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FISSION-PRODUCT SECURITY PROBLEM -

PRELIMINARY EVALUATION

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FISSION-PRODUCT SECURITY PROBLEM - PRELIMINARY EVALUATION

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FISSION-PRODUCT SECURITY PROBLEM - PRELIMINARY EVALUATION

INTRODUCTION

Hanford is currently involved in the initial phases of a fission-product recovery program involving the recovery for sale of radiostrontium and radio-cesium. Since the fission products would be released for unclassified uses, there is concern about the possibility of revealing irradiation conditions through knowledge of fission-product compositions. At the request of Advance Process Development Operation, Chemical Processing Department, a study was initiated to determine:

- (1) The possibilities of calculating classified information relating to irradiation history and plutonium production from analyses of fission-product concentrates if they are released as unclassified material; and
- (2) If classified information may possibly be divulged, what steps can be taken to mask or distort the fission-product compositions to prevent calculation of significant results without, at the same time, grossly reducing specific activity of the fission-product mixture.

The purpose of this report is to point out results of preliminary studies which indicate some possible security problems. A more thorough analysis, now in progress, will more precisely define the probable errors inherent in the calculations and will more thoroughly explore possible masking procedures. A completely satisfactory procedure may ultimately be developed to allow unrestricted shipments of these materials. However, since initial shipments of fission product cesium and strontium are imminent, it appears advisable to avoid any possibility of compromising restricted data by taking action to safeguard the information indicated to be sensitive by these preliminary studies.

It is assumed in this study that anyone interested in employing fission-product analyses as a means of determining plutonium capability has the knowledge that only natural uranium irradiation is involved in the fission product recovery program.

The basic classification restrictions of importance to this study are presented in the Appendix as excerpts from the "Hanford Classification Guide".

SUMMARY AND CONCLUSIONS

1. Both the strontium and cesium fission-product mixtures include one stable nuclide that has a substantial fission yield and only short-lived precursors. These are Sr-88 and Cs-133. Neither they nor their precursors have appreciable thermal neutron absorption cross sections and the amount of the stable nuclides present after a short period of cooling can be calculated quite accurately by assuming they occur as a direct fission yield. This permits calculation of total Megawatt Days of exposure for given quantities of the fission product mixtures. The original amount of Cs-133 can be masked to some extent by addition of natural cesium (100% Cs-133) with a consequent sacrifice in specific activity, but the Sr-88 cannot be masked with natural strontium because any addition of Sr-88 as natural strontium is readily detectable through measurement

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of the accompanying Sr-86 or Sr-87. Since total Megawatt Days is a rough measure of plutonium production, it is recommended that the relationship of any given amount of fission product cesium or strontium to total capability should be classified. This information is currently treated as classified data so new safeguards are not involved here.

2. The ratio of Cs-135 to Cs-133 is indicated to bear a simple algebraic relationship to reactor equilibrium specific-power level (MW/T) or the equivalent of neutron flux. The effect of reactor shutdowns and outages on this ratio have not been fully evaluated but preliminary calculations indicate that these effects can be corrected for within relatively narrow error limits (less than $\pm 10\%$ error in the calculated MW/T). The ratio can be distorted to a limited extent by addition of natural cesium with the consequent sacrifice in specific activity. It is recommended that cesium fission-product mixtures be considered as classified material until (a) it is determined that any cesium recovered will not reveal sensitive data because it is of such an age that it is not representative of current production, (b) further study indicates much larger probable error in the calculation, or (c) acceptable masking procedures can be recommended.
3. It is not possible to directly determine age-since-reactor-discharge by measurement of daughter nuclides of radioactive cesium and strontium because these nuclides will be separated from the product during processing. (These daughter nuclides are stable Ba-134, stable Ba-137 for cesium, and stable Y-89 and stable Zr-90 for strontium.) Equations based on the long-lived radionuclides, Cs-137 and Sr-90, are very sensitive to small errors because of the small value of the decay constants and their occurrence as exponential functions. In addition, the procedure of accumulating fission-product solutions in large tanks over a long period of time has the effect of distorting the ratios of the radionuclides relative to their average age because of differences in decay rates, (continuous supply of same aged material and different rates of disappearance). These effects result in the introduction of substantial errors into the solution for irradiation and aging time using simultaneous equations based on isotope ratios which might otherwise be significant. However, if aging time were known it may be possible (1) to calculate, from the relative amounts of Cs-133, Cs-134 and Cs-135, both the flux and irradiation time (the equivalent of MWD/T) as well as tons of uranium irradiated to produce a given quantity of cesium fission products or (2) to calculate irradiation time from the relative amounts of Sr-88 and Sr-89. Although the preliminary studies indicate substantial errors may be inherent in these calculations, even with knowledge of aging time, it is recommended that any reference to actual or average aging time as it relates to activity ratios of Cs-134 to Cs-137 or Sr-89 to Sr-90 (especially calculated ratios) or to specific batches of recovered fission products and any reference to actual details of fission-product accumulation procedures should be classified, at least until further studies permit better definition of the probable errors involved.

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FISSION-PRODUCT CESIUM DISCUSSION

Present plans for cesium recovery involve processing waste solutions that are on the order of 1000 days old. The calculated composition of a cesium fission-product mixture from a 600 MWD/T irradiation at 6 MW/T and 1000 days old is shown in Table I.

TABLE I

FISSION-PRODUCT CESIUM COMPONENTS -
(600 MWD/T at 6 MW/T and 1000 Days Old)

<u>Isotope</u>	<u>g/T</u>	<u>Curies/T</u>
Stable Cs-133	23.56	--
2 y Cs-134	0.052	69.4
Stable Cs-135	7.95	--
13 d Cs-136	$\ll 1 \times 10^{-6}$	$\ll 0.05$
26.6 y Cs-137	20.46	2010.00

Although five cesium isotopes are produced during an irradiation, only four are produced in sufficient abundance for measurement. The Cs-136 is produced principally from neutron absorption by Cs-135 and the amount remaining after 1000 days aging is so small that even its detection is unlikely, let alone an accurate measurement. No consideration has, therefore, been given to possible measurement of Cs-136 quantities. Decay chains for the cesium isotopes are presented in Figure 1(1).

Total MWD from Cs-133 Measurement

All the precursors of Cs-133 are short-lived and have negligible cross sections for thermal neutrons. Since we are only interested in the quantity of Cs-133 present after long aging, and since the equilibrium quantities of all its precursors will decay to Cs-133 after a few days aging, it can be assumed that stable Cs-133 is produced as a direct fission yield. The expression for the amount of Cs-133 formed is then

$$W_{Cs133} = \frac{m'}{A} \Sigma_f (x\phi\tau)y' \quad [1]$$

where W_{Cs133} = grams Cs-133 formed

m' = Cs-133 mol. wt.

A = Avogadro's number, 6.02×10^{23} atoms/mole

Σ_f = macroscopic fission cross section, $9262 \text{ cm}^2/\text{TU}$ (average in Hanford reactors for natural uranium)(2)

x = tons U irradiated

ϕ = neutron flux, $n/\text{cm}^2/\text{sec}$

τ = irradiation time, seconds

y' = fission yield fraction, 0.066 ± 0.001

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The only inherent error in this expression results from the fact that a small quantity of Cs-133 is lost by neutron capture, but this is only on the order of 0.2% and can be neglected.

Knowing the quantity of Cs-133 (W_{Cs-133}) associated with a given quantity of cesium concentrate, as determined by mass spectrographic analysis, equation (1) can be solved for ($x\phi\tau$). This is equivalent to determining the total megawatt days of exposure:

$$3.350 \times 10^{-18} (x\phi\tau) = \text{MWD}$$

The number of reactor startups and outages will have no effect on the accuracy of this calculated value.

The total MWD is not an exact measure of plutonium production, since this also depends on the relative values of x , and $\phi\tau$ and the reactor design. It does, however, provide a fair approximation, particularly if some knowledge on the capabilities of Hanford-type reactors is available. Thus, a fair estimate (probably at least within $\pm 10\%$) of plutonium production associated with a given batch of recovered cesium is possible.

The accuracy of the calculation can be distorted by addition of natural cesium (100% Cs-133). However, since the Cs-133 accounts for nearly 50% of the total fission-product cesium, substantial quantities of natural cesium are required and it is added at the expense of lower specific activity. For example, adding 10% natural cesium to the mixture would increase the calculated MWD figure by approximately 20% and reduce specific activity by 10%. Whether or not a distortion in this direction would be desirable from the standpoint of security philosophy is not known. In any event, gross distortions are not practical.

It is concluded that rather than attempt masking to conceal total MWD, the relationship of any cesium recovered to total capability should be classified. This is in accord with present practice so new safeguards are not required. As long as only relatively small quantities of cesium are recovered, there should then be no problem. If, however, we reach the point where nearly all of the cesium is recovered it would be difficult to conceal plutonium production capability.

Flux Determination from Cs-135/Cs-133 Ratio

Potentially the most serious security problem relating to either cesium or strontium recovery involves the high sensitivity of Cs-135 formation to neutron flux during an irradiation. This phenomenon provides a means for calculation of neutron flux through measurement of the Cs-135/Cs-133 ratio in the recovered fission-product cesium.

The concentration of Cs-135 relative to Cs-133 is uniquely adapted to calculation of reactor equilibrium flux levels for the following reasons:

- (1) The immediate precursor of Cs-135, Xe-135, has an extremely high absorption cross section for thermal neutrons (2.7×10^6 barns) resulting in a rate of Cs-135 formation that is highly dependent on neutron flux;

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- (2) Both Cs-135 and Cs-133 are stable nuclides so that, except for the first few days after reactor discharge, their relative concentrations remain constant irrespective of age at the time of measurement. (Cs-135 is in reality slightly radioactive, but its half-life, 2.6×10^6 years, is so long that for practical purposes, it can be considered stable.)
- (3) The precursors of Cs-135 (see Figure 1) are all short-lived so they reach saturation concentrations very quickly after reactor startup resulting in a virtually constant rate of Cs-135 formation during an irradiation.
- (4) Formation of Cs-133 is not appreciably affected by neutron flux and provides the necessary basis of comparison.
- (5) The relatively high fission yield of both Cs-135 and Cs-133 permits accurate measurement of their relative concentrations by mass spectrometer.

During a period of constant flux irradiation, if the loss of Cs-135 by neutron absorption is neglected (approximately 0.1%), the amount of Cs-135 formed can be expressed as

$$W_{Cs135} = \frac{m}{A} \Sigma_f (x\phi\tau) \left[\frac{\lambda_3 y_3}{(\lambda_3 + \sigma_3\phi)} + y_4 \right] \quad [2]$$

where W_{Cs135} = grams Cs-135 formed,

m = Cs-135 mol. wt.,

λ_3 = Xe-135 decay constant, $(2.10 \pm 0.01) \times 10^{-5} \text{ sec}^{-1}$,

σ_3 = Xe-135 absorption cross section, $(2.7 \pm 0.1) \times 10^{-18} \text{ cm}^2$ (3)

y_3 = Xe-135 fission yield fraction, 0.063 ± 0.001 ,

y_4 = Cs-135 independent fission yield fraction, 0.01 ± 0.0005 ,

and other terms are the same as in equation [1].

This expression is exact (except for the small Cs-135 neutron capture effect) during a period of constant flux but in an actual irradiation, the effects of reactor startups and shutdowns must also be factored in. A preliminary evaluation of these outage effects has been made and a method developed for correction of equation [2].

When a reactor shuts down, the saturation concentrations of 6.7 h I-135 and 9.2 h Xe-135 begin to decay to Cs-135. After 48 hours, approximately 95% of the I-135 and Xe-135 has decayed to Cs-135 and further downtime will have little effect on Xe-135 accumulation. Hanford reactor outages average 52 hours (4), so essentially all of the I-135 and Xe-135 backlog decays to Cs-135 at each outage. It is estimated that this Cs-135 accumulation amounts to approximately 2% additional Cs-135 per outage for an average Hanford irradiation.

When a reactor starts up, it takes a period of time for the reactor to reach equilibrium power level and for the I-135 and Xe-135 to reach saturation concentrations, during which time the rate of Cs-135 formation is gradually approaching

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its equilibrium rate of formation. Since equation [2] assumes an equilibrium rate of Cs-135 formation during the entire irradiation, it will indicate more Cs-135 formation than is actually the case. It has been estimated that the difference between the integrated amount of Cs-135 formation during startup period and the amount of Cs-135 that would be formed during an equilibrium operation over the same period, amounts to approximately 0.5% per outage for an average Hanford irradiation.

To complete formulation of an outage correction factor, the number of outages per irradiation must be defined. The number of outages occurring during irradiation of any specific batch of uranium is likely to be widely different from any other specific batch. However, the average number of outages for all discharges from all reactors over a period of time can be stated with fair precision -- the longer the time period involved, the better the precision. Since cesium recovery operations involve processing waste solutions accumulated over a period of months, the average number of outages represented in quantities of recovered cesium can be stated with reasonable precision. The number of outages per irradiation used in this study was taken as 10.4 ± 2.9 . The error limits are based on 95% confidence limits for a 3 month accumulation and should be quite conservative since the waste tanks generally represent at least a 12 month accumulation.

The deviation of an actual irradiation from an equilibrium irradiation as defined by equation [2] can then be stated as

$$(\% \text{ increase per shutdown} - \% \text{ loss per startup}) \times \text{number of outages} =$$

$$(2.0 - 0.5) \times (10.4 \pm 2.9) = + 16 \pm 4\%$$

Equation [2] can then be restated as

$$W_{Cs135} = \frac{m}{A} \Sigma_f (x\phi\tau) \left[\frac{\lambda_3 y_3}{(\lambda_3 + \sigma_3\phi)} + y_4 \right] C \quad [2']$$

where $C = 1.16 \pm 0.04$, a correction factor for outage effects.

Now, dividing equation [2'] by equation [1] and solving for ϕ , we obtain

$$\phi = \frac{\lambda_3}{\sigma_3} \left[\frac{y_3}{\left(\frac{W_{Cs135}}{W_{Cs133}} \times \frac{m'}{m} \times \frac{y'}{C} - y_4 \right)} - 1 \right] \pm 8.4\% \quad [3]$$

where all terms are as previously defined and the error limits are based on 95% confidence limits and a $\pm 1\%$ error in the W_{Cs135}/W_{Cs133} ratio. The flux, ϕ , can be readily converted to specific power:

$$P = 2.894 \times 10^{-13} \phi, \quad \text{where } P = \text{specific power in MW/T.}$$

The composite fission yields for Cs-133 and Cs-135 were calculated considering Pu-239 and U-238 as well as U-235 fission, and found to be well within the error limits previously stated for fission yields.

It has been possible to date to check equation [3] with data from one mass spectrometer analysis on a sample from the Purex 103-A waste tank. Results of the analysis are shown in Table II.

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TABLE II

MASS SPECTROMETER ANALYSIS FOR CESIUM - 103-A TANK⁽⁵⁾

<u>Isotope</u>	<u>Wt. %</u>
Cs-133	47.0 ± 0.3
Cs-134	0.07 ± 0.02
Cs-135	16.2 ± 0.2
Cs-137	36.7 ± 0.3

Based on these data, the average reactor specific power during the period this material was formed was calculated by equation [3] to be 5.47 ± 0.46 MW/T. The 103-A tank was filled during the period from May 1956 to June 1957, representing reactor operations from November 1955 through March 1957. The weighted averaged specific power during this period was determined as 5.32 MW/T and well within the estimated error limits of the calculation based on the sample analysis.

Additional work is planned to more precisely define the reliability of the correlation expressed by equation [3]. This work includes:

- (1) Checking validity of some simplifying assumptions employed in defining the magnitude of the correction factor C.
- (2) Defining the effect of reactor shutdowns that were recovered, that is, startup achieved after brief shutdown and not considered as an outage.
- (3) Defining the effect of variations in the average number of outages over the past 4 to 5 years.
- (4) Checking the correlation with sample analyses on the other 3 Purex waste tanks.
- (5) Exploring the possibility that natural cesium introduced during processing may distort results.

After this work is completed and if results continue to check actual reactor operations, it will be necessary to estimate how closely someone outside Hanford (with extensive reactor experience but with relatively superficial knowledge of our reactor operations) might be able to define the magnitude of the outage correction, C. The absolute security risk can then be established.

For the present, based on the preliminary work reported here, any cesium recovered should be treated as classified material with appropriate safeguards imposed, at least until such time as the security implications can be thoroughly investigated. It may be possible to release material recovered from the 103-A tank since power levels have increased substantially since this material was processed. Whether or not this would be compatible with current security philosophy apparently cannot be determined locally. Masking the true Cs-135/Cs-133 ratio by addition of natural Cs-133 is also a possibility but does not look attractive because of consequent reduction in specific beta and gamma activity.

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Irradiation Time and Aging Time

To calculate anything further than total exposure and flux, it is necessary to develop other relationships that utilize analyses for the radioisotopes Cs-134 and Cs-137. This introduces another unknown, the aging time. Aging time cannot be determined directly by measurement of daughter nuclides since these, stable Ba-134 and stable Ba-137, will be separated from the recovered cesium. Only the amount of daughter nuclides that grow in after recovery operations will be present. Thus, it is necessary either to have aging time provided or to factor it out through use of simultaneous equations if further calculations are to be made.

The quantity of Cs-137 present in a given batch of fission-product cesium can be expressed as

$$W_{Cs137} = \frac{m''}{A} \frac{\Sigma_f}{\lambda} x\phi'y'' (1-e^{-\lambda\tau'}) (e^{-\lambda t}) \quad [4]$$

where W_{Cs137} = grams Cs-137 from irradiation of x tons of uranium,

m'' = Cs-137 mol. wt.,

ϕ' = apparent flux = ϕE , where E = production efficiency,

τ' = apparent irradiation time = τ/E ,

y'' = Cs-137 fission-yield fraction = 0.061 ± 0.001 ,

λ = Cs-137 decay constant = $8.26 \times 10^{-10} \text{ sec}^{-1}$,

t = age since reactor discharge, seconds,

and other terms are the same as previously stated.

Use of the terms, apparent flux and apparent irradiation time, is a means of correcting for the effect of down time on the amount of a radionuclide present at termination of an irradiation. It is not an exact correction but when a large number of reactor discharges are lumped together as is the case here, the error would be very small. It introduces still another unknown, E , but one that can probably be guessed fairly closely -- say 0.75 ± 0.05 .

After solving for $(x\phi\tau)$ from equation [1], a substitution can be made in equation [4]:

$$E \left(\frac{x\phi\tau}{\tau'} \right) = x\phi'$$

$$\text{or } \left(\frac{x\phi\tau}{\tau'} \right) = x\phi'$$

$$\text{and } W_{Cs137} = \frac{m''}{A} \frac{\Sigma_f}{\lambda} \left(\frac{x\phi\tau}{\tau'} \right) y'' (1-e^{-\lambda\tau'}) (e^{-\lambda t}) \quad [4']$$

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If data on age of the fission-product cesium is available, equation [4'] could then be solved for τ' (or τ/E). With a fair guess at the value of E , it would appear that τ and $\phi\tau$, or specific exposure, could be obtained within the limits of error on E :

$$3.350 \times 10^{-18} (\phi\tau) = \text{MWD}/T$$

However, it turns out that extreme accuracy is required for the known values in equation [4']; $W_{\text{Cs-137}}$, $(x\phi\tau)$, y'' , etc. This is due to the insensitivity of the term $(1-e^{-\lambda\tau'})/\tau'$ to small changes in τ' . The decay constant, λ , is a very small number and large changes in τ' are required to change the value of $(1-e^{-\lambda\tau'})/\tau'$. For example, a 3% error, will cause a 10-fold error in the solution for τ' ; an over-all error in the known values on the order of 0.05% must be obtained to limit the error in τ' to around 10%. Just the uncertainty in y'' alone would be enough to destroy the value of equation [4'].

Another possible approach would be through measurement of the Cs-134 and calculation of τ if aging time is known. Although the amount of Cs-134 in the fission-product cesium is quite small, enough is present for determination within less than $\pm 5\%$ error limits by radiochemical methods⁽⁶⁾. Because of the shorter half-life (2.0 years) of Cs-134, the solution for τ using the Cs-134 equation (with age known) is much less sensitive to the type of error inherent in the Cs-137 equation. However, an additional uncertainty is involved since the Cs-134 is all formed through neutron absorption by Cs-133 whose total absorption cross section has a relatively-very-large epithermal component. It may be difficult for someone outside of Hanford to define the effective cross section of Cs-133 with sufficient accuracy. Error limits inherent in this calculation have not been fully evaluated and require further study to define the security risk.

A third possibility for determining both irradiation time and aging time would be through solution of simultaneous equations representing Cs-134 and Cs-137 formation in conjunction with the Cs-133 and Cs-135 equations. It would still be necessary though to estimate production efficiency, or if aging time is known it would be possible, theoretically at least, to solve for production efficiency. Still another difficulty is involved, however. This is the effect of accumulation, in a large tank over a long period of time, on the relative amounts of two radionuclides relative to their average age -- a continuous supply of same aged material and different rates of disappearance. This effect can be illustrated by the following equations:

$$(W_1/W_2)_1 = (W_1/W_2)_0 (\lambda_2/\lambda_1) \frac{(1-e^{-\lambda_1 t_1})}{(1-e^{-\lambda_2 t_1})} \quad [5]$$

$$\text{and } (W_1/W_2)_2 = (W_1/W_2)_0 (e^{-\lambda_1 t_2}/e^{-\lambda_2 t_2}) \quad [6]$$

where a constant rate of accumulation is assumed and,

$(W_1/W_2)_0$ = ratio of two radionuclides in feed solution,

λ_1 and λ_2 = decay constants of the two nuclides,

t_1 = total accumulation or filling time,

t_2 = average age = $t_1/2$,

$(W_1/W_2)_1$ = ratio of radionuclides at end of filling period,

$(W_1/W_2)_2$ = ratio of radionuclides if average age assumed.

In the case of Cs-134 and Cs-137, the difference between $(W_1/W_2)_1$ and $(W_1/W_2)_2$ for a 400 day accumulation is only about 0.7% and may not introduce a significant error. The over-all error limits for simultaneous equations involving Cs-134 and Cs-137 analyses have yet to be evaluated but considering the uncertainties involved they are likely to be substantial.

Although the preliminary studies indicate small possibilities of determining anything further from a cesium analysis other than reactor flux, and although the preliminary studies also indicate that knowledge of aging time may not help too much in determination of irradiation time and specific exposure, the studies are still incomplete and it appears advisable to withhold information on the age of fission-product cesium batches as a security measure, at least until the present study is completed. Information on waste tank filling periods should also be temporarily classified as this is just one more important piece of the puzzle that may be difficult to estimate.

FISSION-PRODUCT STRONTIUM DISCUSSION

Present plans for strontium recovery involve a crude separation of strontium from Purex waste solutions (1 WW) as it leaves the Purex process; accumulation of the crude strontium solution in storage tanks over approximately 2.5 month periods; further storage for 9 to 15 months depending on inventory and process requirements; further processing to a concentrated $\text{Sr}(\text{NO}_3)_2$ solution or a SrCO_3 powder ready for shipment. The age at time of shipment will be on the order of 400 to 600 days from reactor discharge.

Analysis of Hanford fission-product strontium solutions has indicated that about a 5% contamination with natural strontium is picked up during processing⁽⁷⁾. The composition of natural strontium is shown in Table III.

TABLE III

COMPOSITION OF NATURAL STRONTIUM⁽⁸⁾

Sr-84	0.56 wt. %
Sr-86	9.86 wt. %
Sr-87	7.02 wt. %
Sr-88	82.56 wt. %

The calculated composition of a strontium fission-product mixture from a 600 MWD/T irradiation at 6 MW/T and cooled for 400 days is shown in Table IV.

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TABLE IV

FISSION-PRODUCT STRONTIUM COMPONENTS
(600 MWD/T at 6 MW/T and 400 Days Old)

	From Fission Only		Corrected for Natural Sr Content	
	Wt. %	Curies/100 g	Wt. %	Curies/100 g
Stable Sr-84	--	--	0.028	--
Stable Sr-86	0.00138	--	0.49	--
Stable Sr-87	0.00000022	--	0.35	--
Stable Sr-88	38.2	--	40.4	--
50.5 d Sr-89	0.115	3347	0.109	3172
27 y Sr-90	61.7	8756	58.6	8318

Two conclusions are apparent: (1) Only three strontium isotopes (Sr-88, Sr-89, and Sr-90) can be measured with accuracy since the Sr-86 and Sr-87 quantities are so small and in addition are masked by the natural strontium; and (2) The Sr-88 cannot be masked with natural strontium since the amount of natural strontium present can be determined by Sr-86 or Sr-87 measurements. Decay chains for the three important strontium isotopes are shown in Figure 2.

Since the precursors of all three of the important strontium isotopes are all very short-lived and since the absorption cross sections of all the nuclides involved are small, the equations representing the quantity of each isotope present after aging are relatively simple:

$$W_{\text{Sr}88} = \frac{m}{A} \Sigma_f (x\phi\tau) y \quad [7]$$

$$\text{or } \frac{m}{A} \Sigma_f (x\phi'\tau') y$$

$$W_{\text{Sr}89} = \frac{m_1}{A} \Sigma_f (x\phi') \frac{y_1}{\lambda_1} (1 - e^{-\lambda_1\tau'}) (e^{-\lambda_1 t}) \quad [8]$$

$$W_{\text{Sr}90} = \frac{m_2}{A} \Sigma_f (x\phi') \frac{y_2}{\lambda_2} (1 - e^{-\lambda_2\tau'}) (e^{-\lambda_2 t}) \quad [9]$$

where $W_{\text{Sr}88}$ = grams Sr-88 formed,

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m = Sr-88 mol. wt.,

m_1 = Sr-89 mol. wt.,

m_2 = Sr-90 mol. wt.,

A = Avogadro's number,

Σ_f = macroscopic fission cross section, 9262 cm²/TU,

x = tons uranium irradiated,

ϕ = neutron flux, n/cm²/sec,

ϕ' = apparent flux = ϕE ,

E = production efficiency = total equivalent irradiation time at equilibrium flux/total time in reactor,

τ = total equivalent irradiation time at equilibrium flux, seconds,

τ' = apparent irradiation time, τ/E , seconds,

y = Sr-88 fission yield fraction = 0.0335 $\pm$ 0.0015,

y_1 = Sr-89 fission yield fraction = 0.0445 $\pm$ 0.0015,

y_2 = Sr-90 fission yield fraction = 0.0545 $\pm$ 0.0015,

λ_1 = Sr-89 decay constant = 1.589 $\times 10^{-7}$ sec⁻¹,

λ_2 = Sr-90 decay constant = 7.845 $\times 10^{-10}$ sec⁻¹,

t = age since reactor discharge, seconds.

The fission yields for the strontium nuclides from Pu-239 and U-238 fission are only on the order of one-half the fission yield from U-235 so the amount of Pu-239 and U-238 fissioning has an appreciable effect on the composite or average fission yield. The fission yields and error limits listed above are based on calculated composite fission yields and are valid over the range from 200 MWD/T at 3 MW/T to 800 MWD/T at 8 MW/T.

Total MWD from Sr-88 Measurement

Measurement of the quantity of stable Sr-88 associated with a given quantity of strontium concentrate by mass spectrographic analysis and use of equation [7] leads to a determination of total megawatt days of exposure ($\pm \sim 5\%$) represented by a batch of recovered fission-product strontium. This is the same security problem as that associated with the cesium previously outlined in regard to equation [1]. Here, however, there is no possibility of masking the Sr-88 with natural strontium since the addition could be detected. Thus,

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the relationship of strontium recovered to total capability should remain classified in accordance with present practice. If recovery operations reach the point where essentially all of the strontium is recovered, it would no longer be possible to conceal plutonium production capability in terms of megawatt days.

Irradiation Time and Aging Time

In addition to total megawatt days of exposure, the only other determinations possible from strontium measurements are irradiation time and aging time. This can be shown most easily by writing the strontium relationships in terms of the three isotope weight ratios that can be measured:

$$\frac{W_{Sr89}}{W_{Sr88}} = \frac{m_1}{m} \frac{y_1}{y} \frac{1}{\lambda_1} \frac{(1-e^{-\lambda_1 \tau'})}{\tau'} (e^{-\lambda_1 t}) \quad [10]$$

$$\frac{W_{Sr90}}{W_{Sr88}} = \frac{m_2}{m} \frac{y_2}{y} \frac{1}{\lambda_2} \frac{(1-e^{-\lambda_2 \tau'})}{\tau'} (e^{-\lambda_2 t}) \quad [11]$$

$$\frac{W_{Sr89}}{W_{Sr90}} = \frac{m_1}{m_2} \frac{y_1}{y_2} \frac{\lambda_2}{\lambda_1} \frac{(1-e^{-\lambda_1 \tau'})}{(1-e^{-\lambda_2 \tau'})} \frac{(e^{-\lambda_1 t})}{(e^{-\lambda_2 t})} \quad [12]$$

Where all terms are as previously defined for equations [7], [8], and [9]. The only unknowns, and they occur in all three expressions, are τ' and t . Theoretically, it should be possible to solve for both τ' and t using any two of these three equations. The error limits have not been fully defined yet, but, based on the preliminary work, it is apparent that small errors in the basic data can result in gross errors in the solution for τ' and t . The basic difficulty is the same as that involved in equation [4]; that is, the small value of λ_2 and the insensitivity of the term $(1-e^{-\lambda_2 \tau'})/\tau'$ to small changes in τ' . The term $(1-e^{-\lambda_2 \tau'})/\tau'$ is inherent in the solution of simultaneous equations involving any two of the above three equations [10], [11], and [12].

There are three principal sources of error involved in the solution of simultaneous equations based on equations [10], [11], and [12], any one of which is sufficient to result in gross errors in the solution for τ' and t :

- (1) The uncertainty in the value of the fission yields -- approximately $\pm 5\%$;
- (2) The precision involved in establishing the ratio of Sr-89 to Sr-90 by radiochemical methods -- at least $\pm 1\%$ and perhaps as much as $\pm 20\%$ after 500 days aging⁽⁶⁾; and
- (3) The accumulation effect illustrated previously by equations [5] and [6] -- because of the large difference in half-lives of Sr-89 and Sr-90, this effect results in about a 5% distortion in the W_{Sr89}/W_{Sr90} ratio relative to average age for a 2.5 month accumulation period.

It is concluded that there is no need to treat recovered fission-product strontium as classified material except in the sense that total capability should not be disclosed.

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If data on age of fission-product strontium batches were disclosed, it may be possible to calculate apparent irradiation time, τ' , through use of equation [10]. Since the decay constant for Sr-89 is relatively large, a solution for τ' using equation [10] is not too sensitive to errors in the constants and the W_{Sr89}/W_{Sr88} ratio used. The inherent error in a solution for τ' using [10] is approximately $\pm 10\%$ assuming an error in the W_{Sr89}/W_{Sr88} ratio of $\pm 5\%$ and with full knowledge of the aging history. Of course, to obtain actual irradiation time, an estimate of production efficiency must be made but as mentioned previously this could most likely be guessed fairly closely. Since there is a possibility that significant information might be disclosed if age is disclosed, it is recommended that aging time be classified.

Without full knowledge of aging history, in particular the accumulation procedure, the probable error in a calculation for τ' is increased substantially. The effect of product accumulation can be illustrated by the following equations:

$$(W_{Sr89}/W_{Sr88})_1 = (W_{Sr89}/W_{Sr88})_0 \frac{(1 - e^{-\lambda_1 t_1})}{\lambda_1 t_1} \quad [13]$$

$$\text{and } (W_{Sr89}/W_{Sr88})_2 = (W_{Sr89}/W_{Sr88})_0 (e^{-\lambda_1 t_2}) \quad [14]$$

where a constant rate of accumulation is assumed and,

$(W_{Sr89}/W_{Sr88})_0$ = weight ratio of Sr-89 to Sr-88 in feed solution,

$(W_{Sr89}/W_{Sr88})_1$ = weight ratio at end of filling period,

$(W_{Sr89}/W_{Sr88})_2$ = weight ratio if average age assumed,

λ_1 = decay constant for Sr-89,

t_1 = total accumulation or filling time,

t_2 = average increase in age = $t_1/2$.

For a 2.5 month accumulation, the difference between $(W_{Sr89}/W_{Sr88})_1$ and $(W_{Sr89}/W_{Sr88})_2$ is about 5% which doesn't introduce a particularly large error in the solution for τ' . However, for someone outside Hanford who does not have knowledge of the accumulation procedure and does not know whether the accumulation period is one month, six months or a year, the uncertainty to him is probably more on the order of $\pm 50\%$. Thus, this becomes a very important factor and it is recommended that information relating to strontium accumulation procedures be classified as an additional safeguard.

In the discussion above, it was assumed that the W_{Sr89}/W_{Sr88} ratio would be measured within $\pm 5\%$. The assumed analytical procedure was: determination of the W_{Sr89}/W_{Sr90} ratio by radiochemical means and conversion to a W_{Sr89}/W_{Sr88} ratio by means of a mass spectrographic analysis for Sr-88 and Sr-90. This magnitude of precision is probable for strontium up to about 400 days old; at around 500 days old, the precision is estimated to be only on the order of $\pm 20\%$ (6). If this is true, then it may be possible to eliminate entirely the security problem associated with

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age by specifying no strontium shipments at less than 500 days old. The possible precision for these measurements will be further investigated before firm recommendations are made along this line.

Another aspect of this problem relates to possible quotations of calculated Sr-89/Sr-90 activity ratios in unclassified project proposals and specifications or to potential customers. If the age of the strontium is stated as well as the calculated activity ratio, and if it is obvious from the presentation that the quotation is a calculated value, it is a relatively simple matter for someone else to employ equation [12] and determine the irradiation time used in the original calculation. The only uncertainty would be in the y_1/y_2 ratio. The uncertainty in the y_1/y_2 ratio is only about $\pm 5\%$ which results in a $\pm 10\%$ uncertainty in the determination of irradiation time. Therefore, care should be exercised not to disclose the age associated with calculated Sr-89/Sr-90 activity ratios.

FIGURE 1

DECAY CHAINS FOR FISSION-PRODUCT CESIUM ISOTOPES⁽¹⁾

Notes: Underlined numbers give experimental U-235 fission yields;

() indicate probable nuclide occurrence in fission.

FIGURE 2

DECAY CHAINS FOR FISSION PRODUCT STRONTIUM ISOTOPES⁽¹⁾

Note: Underlined numbers give experimental U-235 fission yields.

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REFERENCES

- (1) Katcoff, Seymour. "Fission-Product Yields from Neutron-Induced Fission", Nucleonics 11: 201-210, November, 1960.
- (2) Bowers, C. E. Personal Communication, 1/26/61.
- (3) Walker, W. H. Yields and Effective Cross Sections of Fission Products and Pseudo-Fission-Products, CRRP-913. March, 1960.
- (4) Jaech, J. L. Personal Communication, 1/28/61.
- (5) Street, E. B. Personal Communication, 2/1/61.
- (6) Leaf, A. C. Personal Communication, 1/6/61 and 2/15/61.
- (7) Bupp, L. P. Monthly Report - October, 1960, Chemical Research and Development Operation, HW-67254-C. November 1960 (SECRET).
- (8) Friedlander, G., and J. W. Kennedy. Nuclear and Radiochemistry, 422. New York: John Wiley and Sons, 1955.

APPENDIXExcerpts from HW-37965, "Hanford Classification Guide"

<u>Section</u>	<u>Classification</u>
201.7.10 IRRADIATION (Natural Uranium)	
d. Irradiation time	S-RD
e. Integrated flux	S-RD
k. Exposure levels	S-RD
301.12.1 Plutonium and/or tritium production data, actual or forecast, expressed as megawatt days or equivalent weight or amount of product:	
a. For any one (except NPR) or all reactors for any period of time subsequent to June 30, 1958	S-RD
401.13.1 Any information which discloses irradiation history, for example:	
a. G/T or MWD/T levels	S-RD
d. Fission product compositions which disclose irradiation history	S-RD
401.14 <u>Liquid Waste Streams</u> - technical data revealing the following:	
401.14.1 Cumulation ^{ice} total amounts of specific fission products	S-RD
405.1.4 Exposure level of fission product waste material expressed in MWD/T or MW/T	S-RD
405.1.6 Amount of product recoverable (megacuries) per unit time	S-RD

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