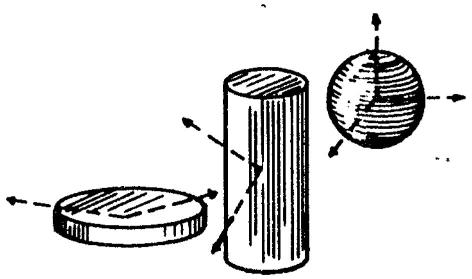

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CRITICALITY SAFETY IN THE STORAGE OF FISSILE MATERIAL

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PLUTONIUM SOLUTION STORAGE IN ARRAYS OF SIX-INCH PIPES

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The storage of Pu-239 in solution in an array of 6-inch pipes is very economical and convenient. It allows the material to be stored close to the work station. If the arrays contain more than six 6-inch pipes and have a concentration greater than 8 grams of Pu-239 per liter, an in-depth study should be conducted. Since most facilities are built with concrete walls, the safety margin of no reflection does not exist. At Los Alamos, a 8 x 5 x 1 array was calculated to be critical with concentrations between 30 and 40 grams of plutonium containing five percent Pu-240 per liter and a proposed 6 x 5 x 4 array at 60 grams per liter. A 5-inch pipe array modeled the same as the 6 x 5 x 4 array would be critical at 100 grams per liter. To prevent a problem with this storage, neutron interaction between tanks may be reduced by the use of boron loaded material which can be castable and non-combustible. If this isolation is not possible then very reliable administration controls must be implemented.

INTRODUCTION

A criticality safety study of plutonium nitrate storage in 6 inch diameter stainless steel tubing and piping in tank farms was initiated since Monte Carlo computer codes can now handle these large geometries at reasonable cost.

The study examined three sets of tank farms in the Plutonium Facility at the Los Alamos National Laboratory. The configurations were: a) a single unit; b) 6 x 5 x 4 array; and c) 8 x 5 x 1 array. The 6 x 5 x 4 array tube tank farm was chosen because it would contain the largest number of individual 6-inch tubes. The 8 x 5 x 1 array 6-inch pipe tank farm was selected due to its closeness to the concrete walls.

The range of concentrations of plutonium in nitrate solutions in processing facilities is very large. Most solutions for storage will have concentrations between 1 gram per liter and 20 grams per liter at Los Alamos. Discussions with personnel at Barnwell indicated that 500 grams per liter solutions was proposed to be pumped through pipes to flat storage tanks. Eldon Christensen at Los Alamos has made solutions containing 750 grams per liter under highly controlled conditions. In this study, concentrations were investigated in the range of 10 grams of plutonium per liter to 600 grams of plutonium per liter so the results could be applied to a variety of processing facilities.

Pipe and tube storage is a very economical and convenient method for plutonium nitrate solution storage. Stainless 304 steel is readily available in commercial tube and pipe and is easily welded which makes construction of arrays straightforward. By using commercial pipe and tube, physical characteristics have already been tested and documented and many suppliers have available a uniform product.

Six-inch diameter tube and pipe was used to minimize the cost of the materials and the amount of floor area per given volume of storage. As an example, the difference in volume between a 6-inch schedule 10 tube and a 5-inch schedule 10 tube is 46.7% with only a difference of 20.6% in the quantity of material.

Storage arrays were all modeled containing 6-inch schedule 10 pipe because the increase in volume in 6-inch pipe compared to 6-inch tube would give more conservative results. The characteristics of a single pipe were 16.15 cm inner diameter, .34 cm wall thickness, 304.8 cm (10 feet) length, and 62.41 liters in volume. The pipes in the arrays were on 45.72 cm (18 inch) centers.

In low molar HNO_3 solutions, the molar density of NO_3 density becomes significant since an increase in NO_3 causes a decrease in the hydrogen atom density and more neutrons are absorbed by the nitrogen atoms. Plutonium solutions can have a predominance of $Pu(NO_3)_4$ or $Pu(NO_3)_3$ depending on the length of time the solution has been mixed, solution temperature, solution temperature changes, acid concentration of the solution, and alpha activity of the plutonium. Since measuring and controlling ratios of $Pu(NO_3)_3$ and $Pu(NO_3)_4$ in a production facility would not be practical because of the above mentioned conditions, only solutions of $Pu(NO_3)_3$ were thoroughly analyzed so the results would always be conservative.

The Keno Monte Carlo code[1] used at Los Alamos was benchmarked on critical arrays of cylinders of a uranium solution with a concentration of 410 grams of 92.6% 235 enriched uranium assembled by Joe Thomas at Oak Ridge National Laboratory[2]. Code verification was done using Joe Thomas's uranium experiments because there have not been any experiments performed on arrays containing plutonium solutions. The Benchmark experiment of Joe Thomas consisted of uranium nitrate solutions in plexiglas cylinders in a 5 x 5 x 5 array. The major differencs of the experiments at Oak Ridge and the tank farms at PF-4 are: The experiments do not contain plutonium; the cylinders are made with plexiglas instead of stainless steel type 304 and the distance from the concrete walls is much less for the Los Alamos arrays. The Keno code calculated a k-eff of about .99 for the experimental 5 x 5 x 5 critical assembly which is about 1% less than Joe Thomas's calculations.

RESULTS

The single unit was studied to investigate how a single pipe would react without outside neutron drivers. Using the ONEDANT Code[3], a single pipe was reflected with 30 cm of water to simulate the most reactive condition. This reflection would also simulate floodings of the tank farm. The highest k-eff was 0.95 at 300 grams of plutonium per liter. Table 1 presents these results.

Table 1

6-Inch Pipe
95% Pu-239, 5% Pu-240, No HNO_3

Grams of Pu	k eff Bare	k eff Water Reflected	Kg of Pu
10	.353	.485	.62
20	.484	.660	1.24
30	.551	.753	1.87
40	.587	.803	2.50
50	.611	.849	3.12
100	.654	.909	6.24
150	.663	.920	9.36
200	.658	.931	12.48
250	.664	.951	15.60
300	.663	.954	18.72
350	.661	.949	21.84
400	.659	.948	24.96
500	.655	.947	31.20
600	.652	.946	37.44
700	.650	.945	43.68
750	.649	.943	46.80

The 6 x 5 x 4 tube schedule 10 array would be critical with a concentration of about 40 grams of plutonium per liter or about 300 kg of plutonium total. The density of this system equates to 0.10 kg of plutonium per cubic foot.

The 8 x 5 x 1 pipe array would be critical with a concentration of about 40 grams of plutonium per liter but with only a total mass of plutonium of about 100 kg. The mass density of the critical 8 x 5 x 1 array was 0.11 kg of plutonium per cubic foot.

K-eff is about 1.11% larger if $Pu(NO_3)_3$ is used instead of $Pu(NO_3)_4$ over the range of concentrations studied.

Water was modeled from 1 to 100% density between the pipes. At densities between 1 and 2% density, k-eff increased about 1%.

When the pipes were modeled with schedule 40 walls, k-eff decreased by about 6%. With schedule 80 walls, k-eff decreased by 11%.

A decrease in Pu-240 was investigated to see how significant small changes in isotopic composition of the plutonium would be. Using 3% Pu-240 gives an average increase of 2.8% in k-eff for the 6 x 5 x 4 tube array. Using 3% Pu-240 gives an average increase of 1.7% in the k-eff for 8 x 5 x 1 pipe array.

To simulate more realistic solutions, calculations were made with solutions containing plutonium nitrate and one molar and three molar nitric acid. The 1 molar nitric acid solution gives an average decrease of 0.95% in k-eff and the 3 molar solutions give an average decrease of 4.1% in k-eff over the range of concentrations studied.

SOLUTIONS

In all the above calculations a 6-inch schedule 10 pipe was used. If the 6-inch pipes are replaced by 6-inch tubes in the 6 x 5 x 4 array, the system will be critical at about 60 grams of plutonium per liter instead of 40 grams of plutonium per liter but the total amount of plutonium storage is decreased by 18.7%. If the 6-inch pipes are replaced by 5-inch schedule 10 pipe in the 6 x 5 x 4 array, the 5-inch pipe system will be critical at about 100 grams of plutonium per liter with a decrease in plutonium storage capability of 30.62%. The significance of these calculations is that the systems can become critical at concentrations found in processing facilities and the 6-inch pipe, 6-inch tube and the 5-inch pipe 6 x 5 x 4 and 8 x 5 x 1 arrays must have administrative concentration controls to ensure that they will remain subcritical.

To avoid the reliance on concentration control each pipe or tube can be neutronically isolated. Flexible material containing boron or cadmium as a wrapper for the pipes was studied. Table 2 gives the comparisons and the flexible material contents. Tubes wrapped with cadmium also gives a sufficient reduction of k-eff but the use of cadmium is not recommended due to the increase in gamma dose. Although these flexible materials give a sufficient reduction of k-eff and effectively isolate each pipe from low energy neutrons, the ability to remove the material from the tubes give a false sense of adequate safety.

Table 3 gives the results of a non-removable castable neutron moderator and poison being installed around the pipes. The 2% boron 1-inch thick material assures the system would be subcritical at any concentration of plutonium.

ADMINISTRATIVE PRACTICES

Administrative controls are practiced to ensure that concentrations of no more than 20 grams/liter are in a single tank at any time. Solution concentrations are verified through non-destructive assay (NDA) measurements. Each time solution is introduced into a tank, a total Pu-value is recorded so that at all times total Pu and total concentration is known.

Table 2

6 x 5 x 4 Array
60 Grams Plutonium/Liter
95% Pu-239, 5% Pu-240, 1 molar HNO_3
Concrete Walls as Exist
Water Equivalent Man on Open Side

Configuration	k-eff	Decrease in k-eff %
Tubes in column next to wall wrapped with 1/8" boron material	0.95	4.5
Outer tubes wrapped with 1/8" boron material	0.87	12.7
All tubes wrapped with 1/8" boron material	0.82	18.0
All tubes wrapped with 1/2" boron material	0.76	23.5
All tubes wrapped with 1/8" cadmium	0.86	13.8

Note: Flexible Boron Material

element	atom number density atoms/cc
boron	2.32×10^{22}
hydrogen	2.70×10^{22}
carbon	1.65×10^{22}
oxygen	1.49×10^{22}
silicon	9.47×10^{21}

Table 3

6-Inch Tube Farm with Neutron Poisons
6 x 5 x 4 Array
60 Grams Plutonium/Liter
95% Pu-239, 5% Pu-240, 1 molar HNO_3
Concrete Wall as Exist
Water Equivalent Man on Open Side

Concentration	1/2 Inch Thick 1% Boron	1 Inch Thick 1% Boron	1 Inch Thick 2% Boron	Mass
Grams of Pu/Liter	k-eff	k-eff	k-eff	kg
100	1.00	0.92	0.90	749
200	1.00	0.94	0.92	1498
300	1.04	0.96	0.93	2247
500	1.03	0.95	0.93	3745

Tanks are visually inspected and NDA measurements are performed to indicate any hold-up of Pu in a tank. A hold-up is usually due to post-precipitation of plutonium oxalate. When this occurs, a HNO_3/HF wash will dissolve the oxalate, cleaning the tank.

Los Alamos is currently replacing the horizontal 6 inch diameter tanks with vertical tanks to alleviate the post-precipitation build-up problem.

CONCLUSION

Once it is decided to store or transport plutonium and uranium nitrate in pipes or tubes, extensive analyses are necessary since reflection of concrete walls and interaction between the pipes can lead to a system being critical at modest concentrations. The 8 x 5 x 1 array of 6-inch pipes as built would be critical with concentrations between 30 and 40 grams of plutonium per liter. The 6 x 5 x 4 array of 6-inch tubes if built, would be critical at about 60 grams of plutonium per liter. If 5-inch pipe was used in the 6 x 5 x 4 array, the system would be critical at a concentration of about 100 grams of plutonium per liter.

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