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CRITICAL MASS STUDY OF 231 PROCESS TANKSABSTRACT

An estimated minimum critical mass for each of the process vessels in the 231 building has been calculated on the basis of critical mass data given in the P-11 project document HW-24514. The calculations are made assuming the plutonium to be a homogeneous mixture of precipitate and water with some slight neutron poisoning due to other elements. The precipitate is further assumed to have partially settled making an effectively infinite water reflector above the plutonium and hence reducing the critical mass.

A summary of the calculated minimum critical masses for plutonium of 400 MWD/T exposure is given in the following table:

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Process TankEstimated
Min. Critical Mass
for 400 MWD/T Plutonium*Distance to
bottom reflector*

PR (in water filled jacket)	850 grams	
✓ N-1 (floor)	1,100	4" none
✓ F-2	1,100	7" (no floor)
✓ NR-1, PR-1, PR-2, WR-2 (2x 2" angle iron only)	1,000	
✓ P-1, P-2, CT-2 (3" floor)	910	
✓ (4") (4") CT-1 (2" from floor)	1,250	
✓ N-2 (6" from floor)	860	
✓ WR-1 (2" from floor - 7" from bottom)	1,400	
✓ PR-3 16" from 2" iron	1,800	
S-1	1,100	
ST-2 (8" from floor)	900	
R-1, SR-1 OK	No limiting mass	
AT, WT, SC, Filter Boat	No limiting mass	

The minimum critical masses for 0, 200, 500, and 600 MWD/T are also calculated.

Because of the limiting critical mass of the N-2 filter, an increase of the present maximum batch size of 375 grams to 400 grams can be considered while still permitting the possibility of double batching.

INTRODUCTION

A study of the criticality hazards in each of the process tanks of the 231 building has been made to review the nuclear safety in this building. The minimum critical mass estimated for plutonium solutions in these tanks is based on an extrapolation of critical mass data found experimentally.

CALCULATIONS

In order to calculate the critical masses of the vessels, an empirical formula described in HW-24514 was used. This formula is based on the experimental fact that the critical thermal utilization (f) seems to vary linearly with changes in buckling and changes in hydrogen concentration (H), which gives a method of determining an empirical critical (f). The constants of such an equation will depend upon the cross sections used to calculate the critical thermal utilization and the extrapolation length used in calculating the buckling (B). Two such equations have been worked out using the same cross sections except for the Pu^{240} cross section and using different extrapolation lengths. The values used were: (1) $l = 4.0$ cm, $\text{Pu}^{240} = 1000$ barns and (2) $l = 6.0$ cm, $\text{Pu}^{240} = 925$ barns where for both cases the extrapolation length (l) is for an effectively infinite water reflector around thin walled stainless steel reactors. For case (1)

$$f = (38.897 - .2494 H) B + 2.743 (10)^{-3} H + .2221$$

and for case (2)

$$f = [54.68 - 36.24 H (10)^{-2}] B + 3.812 (10)^{-3} H + .08616$$

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These equations fit the experimental data within the experimental error and both extrapolate to the same value of the minimum homogeneous critical mass of plutonium in water. Figure I shows a plot of the minimum critical mass versus tamped SS sphere radius as calculated by the two methods.

Actual experiments were made for the critical mass in spheres of radius 15 cm to 19 cm. However, no experimental points could be made for the hydrogen concentration as high as water since the plutonium nitrate must be kept in at least one normal HNO_3 in order to stay in solution, and no experiments could be run without the presence of Pu^{240} . Thus Figure I is an extrapolation of the data and from the graph it is seen that over the range of spheres actually used the difference in the critical mass predicted by the two formulas is less than one per cent. However for radii less than 15 cm the difference becomes considerably larger.

In calculating the critical masses for the 231 process tanks the two formulas agreed in general to within three to five per cent. The lowest critical mass predicted by either formula was taken as the minimum critical mass. This for most of these cases is believed to be a conservative answer, so that the actual critical mass may be slightly larger than the results stated here.

Analysis of the possible concentrations in the process vessels show that in most cases it would be possible to have hydrogen concentrations very close to that of water. Thus, for these calculations the hydrogen concentration was taken to be 111.9 grams/liter. For all solutions there will be some other neutron absorbers such as nitrate associated with the Pu, SO_4 , La, and others. The critical mass will be raised slightly by these even in the solutions of nearly pure water, hence the total absorption cross section of these absorbers plus hydrogen was taken to be a minimum of $\Sigma_{\text{poison}} = 0.0220 \text{ cm}^{-1}$.

All of the calculations are based on the assumption that by some means either intentional or accidental the plutonium might start precipitating out of solution, effectively making an infinite water tamper above the plutonium, hence lowering the critical mass.

The extrapolation lengths used for the various thicknesses of stainless steel and water jackets were based on experiments made at Oak Ridge as reported in document K-643, where for a fuel of a given material buckling different amounts of lateral reflector thicknesses were used and the critical height (H_x), of the right cylinders, measured. Thus assuming a value of the extrapolation length with no reflector (l_0), l_x is found approximately by the formula

$$l_x = \frac{2.4048}{\sqrt{B_0 - B_{H_x}}} - R$$

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FIGURE 1

CRITICAL MASS VS TAMPED S.S. SPHERE RADIUS,
AT 0.0MWD/T, 0.0 NITRATE, 111.9 G/L HYDROGEN.

○ USING $l = 6$ CM

$\sigma_{\text{PU}240} = 925$ BARNES

△ USING $l = 4$ CM

$\sigma_{\text{PU}240} = 1000$ BARNES

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where R is the radius of the cylinder, B_0 is the buckling with no reflector and

$$B_{H_x} = \left(\frac{\pi}{H_x + l_0} \right)^2$$

Figure II is a plot of the results where $l_0 = 2.68$ cm. This gives for the effectively infinite water reflector $l = 6.1$ cm. The value of l_0 was picked arbitrarily in order to make the l_x at 0.05 inch of stainless steel (SS) be 2.84 cm, a value predicted by bare SS sphere data reported in HW-24514, using the formula made up with $l = 6.0$ cm for an infinite water reflector around a SS sphere.

If l_0 is taken to be ~ 0.65 as predicted by theory ($l_0 = 0.71 \lambda_t$), then the l_x for an infinite water reflector as calculated by the foregoing method becomes a little less than 4 cm. However, the formula based on $l_0 = 2.68$ cm predicts the lowest critical masses for cylinders and is the formula used in these calculations.

Using the value of 111.9 g/l for the hydrogen concentration, the critical thermal utilization becomes

$$f = 14.127 B + 0.51272$$

and the critical concentration is given by:

$$C = \frac{M}{V} = \frac{(10)^4}{28.97(1-w)} \left\{ \frac{\sum_{\text{poison}}}{1/f - \left[1 + \frac{w}{1-w} (0.801) \right]} \right\} \frac{\text{Grams}}{\text{Liter}}$$

where w is the fraction of Pu^{240} associated with the plutonium and for these calculations the approximate values used are:

<u>w %</u>	<u>$\sim \text{MWD/T}$</u>
0	0
1.46	200
2.95	400
3.65	500
4.40	600

The buckling for a sphere is simply

$$B = \left(\frac{\pi}{R'} \right)^2$$

where R' includes an extrapolation distance added to the radius. Hence using the critical mass formula the critical concentration for a given B corresponding to an R' can be calculated. Taking $\sum_{\text{poison}} = 0.0220$ then gives the following table:

FIGURE 2

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Critical Concentration g/l

R'	B	0 MWD/T	200 MWD/T	400 MWD/T	500 MWD/T	600 MWD/T
16 cm	.03855	∞	∞	∞	∞	∞
17	.03415	1563.0	∞	∞	∞	∞
18	.03046	125.7	158.7	216.9	262.1	337.3
19	.02734	67.56	76.65	88.86	96.05	105.2
20	.02467	47.13	51.63	57.20	60.26	63.92
21	.02238	36.79	39.61	42.97	44.76	46.84
22	.02039	30.52	32.53	34.86	36.08	37.48
23	.01866	26.36	27.90	29.67	30.58	31.62
24	.01713	23.37	24.61	26.03	26.75	27.57
25	.01579	21.15	22.19	23.37	23.97	24.65
26	.01460	19.43	20.33	21.35	21.86	22.44
27	.01354	18.06	18.86	19.75	20.20	20.71
28	.01259	16.95	17.67	18.47	18.87	19.32

For a cylinder the buckling is

$$B = \left(\frac{2.4048}{R + l_r} \right)^2 + \left(\frac{\pi}{H_c + l_t + l_b} \right)^2$$

where l_r = radial extrapolation length

l_t = top extrapolation length (used as 6.1 cm in these calculations)

l_b = bottom extrapolation length

H_c = critical height of the precipitate.

Then in a given process vessel the radius R and l_r , l_t , l_b are all known so the problem is to solve the critical mass equation for the height which gives the minimum critical mass. This can be done by equating the buckling of the cylinder to the buckling for which the critical concentration is known, solving the equation for the H_c , calculating the corresponding volume and hence the critical mass. This calculation is made for a series of heights until the minimum critical mass is found for the vessel.

RESULTS

Figure III shows the general layout of the tanks in a cell. The following are the critical masses calculated for each of these tanks.

PR (Process piece 224-W-310 Print D64263)

PR is transfer vessel in which the product is brought to the 231 building. It has an I.D. of 18-1/2 inch with 1/8 inch SS walls. The vessel is carried in a loose fitting jacket described in print D64264. Although highly improbable it is conceivable that the space of 5.1 cm between the vessel and the jacket could

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FIGURE 3
PROCESS TANKS IN 231 BUILDING

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become filled with water. Figure II gives $l = 5.75$ for this case.

For this and the following tables R^* is the radius of the critical sphere, H_c is the height of the cylinder of corresponding buckling and V_c is the volume of the precipitate solution at this H_c so that the critical mass is the product of V_c times the critical concentration of the equivalent buckling.

This gives for PR¹

R^*	H_c	V_c	Critical Mass for Various MWD/T				
			0	200	400	500	600
23 cm 18.1"	16.95 cm 6.67	29.39 l	775 g	820 g	872 g	898 g	929 g
24 18.9"	19.00 7.48	32.95	770	811	857	881	908
25 19.7"	21.21 8.35	36.78	778	816	859	882	906

N-1 (Process equipment piece 231-W-112 print D64458)

The solution is taken from the transfer vessel to the filter N-1. This piece has a 22 inch I.D., and 1/4 inch SS wall. It is 25 inches high and has a filter 3-3/4 inches from the bottom. Some plutonium may be caught in the filter but it is believed that the total critical mass will not be effectively decreased by a spot of increased concentration near the edge of the vessel. Thus this calculation is based on the possibility of a homogeneous precipitate above the filter so that the extrapolation length for the height is taken to be $l_H = l_T + l_b = 12.2$ cm and the radial l_R as given by the graph for 1/4 inch SS reflector is 3.33 cm.

This gives for N-1 ✓

R^*	H_c	V_c	Critical Mass for Various MWD/T				
			0	200	400	500	600
23 cm	15.62 cm	38.31 l	1,010 g	1,069 g	1,137 g	1,172 g	1,211 g
24	17.46	42.82	1,001	1,054	1,115	1,145	1,181
25	19.41	47.60	1,007	1,056	1,112	1,141	1,173
26	21.50	52.73	1,024	1,072	1,126	1,153	1,183

F-2

This tank has a backwash line and a leach line connected to N-1 filter. It has a 20 inch O.D. with 1/8 inch SS side walls, and is two feet six inches high with a 1/4 inch SS bottom. Thus $l_R = 3.03$ cm and $l_H = 9.43$ cm which gives for F-2

DECLASSIFIEDCritical Mass for Various MWD/T

R'	H _c	V _c	0	200	400	500	600
23 cm	20.29 cm	39.09 l	1,030 g	1,091 g	1,160 g	1,195 g	1,236 g
24	22.57	43.49	1,016	1,070	1,132	1,163	1,199
25	25.06	48.28	1,021	1,071	1,128	1,157	1,190
26	27.83	53.62	1,042	1,090	1,145	1,172	1,203

NR-1, PR-1, WR-2, PR-2 (Process equipment piece 231-W-140 Print D65228)

These vacuum transfer vessels have 17-1/2 inches I.D. with 1/4 inch SS walls and are 27 inches high. These vessels are held in the air and cannot easily be surrounded by further moderating material. Thus $l_R = 3.33$ cm and $l_H = 9.43$ cm, this gives for NR-1, PR-1, WR-2, PR-2

Critical Mass for Various MWD/T

R'	H _c	V _c	0	200	400	500	600
22 cm	19.81 cm	30.74 l	938 g	1,000 g	1,071 g	1,109 g	1,152 g
23	22.29	34.59	912	965	1,026	1,058	1,094
24	25.10	38.95	910	958	1,014	1,042	1,074
25	28.28	43.88	928	973	1,025	1,051	1,081

P-1, P-2, CT-2 (Process equipment piece 231-W-111 Print D64468)

These precipitator tanks have 17-3/4 inches I.D., the side walls are 1/8 inch SS surrounded by a 1/4 inch space that can be filled with water, which is enclosed by another 1/8 inch SS wall. The bottom is made of 3/8 inch SS beneath which is a one inch opening that can be filled with water and is enclosed by another 3/8 inch SS wall. These vessels are agitated, however settling is slow after the agitators are turned off so that only small heaping action of the precipitate is expected. Further, the tanks are small compared with the approximately 19 inch diameter predicted for the minimum critical mass of an infinitely water tamped hemisphere. Thus the homogeneous precipitate calculations seem acceptable in this case also.

For these calculations the absorption effect of a SS layer between a water reflector and the vessel is neglected giving in general a longer, thus more conservative extrapolation length. From the graph $l_R = 3.70$ cm, $l_b = 5.45$ cm so that $l_H = 11.55$ cm. This gives for P-1, P-2, CT-2

Critical Mass for Various MWD/T

R'	H _c	V _c	0	200	400	500	600
22 cm	17.14 cm	27.36 l	835.0 g	890 g	954 g	987 g	1,025 g
23	19.47	31.08	819	867	922	950	983
24	22.08	35.24	823	867	917	942	971
25	24.99	39.89	844	885	932	956	983

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CT-1 (Print H-2-202)

This tank receives the supernatant liquid drawn off from P-1 and usually has very low concentrations of product, however if an error were made it would be possible to draw a complete batch of plutonium into this tank. The tank has an I.D. of 23-5/8 inches, is 29-1/2 inches high and has a 3/16 inch SS wall and bottom which is surrounded by 1/4 inch opening that can be filled with water, enclosed by 1/8 inch SS outside wall. Thus $l_R = 4.75$ cm and $l_H = 10.85$ cm which gives for CT-1;

OK

R'	H_c	V_c	Critical Mass for Various MWD/T				
			0	200	400	500	600
24 cm	17.43 cm	49.28 l	1,152 g	1,213 g	1,283 g	1,318 g	1,359 g
25	19.10	54.00	1,142	1,198	1,262	1,294	1,331
26	20.87	59.01	1,146	1,200	1,260	1,290	1,324
27	22.74	64.29	1,161	1,212	1,270	1,299	1,331

N-2 (Equipment piece number 231-W-212 Print D64900) *(no filter in vessel)*

This filter tank has a 14 inch O.D. with 3/16 inch SS walls and is 20 inches high. There is a filter six inches from the bottom. As in N-1, there is a possibility of precipitation such that the plutonium has an effectively infinite water tamping above and below so that $l_H = 12.2$ cm and from Figure II for 3/16 inch SS reflector $l_R = 3.18$ cm. This gives for N-2:

OK

R'	H_c	V_c	Critical Mass for Various MWD/T				
			0	200	400	500	600
20 cm	17.91 cm	16.84 l	793 g	869 g	963 g	1,015 g	1,076 g
21	21.70	20.40	750	808	876	913	955
22	26.47	24.89	759	809	867	898	932
23	32.82	30.86	813	861	915	943	975

WR-1 (Print H-2-765)

WR-1 is a larger vacuum transfer vessel with a 24-1/2 inch O.D., 1/4 inch SS wall and is 25-1/2 inches high, hence $l_R = 3.33$ cm and $l_H = 9.43$ cm, thus:

R'	H_c	V_c	Critical Mass for Various MWD/T				
			0	200	400	500	600
24 cm	19.17 cm	55.95 l	1,307 g	1,377 g	1,456 g	1,496 g	1,542 g
25	20.90	61.00	1,290	1,353	1,425	1,462	1,503
26	22.74	66.37	1,289	1,349	1,417	1,450	1,489
27	24.68	72.03	1,301	1,358	1,422	1,455	1,492

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PR-3 (Print H-2-204)

PR-3 is a small vacuum transfer vessel with an I.D. of ten inches and a height of 16-1/2 inches. The walls are 1/8 inch SS so that $l_R = 3.03$ cm and $l_H = 9.13$ cm, thus:

			<u>Critical Mass for Various MWD/T</u>				
R'	H_c	V_c	0	200	400	500	600
18 cm	28.18 cm	14.28 l	1,795g	2,266g	3,097 g	3,743 g	4,817 g
19	40.73	20.64	1,394	1,582	1,834	1,982	2,170
20	78.02	39.53	1,863	2,041	2,261	2,382	2,527

(Vessels cannot be filled this high but this indicates where the minimum mass is found.)

S-1 (Equipment piece 116 Print D64497)

S-1 is a still with an inside dimension of: height 12 inches and diameter 11-3/4 inches. The side walls of the vessel are 1/8 inch SS and the bottom is constructed the same as P-1 where it is possible for a layer of water one inch thick to fill an opening between two enclosing layers of 3/8 inch SS, thus $l_R = 3.03$ cm and $l_H = 5.45$ 1.1 = 11.55 cm.

In this case however, the height of the hypothetical homogenous precipitate becomes large enough that there is no longer an effectively infinite water reflector above the precipitate since the tank is filled before the height for a minimum critical mass in this size cylinder can be reached. This necessitates a solution for the extrapolation length as the tank becomes filled.

Since $l_H = l_b + l_T$, in this calculation l_b is a known constant and the problem is to find l_T for a given value of $H' (= l_H + l_c)$ which is found when the cylindrical buckling is set equal to the buckling corresponding to a known critical concentration and the result solved for H' . Figure II shows that l is some function of the reflector thickness (rt) and since

$$H_{\max} - H_c = (rt)$$

where $H_{\max}$ is the height of the tank then H_c can be eliminated giving

$$l_T - (rt) = (H' - H_{\max}) - l_b$$

Figure IV shows a plot of l versus $l - rt$ so that as the critical height (H_c) approaches the top of the vessel values of l_T can be found as it varies from 6.1 to 2.68 cm.

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Thus for S-1:

			Critical Mass for Various MWD/T				
l_H	H_c	V_c	0	200	400	500	600
11.55 cm	20.85 cm	14.59 l	985 g	1,118 g	1,296 g	1,401 g	1,534 g
11.20	25.55	17.87	903	995	1,111	1,175	1,253
10.95	26.40	18.47	907	997	1,109	1,172	1,246
10.15	28.14	19.68	927	1,016	1,125	1,186	1,258

For the filled tank

8.48	30.48	21.32	978	1,069	1,181	1,243	1,316
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ST-2

This tank receives the overflow from a volume adjustment in PR-3. ST-2 has a 11-3/4 inch I.D. and is about 15 inches high. The walls and jacket are constructed the same as P-1 tank, hence $l_R = 3.70$ cm and $l_H = 11.55$ cm. Thus for ST-2:

			Critical Mass for Various MWD/T				
R^1	H_c	V_c	0	200	400	500	600
20 cm	23.58 cm	16.50 l	777 g	851 g	943 g	994 g	1,055 g
21	30.03	21.01	772	832	902	940	984
22	39.96	27.95	853	909	974	1,008	1,048

R-1, SR-1, AT, WT, S_c , and Filter Boat

The Limiting Volume and Cylinder Radius:

Using the critical mass equation it can be shown that for a large enough B the denominator becomes zero so that the critical mass becomes some very large amount. Any volume, or cylinder radius, smaller than this limiting size will give a larger buckling and thus will be safe from a thermal chain reaction for any concentration of plutonium. Imagining the most dangerous case of sphere filled with a homogeneous mixture of water and plutonium with no Pu^{240} (0 MWD/T) and the sphere having an infinite water reflector surrounding it, the limiting volume can be solved for since then $1/f = 1$ where $f = 14.127 B$ and .51272 and

$$B = \left(\frac{\pi}{R + 6.1} \right)^2$$

so $R = 10.82$ and

the limiting volume is then $V = \frac{4}{3} \pi r^3 = 5.3$ liters.

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Any volume smaller than this is said to be always safe from a thermal chain reaction.

To find the limiting cylinder radius the radial buckling for a cylinder surrounded by an infinite water reflector is set equal to the limiting B. So that in this case

$$f = 14.127 \left(\frac{2.4048}{R + 6.1} \right)^2 + .51272 = 1$$

Solving this gives $R = 6.85$ cm (5.4" diameter) and for cases when the cylinder does not have infinite water tamping the limiting radius is $R_{M \rightarrow \infty} = 12.95 - l_R$ cm. Any cylinder with a radius smaller than this is safe from a thermal chain reaction no matter to what height or concentration the solution in the cylinder may be.

R-1 (~~Print H-2-198~~)

new vessel 6" O.D. tubing

R-1 is the condensate receiver and usually never contains more than a few micrograms of product per liter received from the still. However, the critical mass in this tank can be noted. The tank has an eight inch O.D. with #16 gage SS walls and is 13-15/16 inches high. Thus $l_R = 2.85$ cm and the inside radius is 10.01 cm.

Since $R_{M \rightarrow \infty} = 12.95 - 2.85 = 10.10$ cm

then the radius of R-1 is smaller than the limiting radius so R-1 has no thermal critical mass limit.

SR-1 (Equipment piece 231-W-127

Print B64861)

new vessel 5 1/2" O.D.

This tank receives the heel from the still. It has a six inch O.D. with 1/8 inch SS walls and is 16 inches high. Since the inside radius is 7.30 cm and $l_R = 3.03$ cm then

$$R_{M \rightarrow \infty} = 12.95 - 3.03 = 9.92 > 7.30$$

hence this tank is always safe from a thermal chain reaction.

AT, WT, SC, Filter Boat

These vessels are the final stages of the 231 process and are of small volume. The adjustment tank, AT, has a one or two liter capacity, the WT weight tank has a volume of about 3-1/2 liters. The SC final loading vessel has a volume of 1.3 liters, and the filter boat which is the final loading for the precipitate form of plutonium, has a volume of less than three liters.

All of these volumes are less than the limiting volume of 5.3 liters so that these vessels by themselves may contain large amounts of product and be safe from a thermal chain reaction.

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It has been considered to load the SC cans with 350 grams of plutonium each, so it is of some interest to calculate how many such cans could be stored together safely. Assume that the outside carrying jackets for the SC cans could accidentally become mashed so that the SC cans are all very closely packed together and further that the 1/4 inch SS walls of the cans have a moderating effect similar to water and that this assembly can be approximated by a homogeneous mixture of solution and stainless steel in the shape of a large ball, then the critical mass formula can be used to calculate the final volume of such an assembly which will be critical.

This has been worked out using plutonium nitrate in 11 normal nitric acid. The formula predicts that a little more than 14 SC cans could cause such an assembly to go critical. This is of course, a very pessimistic estimate since it is highly improbable that the SC cans could ever be forced so closely together. However the result does indicate that the total number of SC cans stored in one place with this loading should be limited until multiplication experiments have been made on the number of cans that could be stored together.

SUMMARY

The minimum critical masses of plutonium in the process vessels of the 231 building are estimated to be for plutonium of 400 MWD/T exposure in the pile:

<u>231 Bldg. Process Tank</u>	<u>Estimated Min. Critical Mass for 400 MWD/T Plutonium</u>
PR (in water filled jacket)	850 grams
N-1	1,100
F-2	1,100
NR-1, PR-1, PR-2, WR-2	1,000
P-1, P-2, CT-2	910
CT-1	1,250
N-2	860
WR-1	1,400
PR-3	1,800
S-1	1,100
ST-2	900
R-1, SR-1	No limiting mass
AT, WT, SC, Filter Boat	No limiting mass

No considerations have been made in these calculations for neutron interaction between nearby vessels lowering the critical mass. However, as shown in an Oak Ridge critical mass study document K-406, for tanks of diameters greater than 15 inches there is very little interaction if the tanks are separated by 50 cm or more. There can be interaction even at this distance for tanks of smaller diameter. For these calculations the important exception is the N-2 Filter tank which has a diameter less than 15 inches, hence this tank could have some interaction with nearby tanks which would lower the minimum critical

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mass. However the tanks which are close to N-2 have large diameters so it is believed that this effect may not be large.

Because of the limiting critical mass of the N-2 filter, an increase of the present maximum batch size of 375 grams to 400 grams can be considered while still permitting the possibility of double batching.

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