Title:

**Isotopic Compositions of Inventory
Designations, 'Mixed Fission Product' and
'Mixed Activation Product'**

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LA-UR--96-4872

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Submitted to:

U.S. Department of Energy,
as support documentation for the
Los Alamos Area G Performance Assessment

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LANL Area G Performance Assessment Appendix:

**Isotopic Compositions of Inventory Designations,
'Mixed Fission Product' and 'Mixed Activation Product'**

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Abstract

The Area G Performance Assessment requires that the entire inventory be specified in terms of specific nuclide activities. The inventory data base has historically allowed some less specific inventory designations including Mixed Fission Products (MFP), Mixed Activation Products (MAP), several Material Type (MT) designations and others. This report describes the assignment of specific nuclide activities from the listed activity amounts in two of the inventory data base categories, MFP and MAP. The MFP nuclide assignment is based on standard fission product yield data and normalized to total activity at two years post-fission. The MAP assignment is based on site-specific analyses conducted by the major generator of the activation products at Los Alamos, the LAMPF.

Isotopic Compositions of Inventory Designations, 'Mixed Fission Product' and 'Mixed Activation Product'

Mixed Fission Product

The inventory data base for the Los Alamos Area G low level radioactive waste disposal facility contains the entry, 'Mixed Fission Products' (MFP), for waste disposed of prior to 1992. Each MFP waste disposal was assigned a total Curie activity quantity which must be interpreted in terms of activities of specific nuclides for the Performance Assessment.

The data used to define the nuclide composition of MFP is summarized in the Table (next page), *Mixed Fission Product Data Sheet*. The listed nuclides were taken from the standard Table of Isotopes (or Chart of the Nuclides, GE Nuclear Energy, San Jose, CA, 1996) including all isotopes in the fission product yield range, atomic mass numbers 78-160, which have a half-life of at least one year. This was selected as a practical cut-off to avoid tracking too many shorter lived isotopes. While the PA follows consequences only for nuclides with half life greater than 5 years, the nuclides with halflife between one and five years are important in determining the total activity in the inventory as reported for the waste disposal records.

The column, *FPY/M# - ave.* in the Table, is the fission product yield (fractional yield per fission) for each atomic mass number. This was taken from data plots (available in most standard nuclear engineering texts) which show yields for thermal and fast neutrons. To be conservative for an unknown fission source we assumed the maximum of the thermal or the fast yield curve. Differences between different fissionable materials are small and neglected. Errors in this term, *FPY/M# - ave.*, are small, less than a few tens of a percent.

The next column, *FPY isot.split*, shows the fractional correction for isotopic yield for a given atomic number. This assumes conservatively that all the yield at a given atomic number results in the production (or decay to) the long lived species listed in the table. This assumes no stable isotopes are produced in the yield. In those cases where two long lived nuclides exist at the same atomic mass, one on each side of the stable nuclide curve, then it is assumed that the isotopic yield of each is half. Errors in this term are

Mixed Fission Product Data Sheet

nuclide, i	mass number	FPY/M# - ave.	FPY-isot. split	half-life (yrs)	Activity_per_FY	Act.i/sumAct0	Act/FY@2yr	(Act.i/sum)@2yr
Se-79	79	0.0002	1	6.50E+04	2.13E-09	1.58E-08	2.13E-09	4.18E-08
Kr-81	81	0.007	1	2.10E+05	2.31E-08	1.71E-07	2.31E-08	4.53E-07
Kr-85	85	0.02	1	10.7	1.30E-03	9.60E-03	1.14E-03	2.23E-02
Sr-90	90	0.065	1	29	1.55E-03	1.15E-02	1.48E-03	2.90E-02
Nb-92	92	0.065	1	2.00E+07	2.25E-09	1.67E-08	2.25E-09	4.42E-08
Zr-93	93	0.065	0.5	9.50E+05	2.37E-08	1.76E-07	2.37E-08	4.65E-07
Mo-93	93	0.065	0.5	3.00E+03	7.51E-06	5.56E-05	7.50E-06	1.47E-04
Nb-94	94	0.065	1	2.00E+04	2.25E-06	1.67E-05	2.25E-06	4.42E-05
Tc-97	97	0.065	1	2.60E+06	1.73E-08	1.28E-07	1.73E-08	3.40E-07
Tc-99	99	0.065	1	2.13E+05	2.11E-07	1.57E-06	2.11E-07	4.15E-06
Ru-106	106	0.045	1	1.05	2.97E-02	2.20E-01	7.93E-03	1.56E-01
Pd-107	107	0.03	1	6.50E+06	3.20E-09	2.37E-08	3.20E-09	6.27E-08
Ag-108m	108	0.02	1	130	1.07E-04	7.90E-04	1.05E-04	2.07E-03
Cd-109	109	0.01	1	1.24	5.59E-03	4.14E-02	1.83E-03	3.58E-02
Cd-113m	113	0.01	1	14.6	4.75E-04	3.52E-03	4.32E-04	8.46E-03
-								
Sn-121	121	0.015	1	50	2.08E-04	1.54E-03	2.02E-04	3.97E-03
Sb-125	125	0.02	1	2.7	5.13E-03	3.80E-02	3.07E-03	6.02E-02
I-129	129	0.03	1	1.60E+07	1.30E-09	9.63E-09	1.30E-09	2.55E-08
Ba-133	133	0.065	1	10.4	4.33E-03	3.21E-02	3.79E-03	7.43E-02
Cs-134	134	0.065	1	2.1	2.15E-02	1.59E-01	1.11E-02	2.17E-01
Cs-135	135	0.065	1	2.30E+06	1.96E-08	1.45E-07	1.96E-08	3.84E-07
Cs-137	137	0.065	0.5	30.1	7.48E-04	5.54E-03	7.15E-04	1.40E-02
La-137	137	0.065	0.5	6.00E+04	3.75E-07	2.78E-06	3.75E-07	7.36E-06
Ce-144	144	0.065	0.5	0.78	2.89E-02	2.14E-01	4.88E-03	9.58E-02
Pm-144	144	0.065	0.5	1	2.25E-02	1.67E-01	5.63E-03	1.10E-01
Pm-146	146	0.05	0.5	5.53	3.13E-03	2.32E-02	2.44E-03	4.78E-02
Sm-146	146	0.05	0.5	1.00E+08	1.73E-10	1.28E-09	1.73E-10	3.40E-09
Pm-147	147	0.035	1	2.62	9.26E-03	6.86E-02	5.45E-03	1.07E-01
Gd-148	148	0.025	1	93	1.86E-04	1.38E-03	1.84E-04	3.60E-03
Eu-150	150	0.01	0.5	35	9.90E-05	7.33E-04	9.52E-05	1.87E-03
Gd-150	150	0.01	0.5	1.80E+06	1.93E-09	1.43E-08	1.92E-09	3.77E-08
Sm-151	151	0.008	1	93	5.96E-05	4.42E-04	5.87E-05	1.15E-03
Eu-152	152	0.005	1	13	2.67E-04	1.97E-03	2.40E-04	4.70E-03
Eu-154	154	0.002	0.5	8.6	8.06E-05	5.97E-04	6.86E-05	1.34E-03
Dy-154	154	0.002	0.5	1.00E+06	6.93E-10	5.13E-09	6.93E-10	1.36E-08
Eu-155	155	0.001	1	4.8	1.44E-04	1.07E-03	1.08E-04	2.12E-03
Tb-157	157	0.0005	1	160	2.17E-06	1.60E-05	2.15E-06	4.21E-05
Tb-158	158	0.0002	1	150	9.24E-07	6.84E-06	9.16E-07	1.80E-05

sum(Act/FPY(t0))=0.135

sum(Act/FPY(2yr))=0.051

uncertain but the estimate tends to overestimate production of the long-lived nuclides in the PA.

One possibility wherein this approximation would not be conservative is if a short lived nuclide decays by a process which changes its mass number, then it might contribute to a long lived daughter product at a different mass number. These decay processes were assumed rare and neglected but followup work should confirm this. The error associated with this type of decay if it occurs is of the same order as the solution.

The half-lives are taken from the Table of Isotopes (or Chart of the Nuclides, GE Nuclear Energy, San Jose, CA, 1996) and from various editions of the Handbook of Chemistry and Physics (CRC Press). There is not good agreement for this fundamental quantity for some of the more exotic species. With this data, the activity per fission is tabulated with no decay correction in the column, *Activity_per_FY*. This is summed to obtain total activity per fission and used to normalize each nuclide contribution to the total activity listed in column, *Act.i/sumAct0*.

It is important to normalize to the total activity at the time that best represents the actual age of the MFP as characterized in the disposed waste. It is assumed that some waste materials are processed in the lab and disposed of in about one year while another significant contribution to the waste will be from clean-up and D&D operations which will occur several years after the MFP was created. A single representative average age for all the MFP is taken to be two years. Thus, the activity per fission for each nuclide was decay corrected to two years in column, *Act/FY@2yr*. This was summed and again used to normalize the nuclide activities per total activity at two years, as summarized in column, *(Act.i/sum)@2yr*.

Figure 1 shows a log scale of nuclide activity normalized to the total activity evaluated at $t = 0$ years (top) and at $t = 2$ years (bottom). A different set of nuclides dominate each case; the shortest lived nuclides which were considered ($\tau > 1$ yr) dominate the evaluation normalized at $t = 0$. Similar results evaluated at $t = 0$, are shown on a linear scale in Fig.2(top) for comparison. The results normalized at $t = 2$ years were taken to define the MFP content for the analyses in the PA. The activity remaining at 100 years and normalized to the total activity at 2 years is seen in Fig.2(bottom). These numbers, *(Act.i/sum)@2yr*, times the activity listed as MFP in the disposal

inventory represents the present best estimate of the actual nuclide inventory comprising MFP. This is the nuclide assignment used in the site PA.

Figure 3 shows a log scale of nuclide activity remaining at subsequent times, $t = 100$ yrs (top) and $t = 10,000$ yrs (bottom) normalized to the total activity evaluated at $t = 2$ years. Note that Fig.2(bottom) and Fig.3(top) show the same data on linear and log plots, respectively. These are the nuclides which could potentially contribute at later times in the assessment period and beyond.

MFP Uncertainty

The nuclide activity sum drops from 0.135 at zero time (no decay correction) to 0.051 with the two year decay correction, or by a factor of almost three, as seen in the Table. This shows the total MFP inventory is sensitive to the estimate of MFP age, and could be used to estimate the inventory uncertainty resulting from a specified age uncertainty.

The estimate for the MFP distribution assumes that the MFP 'total Curie' listed inventory amount is accurately related to the total MFP activity, so that the total activity (at 2 years) is the correct sum for normalizing nuclide amounts. The error in this assumption will depend upon the method used to determine the total MFP activity listed on the waste manifest. For example, calculating total MFP activity from gamma radiation measurements at a distance might reflect mostly just Cs-137 activity, and so the Cs-137 activity would be a better normalization constant in that case. Resolution of this detail would require interviews with waste generators from the past to determine their method of estimating MFP from some actual measurement.

Mixed Activation Products - MAP

The inventory data base contains the entry, 'Mixed Activation Products' (MAP), for waste disposed of prior to 1992. This needs to be defined in terms of specific nuclides for the Performance Assessment. A review of disposal records indicates MAP was primarily generated at LAMPF (Los

Alamos Meson Physics Facility), a high energy accelerator facility. Conversations with current LAMPF staff (John Roberts, LANL, personal communication) lead to the following summary of their waste inventory.

There are three main waste streams from LAMPF, including trash, targets and beam line 'inserts', and targets. Each stream has a similar but distinct waste profile or nuclide composition. Materials in each stream can include activated steel, aluminum, and graphite. Only the 'trash' waste profile has been quantified to date in an ongoing effort between LAMPF and the LANL Health and Safety groups (John Roberts, personal communication). Because the waste streams are similar, this profile will be taken to represent the total profile for inventory listed as MAP.

The MAP profile has been determined to be comprised of the following isotopes by mass fraction.

<u>Nuclide</u>	<u>mass fraction</u>
Co-57	0.12
Co-60	0.44
Mn-54	0.18
Zn-65	0.13
Na-22	0.10
Be-7	0.03

Based on this mass fraction distribution, the approximate distribution by activity follows.

<u>Nuclide</u>	<u>Ci/gram</u>	<u>activity fraction</u>
Co-57	1000	0.065
Co-60	500	0.033
Mn-54	1450	0.095
Zn-65	1080	0.072
Na-22	630	0.042
Be-7	10600	0.693
Total	15300	

The short half-life of Be-7 ($\tau = 54\text{d}$) implies that the normalization constant for total activity is changing rapidly in time. It is assumed that the waste inventory previously characterized as MAP was normalized to total activity in the same way as described here, based on the activity fractions shown above. This implies that the waste was previously characterized in the same time frame that the above mass fractions were determined.

Each of these listed nuclides have half-lives less than 5yr except Co-60. The half-life for Na-22 is 2.6yr, so it is marginally of interest in a PA. If it is retained as a listed nuclide, the effective PA inventory for N_{MAP} Curies of mixed activation products is $(0.033 \cdot N_{\text{MAP}})$ Curies of Co-60 and $(0.042 \cdot N_{\text{MAP}})$ Curies of Na-22. If Na-22 is neglected, the effective PA inventory is just $(0.033 \cdot N_{\text{MAP}})$ Curies of Co-60.

MAP Uncertainty

The major uncertainty in this inventory is the time at which one chooses to categorize the waste because the total activity depends primarily on the short lived nuclide, Be-7.

The beryllium results from graphite activation, sodium from aluminum and the other nuclides result from steel activation. The distribution of activation products is not expected to vary greatly in time or between the different waste streams from LAMPF so this variation introduces smaller uncertainty into the inventory.

Fig.1 Log scale of nuclide activity normalized to the total activity evaluated at $t = 0$ years (top) and at $t = 2$ years (bottom). The evaluation normalized at $t = 2$ years was taken to define the MFP content for the analyses in the PA.

Fig.2 Linear scale of nuclide activity normalized to the total activity evaluated at $t = 0$ years (top) and the activity remaining after 100 years, normalized to the activity present at $t = 2$ years (bottom). A different set of nuclides dominate each case; the shortest lived nuclides which were considered ($\tau > 1$ yr) dominate the evaluation normalized at $t = 0$.

Fig.3 Log scale of nuclide activity remaining at subsequent times, $t = 100$ yrs (top) and $t = 10,000$ yrs (bottom) normalized to the total activity evaluated at $t = 2$ years.