K.	O.K.	O.K.	Same comments as 4.
6	BWR 1	12			High	Low		Low	O.K.	O.K.	O.K.	O.K.	O.K.	A 6.8% increase of the $\frac{240}{239}\text{Pu}$ ratio provides the most consistent estimated data. The ^{239}Pu is thus high and its weight percent decrease 1.3%.
7	BWR 1	17			High	Low		High	O.K.	Low	O.K.	Low	O.K.	All the Pu ratios appear to be in question since the batch data are inconsistent in Figure 2, 3, 4, and 5.
8	BWR 2	19			High	Low		Low	O.K.	O.K.	O.K.	O.K.	O.K.	A 28.5% increase of the $\frac{240}{239}\text{Pu}$ ratio (Fig. 3) gives the most consistent estimated data. Although the Pu/U ratio looks good, Figure 4, it is definitely questioned.
9	BWR 3	44			High	Low		O.K.	O.K.	O.K.	O.K.	O.K.	O.K.	Same comments as 3.
10	BWR 2	28								Low	O.K.	Low	O.K.	The isotopic data for uranium and plutonium appear good, Figure 1, 2, and 3, such that the determinations of uranium and plutonium concentration are questioned. These were measured by amperometric titration or spike techniques for uranium and spike techniques for plutonium.
11	BWR 3	38								Low	O.K.	Low	O.K.	
12	BWR 3	41								High	O.K.	High	O.K.

FIGURE 2
 242_{Pu}/239_{Pu} Atom Ratio Data as a Function of 240_{Pu}/239_{Pu} Atom Ratio Data Illustrating the Consistency of Isotopic Plutonium Mass Spectrometry Data

Again, because of the consistency of a large part of the data, it is possible to identify inconsistent data. The error bars shown in Figure 2 are also $\pm 3\%$ and data that lie outside or near these limits are listed in Table III and identified in Figure 2, numbers 3 through 9. Whether or not the ratios are high or low is noted in column B of Table III. (The uranium atom ratio data for these samples are consistent from Figure 1.) Of the three isotopes forming these plutonium atom ratios, ^{239}Pu is the most critical since it is a fissile isotope and the most abundant of the plutonium isotopes. If the ^{239}Pu value is in error, it would affect the remaining plutonium atom ratios, Pu/U ratio, conversion ratio, the number of grams of ^{239}Pu , and total plutonium.

To test the consistency between uranium mass spectrometry determinations and plutonium mass spectrometry determinations, which were done separately, $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio data are shown as a function of $^{235}\text{U}/^{238}\text{U}$ atom ratio data in Figure 3. The curves were drawn by inspection to represent the burnup paths being traced by these two parameters and it appears that the burnup path of the BWR can be approximated by a straight line. The paths representing the burnup of different reactors as well as the burnup of cores of different initial enrichments in the same reactor are separate and distinct. However, the straight line representing the burnup of the BWR group 1 does not agree exactly with the straight line drawn through the data of the second group. Either the burnup path is not a straight line or a possible mass spectrometer bias occurred when fuel having increased concentrations of the higher masses in the plutonium chain were analyzed.

Nevertheless, the consistency of each group of data is good and permits the identification of questionable data, or, as will be used in this case, good

FIGURE 3

240_{Pu}/239_{Pu} Atom Ratio Data as a Function of 235_U/238_U Atom Ratio Data
 Illustrating the Consistency Between Uranium and Plutonium Mass Spectrometry Data

data. Of particular interest are the dissolution batches listed 1 through 9 in Table III. Data from these batches have been marked as inconsistent either from Figure 1 or 2 and it is desired to learn which of the atom ratios considered is actually at fault. If the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios as a function of the $^{235}\text{U}/^{238}\text{U}$ atom ratios appear consistent in Figure 3, then it can be concluded that the $^{242}\text{Pu}/^{239}\text{Pu}$ ratio is inconsistent or the $^{236}\text{U}/^{238}\text{U}$ ratio is inconsistent. If the $^{242}\text{Pu}/^{239}\text{Pu}$ ratio is inconsistent it has been marked inconsistent from information learned from Figure 2 and so noted in Column B of Table III. If the $^{236}\text{U}/^{238}\text{U}$ ratio is inconsistent, it has been marked inconsistent from the information learned from Figure 1 and so noted in Column A of Table III. If the batch data appear inconsistent in Figure 3, then the $^{242}\text{Pu}/^{239}\text{Pu}$ and $^{236}\text{U}/^{238}\text{U}$ ratios can be considered consistent and either the $^{240}\text{Pu}/^{239}\text{Pu}$ or $^{235}\text{U}/^{238}\text{U}$ atom ratios, in Figure 2 and 1 respectively, are inconsistent. From the information learned from Figures 1 and 2 which is noted in Table III, columns A and B, one or the other of these last two ratios can be identified as inconsistent. The ratio at fault has been noted in Column C, Table III in this last case. We have thus eliminated three of four atom ratios as being inconsistent and identified one ratio per batch which is at fault. The $^{235}\text{U}/^{238}\text{U}$ atom ratio is marked inconsistent for two dissolution batches, numbers 1 and 2, Table III, the $^{240}\text{Pu}/^{239}\text{Pu}$ is inconsistent for number 6, 7 and 8, Table III, and the $^{242}\text{Pu}/^{239}\text{Pu}$ atom ratio for 3, 4, 5 and 9.

Since the parameters at fault are isotopic atom ratios, two isotopes are suspect for each ratio. By presenting additional data and examining them for consistency, more information will be learned such that individual isotopes can be identified as inconsistent. (Note that the inconsistency of the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio, number 7, Table III, Column C, is in the opposite direction from that noted in Column B, indicating that more than one Pu isotope is inconsistent or other factors not considered to this point are involved.)

Having checked the isotopic uranium and plutonium data and finding the greater part of these data consistent, it is expected that the uranium and plutonium concentrations measurements for each dissolution batch will exhibit consistency also. Analytical measurements were by amperometric titration or isotopic dilution techniques for uranium concentrations and by isotopic dilution techniques for plutonium concentrations. These additional data can be examined by checking the Pu/U ratio which is shown as a function of ^{235}U fractional depletion in Figure 4. The burnup paths as traced by these parameters are represented by straight lines (drawn by inspection). Data from each reactor and from fuels of differing initial enrichment in the same reactor indicate straight line burnup paths with different slopes.

The dissolution batches 1 through 9, Table III, are identified in Figure 4. Only three of these appear questionable, batches marked 1, 2 and 7, and are thus indicated in Column D. Dissolution batches marked 10, 11 and 12 are also inconsistent and are so indicated in Column D (the isotopic data for these batches were consistent from Figure 1, 2 and 3).

The data of Figure 5 are presented as the last set of parameters illustrating data consistency. Conversion ratio data are shown as a function of ^{235}U fractional depletion to indicate possible variations of the Pu/U ratio. The conversion ratio is essentially a constant for fuel from each reactor, changing only due to reactor type or due to a variation in initial fuel enrichment. This parameter is constant to within $\pm 2\%$ (one standard deviation) when data from each group are averaged and has a smaller uncertainty when questionable samples are eliminated from the average, e.g., the average ratio of PWR group 2 dissolution batches is 0.5697 ± 0.0042 or $\pm 0.74\%$ (one standard deviation).

The same dissolution batches are found questionable from Figure 5 as were indicated in Figure 4, as expected. Thus, with the five data plots, 12

FIGURE 4
Plutonium to Uranium Ratio as a Function of ^{235}U Fractional Depletion Illustrating the Consistency of Plutonium and Uranium Concentrations as Measured

FIGURE 5
Conversion Ratio as a Function ^{235}U Fractional Depletion Illustrating the Consistency of this Burnup Parameter.

dissolution batches have been identified as having some part of their analytical data inconsistent. Three of the batches, 10, 11 and 12 of Table III, are in error because of the measurements made to determine the uranium and plutonium concentrations. This is indicated in the comment column of Table III. The remaining batches have inconsistent isotopic data and some of the isotopes do not greatly affect the Pu/U ratio. To isolate the isotope or isotopes at fault, an estimated value of the suspect atom ratios is obtained from Figure 1, 2 or 3, and all parameters are recalculated based on the estimated value or values that become available. The consistency of each set of estimated parameters determined from the estimated atom ratios will generally identify one isotope which is at fault. For example, consider dissolution batch one, Table III. Estimated values for $^{236}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ atom ratios from Figures 1 and 3 are listed in Table IV. The $^{235}\text{U}/^{238}\text{U}$ ratio has been identified as inconsistent and since estimates of the $^{235}\text{U}/^{238}\text{U}$ ratio from Figure 1 and Figure 3 do not agree, ^{238}U is suspected rather than ^{235}U . Using the $^{235}\text{U}/^{238}\text{U}$ estimated value from Figure 3, it is found to be 5.3% lower than the measured value (also listed in Table IV). The $^{236}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ atom ratios are decreased by 5.3% and all uranium isotopic parameters are redetermined based on the estimated ratios (except the uranium concentration since all necessary data to do this are not available). The recalculated values are listed and can be checked for consistency in Figures 1 through 5. The estimated parameters are found to be consistent. Actually, although the atom ratios changed ~5%, the ^{238}U weight percent increased very little, ~0.5%. Where a noticeable change occurred, there was a 5.3% decrease of ^{235}U weight percent due to ^{238}U being inconsistent. Thus, the number of grams of ^{235}U is high by 5% for this batch which amounts to ~ 400 grams of ^{235}U . The fact that ^{238}U is inconsistent is noted in the comments of Table III.

TABLE IV

Estimated Values of Uranium Atom Ratios from Figures 1 and 3. The Bottom Line Lists the Estimated Parameters Based on the $^{235}\text{U}/^{238}\text{U}$ Ratio Value from Figure 3.

	$\frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{235}\text{U}}{^{238}\text{U}}$	$\frac{^{234}\text{U}}{^{238}\text{U}}$	^{235}U Dep.	C.R.
Figure 1	0.00128	0.00680			
Figure 3	————	0.00755			
MEASURED	0.00145	0.00798	0.000082	0.479	0.631
ESTIMATED	0.00138	0.00755	0.000078	0.508	0.596

Continuing this general procedure for dissolution batches 2 through 9, an isotope or isotopes are identified as inconsistent and the particular isotope has been noted in the comment column. The ^{238}U has also been identified as in error for dissolution batch number 2 Table III, and, in this case, the number of grams of ^{235}U is low by ~ 340 grams. The isotope that is inconsistent for dissolution batches marked 3, 4 and 5 is the ^{242}Pu isotope and is both high and low as noted. The consistency of the data of batch numbered 6 is improved by identifying ^{239}Pu as being high. However, a discrepancy still is present and it is possible that another factor has not been considered. It was not possible by the general procedure to identify any specific plutonium isotope of batch numbered 7 other than to indicate that all are in question. One other possible explanation is that the plutonium isotopics for batches numbered 6 and 7 have been interchanged through some labeling error. If this were the case, ^{242}Pu would be inconsistent and high for both batches, since the $^{240}\text{Pu}/^{239}\text{Pu}$ ratios would appear consistent in Figure 3. Whatever the reason, the Pu/U ratios are in doubt because consistent data cannot be estimated.

Dissolution batch numbered 8, Table III, is made consistent by increasing the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio 28.5% (Figure 3) and ^{239}Pu is at fault. This increase decreases the ^{239}Pu weight percent by 7.3%. Although the Pu/U ratio appears consistent, Figure 4, it is definitely questionable. Based on the amount of total plutonium listed in Table II, at least 340 grams of ^{239}Pu are involved. Dissolution batch numbered 9, Table III, is inconsistent due to ^{242}Pu which is high. The ^{242}Pu does not affect the other plutonium isotopes greatly and has little influence on the Pu/U ratio.

Generally speaking, the estimated values should be verified by remeasurement. This is possible if six of the eight or ten sample vials are still available for the dissolution batches in question. In practice, consistency checks should be applied shortly after the data are taken, when remeasurement can be readily performed.

CONSISTENCY APPLIED TO SAFEGUARDS PROGRAM

It is evident from the consistency checks presented that the accuracy of chemical plant measurements on input dissolver solutions can be substantiated by examining the data for internal consistency. In the case of the above dissolved fuels data outside or near the limits of $\pm 3\%$ for each burnup parameter considered have been identified by a systematic approach of plotting the data and examining each individual dissolution batch. By taking advantage of the consistency of the majority of the data, consistent values for inconsistent parameters were estimated resulting in improved data. Thus, the accuracy of data measurements can be verified by the consistency of the data and, where necessary, the accuracy can be improved by remeasurement of inconsistent data. Or, stated in another way, consistency checks indicate which analytical measurements a safeguards team should verify by surveillance or by independent measurement to verify the accuracy of the whole campaign.

Of value, also, is the fact that consistency checks of mass spectrometry isotopic data will aid plant laboratory personnel or a safeguards team to know if a bias exists in regions of differing isotopic concentrations, where the ^{240}Pu has increased a factor of 1.5, for example, over previous concentrations. (It is possible that a bias is present between the plutonium isotopic data of BWR 1 and BWR 2.) Biases between machines at different sites will also enter into the accuracy of accountability data. Checking the consistency of data taken on a central safeguards machine concurrently with data from various sites will help eliminate any bias between machines and improve the overall accuracy of accountability data.

Examining the consistency of the data demonstrated also that the various burnup parameters considered trace distinct, continuous burnup paths characteristic of a given reactor and its fuel loading. The burnup paths serve then to fingerprint a reactor since the inherent relationships between plutonium formation as a function of uranium and plutonium isotopic compositions do not change for a given reactor unless some reactor parameter, such as the enrichment, is changed. If an inspection team has available the fingerprint of a reactor, data measurements of dissolved fuel from the reactor can be verified. The fingerprint can be developed from accountability data from a first dissolution campaign of a given reactor fuel. Least square fitting codes (for example, program DUBLIN⁽⁴⁾ which fits the burnup equations⁽⁵⁾ to the experimental data) can be used to generate the fingerprint curves rather than using paths drawn by inspection, as was the case with the present data.

We are not restricted just to the burnup parameters presented above for a complete reactor fingerprint. The accountability data can be manipulated to form other parameters, some of which are sensitive to certain reactor parameters and not sensitive to others. The Pu/U ratio could be checked for consistency.

when compared to any burnup parameters to aid verification. For example, the possibility of checking the Pu/U ratio against the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio or against a combination ratio of $(^{240}\text{Pu}/^{239}\text{Pu})/(^{236}\text{U}/^{235}\text{U})$ is shown in Figure 6 (the combination ratio appears to be insensitive to enrichment for the PWR data). The curves shown represent the various burnup paths and were drawn by inspection using the batch data except that the individual batch data are not shown. The data that are indicated are the average values as determined from uranium and plutonium totals and average weight percents, Table II, for each group of fuel. These data illustrate the checks for consistency of the Pu/U ratio against two burnup parameters that would be done for all accountability data as part of verifying the Pu/U ratio measured by a reprocessing plant. The plant results would have to be consistent with all burnup paths to within the accuracy of the paths before the results would be accepted.

Another approach available to a safeguards team once a reactor fingerprint has been developed is to make independent estimates of the plutonium content of the core. This is possible if the team has available the dissolved fuel's isotopic data and a reliable value of the input uranium based on fabrication data. The Pu/U ratio could then be estimated from the fingerprint and the plutonium content determined by multiplying the ratio by the fabrication value of the uranium reduced slightly to account for ^{235}U depletion and plutonium formation. This approach would tend to discourage alteration of dissolved solution volume measurements which can not be checked for consistency in the same manner as the rest of the accountability data. The estimate could be determined on an individual batch measurement basis or on a composite sample measurement representing the total core. To illustrate this procedure, we can estimate the amount of plutonium for batch marked 11, Table III. The isotopic data for the batch have been shown to be consistent and the uranium concentration will be assumed to be correct. An

FIGURE 6

Curves Illustrating the Partial Fingerprint of a PWR and a BWR. The Average Values of Each Core are Shown to Illustrate the Method of Verifying these Values

average Pu/U value of 0.0036 is obtained from Figure 6 corresponding to the $^{240}\text{Pu}/^{239}\text{Pu}$ and $(^{240}\text{Pu}/^{239}\text{Pu})/(^{236}\text{U}/^{235}\text{U})$ values for the batch. Multiplying the Pu/U ratio by 954645.0 gms of uranium, the estimated amount of plutonium is 3437 gms, approximately 8% more plutonium than the amount measured.

These conditions of verification apply not only to data measurements on input dissolution batches, but they also apply to analytical measurements made on the recovered uranium and plutonium solutions as long as the materials from different cores or materials from the same core but from fuel of different initial enrichment are not physically mixed. At the end point in the recovery process, individual batch identity is no longer maintained, and combinations of dissolution batches as recovered material are stored together. The isotopic measurements made on the product solutions should also be consistent and agree with the fingerprint developed for the input isotopic measurement. Even if the materials were stored in containers large enough to represent the total core, the data measurements should agree with the fingerprints. Additional measurements of the product materials are made by whoever receives the recovered fuel from the reprocessing plant. These data should also agree with the fingerprint.

The fingerprint developed from accountability data, then, is a reliable safeguards tool. The consistency of burnup relationships is a verification tool even in the absence of prior information and can be used by itself without surveillance or independent measurements. A necessary requirement would be to have available fuel fabrication data, all the data taken by a fuels reprocessing plant on the input and product solutions, and data taken by a receiver on the product solutions. The data inventory will begin with the fabrication, irradiation, and reprocessing of a first core and be added to as each additional core goes through the fuel cycle. The reactor's fingerprint is developed

empirically from the data of the first core which should not change unless a reactor parameter is changed. Therefore, as the data for a tenth core is taken, they can be verified by data from the first core via the fingerprint.

However, parameters for a given reactor are being changed, as evidenced by the change in enrichment of the initial fuel of both reactors considered. It is expected that even other reactor parameters will change, e.g., fuel cladding, and, each time a change occurs, it is necessary to develop another fingerprint for the given reactor. Rather than depend solely on consistency tests and independent measurements to develop a fingerprint empirically, the new fingerprint could be calculated by using a reactor burnup code.

In practice, reactor burnup predictions are based on approximate methods. The so-called best calculational methods used presently predict burnup parameters to a certain accuracy (for example, $\pm 5\%$ for plutonium concentration).

The accuracy is gradually increasing via advances in base technology such as better measurements of cross sections, improved calculational methods, and more detailed descriptions of reactor systems in a calculation. However, even though discrepancies exist between predicted and measured values, a calculation will provide prior knowledge concerning the shapes of paths traced by burnup parameters that constitute the reactor's fingerprint. Knowledge of the shape of the burnup paths coupled with the consistency of accountability data provides a stronger safeguards tool than either alone. (Having available a calculated shape of the burnup path traced by the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio as a function of the $^{235}\text{U}/^{238}\text{U}$ atom ratio in the case of BWR groups 1 and 2, for example, would help indicate whether or not there actually is a bias between the plutonium isotopic data of the two groups.) Thus, knowledge of the shape of burnup routes through a calculational capability helps the inspection team by aiding the application of consistency checks.

Actually, there is another side to the street we have started down. If the calculated shapes aid an inspection team to apply consistency tests, the accountability data being tested serve to normalize the calculation and are a means of improving the reactor predicted values. Burnup data are taken to experimentally determine the burnup paths for a given reactor and its fuel. These data are used to test the validity of the current computational methods for predicting the production of plutonium and for inferring reactor effective cross section values. The available burnup data have been obtained from incremental samples, i.e., a one-inch fuel sample cut from a rod from a particular position in a fuel bundle, rather than from the dissolution of total elements in one batch as done at a reprocessing plant. Because of neutron spectrum difference as a function of sample position which results in different rates of plutonium generation, identical burnup parameters for example, $^{240}\text{Pu}/^{239}\text{Pu}$, trace several burnup paths within a given reactor. Burnup calculations are performed not only to match the various paths but to predict the average burnup path since the incremental samples which see different spectrums are averaged together when dissolved for reprocessing. Thus, it has not been entirely satisfactory to predict batch dissolution results with calculational codes normalized to experimental data of incremental burnup samples. This is evidenced by the fact that although large groups of incremental burnup data have been obtained, (Yankee Core Evaluation Program) plutonium production has not been predicted to any better than $\pm 5\%$ with confidence. With a safeguards team in place using the data consistency tool to assure the accuracy of accountability data, these data stand a better chance of normalizing a burnup code to the point that the calculations reproduce reality because the actual average parameters a burnup code is to predict are being measured. Then too, a safeguards team would have available accountability data from various reprocessing sites representing fuel from many types of reactors.

The ultimate goal of using one code to predict the plutonium production of all reactors would have the best possibility of being realized if all accountability data are available for code normalization.

Three associated areas are pertinent to aid code normalization processes. Using program DUBLIK, the accountability data will yield ratios of reactor effective cross sections as a result of the least squares analysis.⁽⁶⁾ The ratios of effective cross sections are thus part of the fingerprint and are a means to normalize reactor predictions for plutonium in addition to the isotopic data that are normally compared. The experimental ratios of effective cross sections are one step closer to basic cross section data that are input to a burnup code than are the isotopic data, however, and thus have a greater potential for evaluating the reasons for inaccuracies in burnup predictions and for subsequently reducing the errors of predicted values. An important feature of this general approach is the generation to generation refinement that is possible each time fuel from a core is dissolved.

The second area concerns a major problem that is present in theory-experiment burnup correlations. The calculations are performed in the flux-time domain and utilize total power output as the burnup criteria. The actual reactor values of power are probably not reliable to within 5% at best, and, moreover, are probably systematic in error. Therefore, a reactor needs to be calibrated in terms of measured total fissions versus measured total power output to remove this bias from the calculation. Data taken at a fuels reprocessing plant represent the best solution to knowing the total fissions if fission product data were to be taken, for example, ^{148}Nd . The number of total fissions is also a burnup parameter useful for checking the consistency of burnup data and could be added to Figure 6 as a means of verifying plutonium measurements. A convenient check is made via the isotopic concentrations

versus total fissions, for example, to insure consistency. The purpose of taking fissions product data is then threefold, to assist consistency checks in order to verify plutonium input, to calibrate total power output measured by reactor monitors, and to aid the normalization of burnup predictions. (Without fission product data, it is still possible to determine power output but values of capture to fission ratios must be assumed from reactor calculations for the fissioning isotopes.)

The third area is the significance of obtaining present input accountability data, plus fission product data, as a function of core position. In past dissolution campaigns, fuel elements from a single core have not been dissolved in any predetermined order. The actual order of elements was random and elements of differing initial enrichment were dissolved together. With some preplanning, irradiated elements from a core could be ordered to take advantage of enrichment, total exposure, and final core position of each element. Although preordering the sequence of fuel element dissolution does aid checking for consistency (for example, elements from the center of the core should give a higher Pu/U ratio compared to other fuel from the same core), the main purpose is to provide maximum ease of evaluating theoretical methods used to predict reactor burnup. The isotopic concentrations, plutonium produced, and total power would be available as a function of core position and reactor predictions could be compared region by region to accountability data. This general approach has a large probability of refining reactor calculations.

CONCLUSIONS

Accountability data consistency in concert with the expertise of predicting plutonium production offers an important safeguards tool. The approach is difficult to defeat if a safeguards team is endowed with authority to require:

- A. Standardization of all analytical procedures that are used. An important part of the standardization would be to ensure that biases are eliminated between mass spectrometer machines at various sites.
- B. That all analytical data taken at:
 - 1. A fabrication site, which would include initial enrichment, preirradiation isotopic concentrations, uranium weight per fuel element and element numbers;
 - 2. A reactor site, which would be data to identify core locations of fuel elements, power produced, predicted plutonium production, and dates of charging and discharging fuel elements;
 - 3. A fuel reprocessing plant, which includes all measurements made on input and product solutions plus the identification of fuel elements per dissolution batch; and at
 - 4. A receiving site (may be a fabrication site) which includes all measurements made on product solutions,
 be made available to the team to be checked for consistency.
- C. That accountability codes which handle the analytical data include the same set of conversion constants.
- D. That duplicate analytical samples be retained for analyses by the safeguards team at an independent laboratory or for remeasurement at a designated site.

The safeguards team, equipped with a code that automates the consistency check procedures and with the capability of performing reactor calculations, can then verify the accuracy of data measurements, develop reactor fingerprints, refine reactor calculations, and establish an independent material balance.

In summary, the available accountability data

have been checked for consistency at a level of $\pm 3\%$.

The consistency of the majority of the data made it possible to identify inconsistent data from 12 dissolution batches. Remeasurement of these data would improve the data consistency and drop the level at which inconsistent

data are identified to 1 to 2%. Based on experience with data from other burnup experiments, the accuracy of the bulk of the data taken

is good except for the discrepancy between the plutonium isotopic data of BWR group 1 and group 2. The consistency of burnup parameters has illustrated separate and distinct burnup paths for different reactor types. This result became the basis of developing a reactor's fingerprint as a safeguards tool to prevent and/or detect diversion of plutonium. The tool has a high probability of succeeding.

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