EVALUATION OF THE CURRENT CAPACITY OF THE LOS ALAMOS NATIONAL LABORATORY'S TRU RADIOLOGICAL LIQUID WASTE TREATMENT FACILITY AND RECOMMENDATIONS FOR ITS FUTURE DESIGN

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ABSTRACT

The Radioactive Liquid Waste Treatment Facility (RLTWF) of Los Alamos National Laboratory (LANL) is critical to the lab's nuclear program, as most of the lab's radioactive liquid wastes are treated in this facility. In addition to treating a large amount of industrial wastes that are low in radioactivity, the RLWTF treats transuranic (TRU) waste that come directly from LANL's plutonium processing facility. TRU waste has radioactivity greater than 100 nanocuries per gram of alpha particle emitting nuclides with atomic number greater than 92 and with half-lives greater than 24,000 years. The treatment of the transuranic waste occurs in Room-60 of the RLWTF.

Appropriate mathematical equations were developed and solved numerically to predict the temperature rise of the neutralization process in Room-60 and the cooling rate of the neutralization tank under different cooling scenarios. These results were analyzed to evaluate the current Room-60 design and to determine whether the current RLWTF is capable of handling the maximum rate of TRU waste that could come from the lab's plutonium processing facility.

It has been found that the current RLWTF is well capable of handling the maximum rate of TRU waste as long as the nitric acid waste is below 4 normal. If the acid waste was to exceed 4 N, the current RLWTF would not be capable of handling the maximum rate (4,500 liters a month). However, if the neutralization tank in Room-60 was to be cooled by cooling water, reducing the neutralization cooling time to be less than 24 hours, the RLWTF should be capable of handling the maximum rate even with acid with high normality as 8.

In addition to these findings, a new Room-60 design proposed by the RLWTF staff utilizing ultrafiltration has been re-evaluated. It was discovered that unless the neutralization tank is equipped with a cooling system, the new facility would not be capable of handling the maximum discharge from the plutonium processing facility. Additionally, the model predicts that the neutralization tank, the phase separator tank, and the ultrafilter feed tank, could be smaller (2,050 liters) in size than what had been proposed earlier (7,400 liters).

INTRODUCTION

The Radioactive Liquid Waste Treatment Facility (RLWTF) of Los Alamos National Laboratory treats transuranic (TRU) waste from the laboratory's plutonium processing facility and other industrial radioactive liquid waste from various parts of the lab. The TRU waste is processed in Room-60 of the RLWTF and is composed of two waste streams: acid and caustic. The industrial radioactive liquid waste is treated in the main treatment facility.

The operation of the RLWTF is critical to the laboratory and its nuclear program because the majority of the radioactive liquid waste generated from the lab is treated in this facility.

Objectives

The objectives of this study are two fold:

1. To evaluate the current Room-60 design and determine whether it is capable of handling maximum amount of TRU waste from TA-55, the lab's plutonium processing facility.
2. To evaluate the feasibility of the Room-60 design that has been proposed in the upgrade project and make any recommendation, if possible, of any changes.

Analyzing Room-60 Design and its Process

The current Room-60 process was modeled with EXTEND™. A general flowchart of the process is shown in Fig. 1. As shown in the flowchart, acid and caustic wastes from TA-55 are stored in separate tanks in WM-66 before being processed in Room-60. Once the tank becomes somewhat full, these wastes are neutralized in TK-1. Afterwards, they are treated in the Clarifier and/or TK-7 where the precipitated materials are later tumbled into 55-gallon solid waste drums. The effluent is sent for further purification.

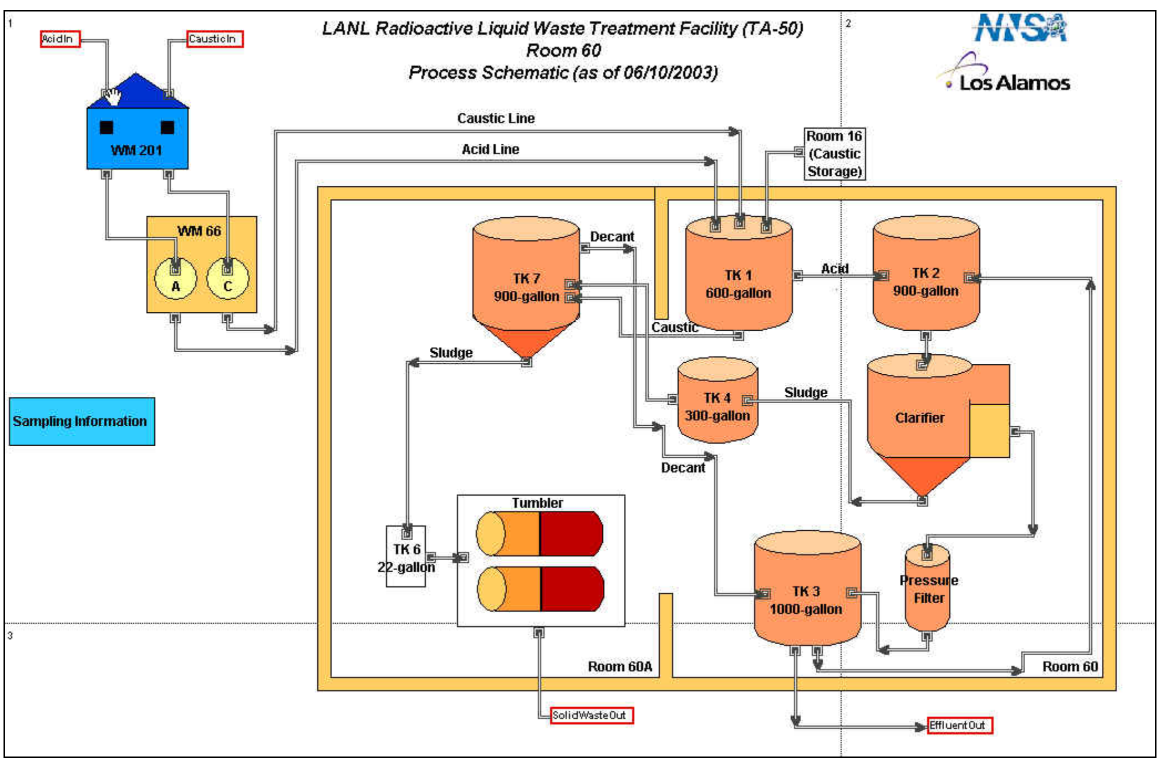

Fig. 1. The process schematic of Room-60 constructed in EXTEND™

When the acid or the caustic waste from TA-55 is neutralized in TK-1, heat is generated. Depending on the normality of the acid or the caustic waste, the generated heat could raise the temperature of the solution up to 85-90 ºC. TK-1 is then cooled for several days before being further processed to TK-7 or the Clarifier.

Since it takes several days for TK-1 to cool and other Room-60 processes take less than a day to complete, this neutralization process becomes the rate limiting step of the whole Room-60 process.

Analytical Considerations

A model was constructed to predict the final temperature of the TK-1 content after the neutralization process and also to predict the cooling-rate of TK-1 under different conditions. This is performed to determine if the current Room-60 is capable of handling the maximum amount of TRU waste from TA-55.

The caustic wastes rarely need neutralization and can be processed straight to TK-7. The heat generated from the neutralization process mainly comes from the following reaction:

$$HNO_{3\,(aq)} + NaOH_{\,(aq)} \rightarrow H_2O_{\,(l)} + NaNO_{3\,(aq)}$$

The heat generated from this reaction is given by the heat of the reaction, which is approximately -13.8 kcal per mole of reacted nitric acid. From this heat of the reaction, the final temperature of the neutralization content is obtained by a simple energy balance.

Afterwards, a transient energy balance was used to predict the rate at which TK-1 cools. The energy balance is shown in equation 1.

$$k\frac{T_{content} - T_{steel}}{t_{steel}}A = \rho c_p V \frac{dT_{content}}{dt} \qquad \text{(Eq. 1)}$$

where

- k = thermal conductivity of steel ($W/m^2\ K$)
- $T_{content}$ = temperature of the tank content (K)
- T_{steel} = outside temperature of the tank (K)
- t_{steel} = thickness of the steel tank (m)
- A = surface area of the tank (m^2)
- ρ = density of the tank content (kg/m^3)
- c_p = heat capacity of the tank content (J/kg K)
- V = volume of the tank content (m^3)
- t = time (s)

Additional energy balance between the tank wall (which looses heat through conduction) and the outside (which involves convection and radiation) gives equation 2.

$$k\frac{T_{content}-T_{steel}}{t_{steel}}A = h_{side}A_{side}(T_{side}-T_{room}) + h_{top}A_{top}(T_{top}-T_{room}) + h_{bottom}A_{bottom}(T_{bottom}-T_{room})$$
$$+\varepsilon\sigma(T_{steel}{}^{4}-T_{surr}{}^{4}) + \rho_{steel}c_{p\_steel}V_{steel}dt \qquad \text{(Eq. 2)}$$

where

A_{side} = surface area of the side of the tank (m^2)
A_{top} = surface area of the top of the tank (m^2)
A_{bottom} = surface area of the bottom of the tank (m^2)
h_{side} = convection coefficient for the side (W/m^2 K)
h_{top} = convection coefficient for the top (W/m^2 K)
h_{bottom} = convection coefficient for the bottom (W/m^2 K)
T_{side} = temperature of the side of the tank (K)
T_{top} = temperature of the top of the tank (K)
T_{bottom} = temperature of the bottom of the tank (K)
ε = emissivity of the tank
σ = radiation coefficient (W/m^2 K^4)
ρ_{steel} = steel density (kg/m^3)
c_{p_steel} = heat capacity of steel (J/kg K)
V_{steel} = volume of steel in the tank (m^3)

For the cooling case scenario, the second energy balance yields equation 3.

$$k\frac{T_{content}-T_{steel}}{t_{steel}}A = F_{cooling}\rho_{water}c_{p\_water}(T_{out}-T_{in}) + h_{top}A_{top}(T_{top}-T_{room}) + h_{bottom}A_{bottom}(T_{bottom}-T_{room})$$
$$+\varepsilon\sigma(T_{steel}{}^{4}-T_{surr}{}^{4}) + \rho_{steel}c_{p\_steel}V_{steel}dt \qquad \text{(Eq. 3)}$$

where

$F_{cooling}$ = flowrate of water in the cooling coil (m^3/s)
ρ_{water} = density of water (kg/m^3)
c_{p_water} = heat capacity of water (J/kg K)
T_{out} = outlet temperature of the cooling water (K)
T_{in} = inlet temperature of the cooling water (K)

These two equations were solved simultaneously to obtain the temperature of the steel and the tank content temperature. The heat convection coefficients for the different scenarios were obtained using empirical heat-transfer correlations demonstrated next.

Heat Transfer Correlations for Natural Convection

The convection coefficient for the top side of the tank was obtained by the following standard equations [1].

$$NU_L = 0.54Ra^{1/4} \quad (10^4 \leq Ra_L \leq 10^7)$$
$$NU_L = 0.15Ra^{1/3} \quad (10^7 \leq Ra_L \leq 10^{11})$$

where

$$NU_L = \frac{hL}{k} \quad \text{(Eq. 4)}, \quad Ra = \frac{g\beta(T_s - T_\infty)L^3}{\alpha\nu} \quad \text{(Eq. 5)}$$

where

h = convection coefficient (W/m^2 K)
L = characteristic length (area/parameter)
k = thermal conductivity of air at the film temperature (W/m K)
g = gravitational constant (m/s^2)
β = gas expansion coefficient (1/K)
T_s = surface temperature (K)
T_∞ = the room temperature (K)
α = thermal diffusion coefficient (m^2/s)
ν = kinematics viscosity (m^2/s)
Nu = Nusselt number (dimensionless)
Ra = Rayleigh number (dimensionless)

The convection coefficients for the lower surface plate of the tank and the side were obtained by using the following standard equations [2].

$$NU_{L\_lower} = 0.27Ra^{1/4} \quad (10^5 \leq Ra_L \leq 10^{10})$$

$$NU_{L\_side} = \left\{ 0.825 + \frac{0.387 Ra_L^{1/6}}{\left[1 + (0.492/\Pr)^{9/16}\right]^{8/27}} \right\}^2$$

Heat Transfer Correlations for Forced Convection

It was assumed that the forced convection would only occur on the side of the tank. For the bottom and the top side of the tank, natural convection was assumed to occur.

For the cylindrical side of the tank, the convection coefficient was obtained by

$$NU_D = C \operatorname{Re}_D{}^m \Pr^{1/3} \quad \text{(Eq.6)}$$

where

Pr = Prandtl number of air at the film temperature (dimensionless)

$$NU_D = \frac{hD}{k} \quad \text{(Eq. 7)}$$

where

D = diameter of the tank (m)

The unitless coefficients C and *m* for the above relation is shown in the Table 1 below.

Table I. Constants of Equation for the Circular Cylinder in Cross-Flow [2]

Re_D	C	*m*
0.4-4	0.989	0.330
4-40	0.911	0.385
40-4,000	0.683	0.466
4,000-40,000	0.193	0.618
40,000-400,000	0.027	0.805

Heat Transfer Correlations for the Cooling Process

Similar to the forced convection scenario, the bottom and the top of the tank were assumed to be cooled by natural convection. It was assumed that the cooling coils were wrapped around the side of the tank.

The outlet temperature of the coolant in the cooling coil was estimated by

$$T_{outlet} = eff \cdot (T_{steel} - T_{in}) + T_{in} \qquad \text{(Eq. 8)}$$

where

T_{outlet} = outlet temperature of the coolant (K)
eff = efficiency of the cooling process (ranges from 0 to 1.0)
T_{steel} = tank steel temperature (K)
T_{in} = inlet temperature of the coolant (K)

Estimation of the Current Room-60 Capacity

The maximum amount of caustic waste that TA-55 can send to the RLWTF is approximately 30,000 liters each year. TK-1 can process approximately 2,300 liters of caustic per time. If one batch of caustic waste is processed per day, 30,000 liters of caustic can be treated in less than three business weeks. Therefore the current Room-60 is capable of handling the maximum amount of caustic waste that could be sent from TA-55.

For the acid waste, the processing time of each acid batch depends on the acid normality since the normality affects the final temperature after the neutralization process, thus the final cooling time. To make a general prediction of the current Room-60 acid treatment capacity, the following assumptions were made:

- The tank, made of 316 Stainless Steel, was assumed to be at a room temperature of 21 ºC initially.

- The acid wastes from WM-66 were assumed to be approximately 27 ºC before entering TK-1
- The tank was assumed to have a radiation emissivity of 0.8 (it turns out that radiation effect is very small anyway).
- The tank was filled up to its 80% full capacity.
- The tank content was assumed to be well mixed. Any heat transfer due to convection around the tank was assumed to cool down the tank content instantly.
- The neutralization content was assumed to have the density and heat capacity of water.
- The tank thickness was assumed to be 3/16 inch
- The TK-1 content was treated when it reached about 40 ºC

The operation factors were taken into consideration as well. Since it is impossible to process the TK-1 content if the cooling finishes in the middle of the night when the facility is empty. Table 2 was used to estimate the number of batches that could be processed each week.

Table II. Predicted Number of TK-1 Operations with Varying Cooling Time

	Cooling time for each TK-1 acid batch				
	0-5 hrs	5-24 hrs	24-48 hrs	48-78 hrs	78-148 hrs
No. of Operations (batches/week)	10	5	3	2	1

For example, if the cooling time of TK-1 batch was under 5 hours, at least two acid batches could be processed in one day. Since there are five operating days each week, this would correspond to at least 10 acid batches a week. On the other hand, if the cooling time was between 5-24 hours, it is most likely that only one acid batch could be processed a day. This would correspond to 5 acid batches a week.

Based on the above assumptions and the operating condition, the Room-60 capacity for the acid treatment was obtained (see Fig. 2).

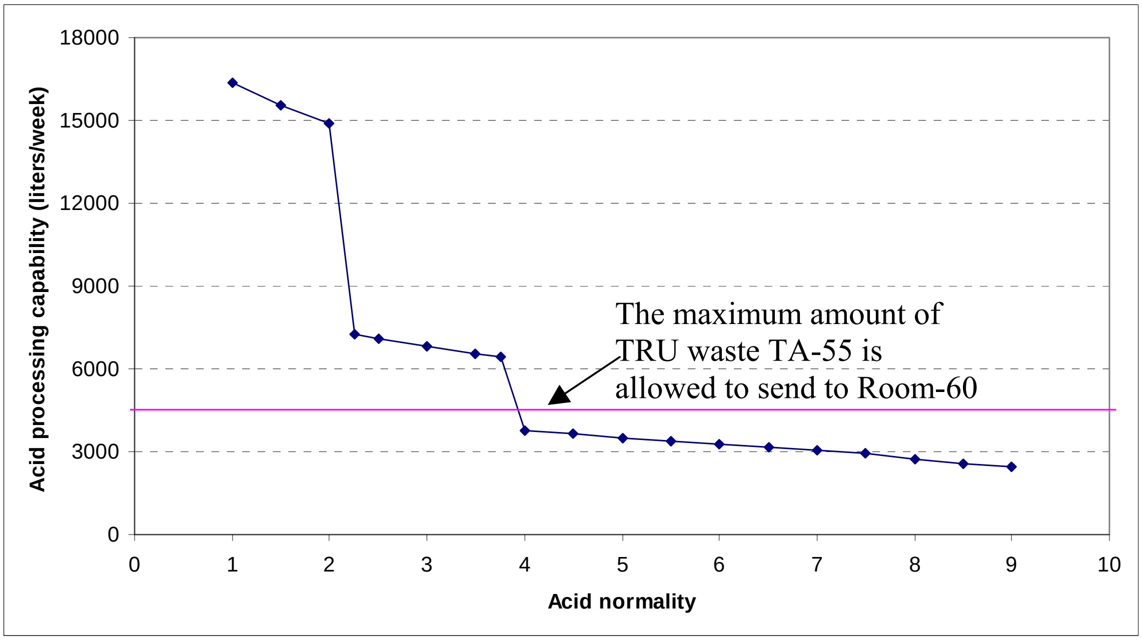

Fig. 2. Current room-60 capacity for the acid treatment with varying incoming acid normality

The maximum amount of acid waste that TA-55 can send to the RLWTF is about 4,500 liters a week (indicated by the solid line on Fig. 2). Although it is not very likely for TA-55 to send 4,500 liters of acid waste every week (approximately 230,000 liters a year), the RLWTF should be capable of handling this amount of acid waste regardless. The blue line on the Figure shows the amount of acid waste that the present Room-60 process is capable of treating for different acid normality.

The Room-60 is predicted to handle the maximum amount of acid waste (4,500 liters) if the acid normality is below 4.0. For example, if the incoming acid waste has a normality of 2.0, Room-60 is expected to be capable of treating up to 15,000 liters of acid waste. However, once the acid normality exceeds 4.0, the facility is not expected to be able to treat 4,500 liters of acid waste each week. As the acid normality increases, the acid processing capability of Room-60 also decreases. For an instance, as the acid normality increases from 4.0 to 6.0, the acid processing capability of Room-60 drops from 3,770 liters to 3,270 liters per week.

The two large decreases in the acid processing capability as the acid normality increases indicate the change in the number of acid batches that are processed each week. For acid waste 2.0N or below, the cooling of the neutralization tank takes less than 5 hours, which constitute an operation of 2 batches a day or 10 batches a week. However for acid waste roughly between 2.2N and 3.8N, the cooling time of the neutralization tank takes between 5-24 hours, which would require an operation of 1 batch a day or 5 batches a week. As the acid exceeds 4N, the cooling takes between 24-48 hours. This results in an operation of only 1 batch every two days or 3 batches a week.

In the past, the average acid normality has been around 2-3 and the RLWTF has been fully capable of handling these wastes as the model correctly predicts. However, with new programs in the laboratory, the acid normality from TA-55 is expected to go up as high as to 6 or 7. If this is the case, the RLWTF would not be capable of treating 4,500 liters of acid waste a week.

What can be concluded from this analysis is that the current Room-60 process would not be capable of handling the maximum amount of TRU waste if the acid waste exceeds 4N. This poses a serious problem since the RLWTF is a critical part to the lab's nuclear program and especially to the operation of TA-55, the lab's plutonium processing facility. If TA-55 starts to discharge its maximum amount of TRU waste every week and if its acid waste exceeds 4N, there is no way that the current RLWTF can process all of its waste.

The critical problem behind this is that it takes too much time to cool TK-1, especially when the acid starts to exceed 4N. Notice that as soon as TK-1 takes more than 24 hours to be cooled, the RLWTF becomes incapable of handling 4,500 liters of acid waste.

Other Ways of Cooling TK-1

There are several other ways of cooling TK-1 than just by natural convection. Three other possibilities were considered in this section. The first was having a fan next to TK-1 that would blow air at a speed of 3 m/s. The air was assumed to be at room temperature. The second scenario was having cooling water around TK-1. The efficiency of the cooling system was assumed to be 0.6 and the flow-rate of the cooling water was assumed to be 2.5 gpm. The third scenario was using process water in TK-9, another tank in the RLWTF facility, to cool TK-1. TK-9 was assumed to be adiabatic, gaining heat only from that absorbed from TK-1.

TK-1 is expected to cool rapidly in the beginning due to the large temperature difference between the tank and the cooling medium. As the temperature difference between the tank and the cooling medium decreases, the rate at which the cooling occurs decreases as well, indicated by the leveling of TK-1 temperature. For example, 50% of the total cooling (TK-1 changing from 90 ºC to 55 ºC) occurs during the first fours hours when cooling water is used. It takes additional 20 hours or more for TK-1 to cool down the other 50% (TK-1 changing from 55 ºC to 20 ºC).

As expected, natural convection is the slowest. Even after 72 hours, TK-1 is predicted to be 37 ºC. Forcing air around the tank causes the tank to be cooled a little faster, cooling TK-1 to 25 ºC in approximately 72 hours. Using TK-9 or cooling water brings TK-1 to below 30 ºC in less than 13 hours.

Figure 3 shows the total amount of time TK-1 would need to cool down from 90 ºC to 40 ºC and from 90 ºC to 27 ºC. For the proposed Room-60 design, an ultra-filtration system was recommended after the neutralization process. The current Room-60 process requires the neutralization content to be approximately 40 ºC in order to process to the next unit. An ultra-filtration system would require the content to cool down to approximately 27 ºC. Therefore two different times are shown in the figure to indicate both possibilities.

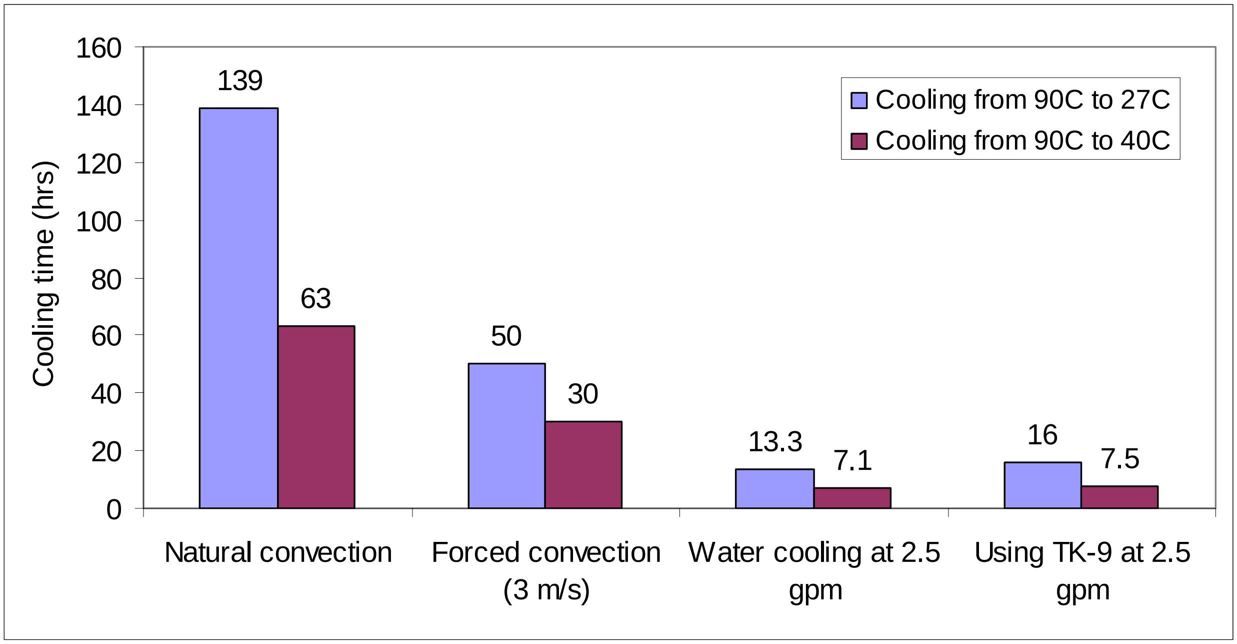

Fig. 3. A summary of cooling time of TK-1 with other cooling-methods

If a fan was to be installed next to TK-1, blowing air on the side of TK-1 at a speed of 3 m/s, and if the tank was initially at 90 ºC, it would take approximately 30 hours for the tank to cool down to 40 ºC. This is a significant decrease in the total cooling time compared to the 63 hours that it would take just by natural convection.

As mentioned before, for the current RLWTF to be able to process the maximum rate of TRU waste from TA-55, the total cooling time for the TK-1 neutralization tank would have to be less than 24 hours. Therefore, cooling TK-1 via forced convection would not satisfy the requirement.

A possibility of cooling TK-1 by water was considered next. The inlet temperature of the cooling water was assumed to be 20 C. To cool from 90 ºC to 40 ºC, TK-1 is predicted to take about 13.3 hours. For it to cool from 90 ºC to 27 ºC, it is expected to take approximately 7 hours.

Therefore, if one needs to cool TK-1 in a few hours instead of a few days, it would be a wise option to use cooling water. A heat exchanger could be installed to cool the heated water and the water could be recycled back. This method would be relatively easy to install and easy to maintain. In case a heat exchanger is to be used, the cooling fluid could be cooled to a temperature far below 20 ºC which would even reduce the cooling time further than shown in the Figure.

Another alternative of cooling TK-1, other than using water at constant temperature, is to use the contents in another tank as a source of cooling water. The possibility of using TK-9 contents to cool TK-1 and recycling the cooling outlet back to TK-9 has been mentioned.

TK-9 was assumed to be adiabatic, gaining heat only from the TK-1 cooling process. In addition, it was assumed that the heat convection on the side of the TK-1 was negligible. The top and the bottom of the TK-1 were still assumed to dissipate heat through natural convection. The cooling time does not change significantly when TK-9 is used (as compared to using water at constant inlet temperature of 20 ºC). The model predicts that it would take 16 hours for the tank to cool from 90 ºC to 40 ºC. Using cooling water would take about 13 hours. The difference is even smaller if the tank were to be cooled down to 27 ºC. By then, there is virtually no difference in time between cooling TK-1 by cooling water or TK-9.

Again, the RLWTF is only capable of handling the maximum rate of TRU waste from TA-55 if the TK-1 neutralization tank can be cooled in less than 24 hours. Since both the second and the third method cools TK-1 in less than 24 hours, it is recommended that either the second or the third method be used to cool TK-1 rather than by natural convection.

Assuming that TK-1 is to be cooled via cooling water or via TK-9 (still cooling the solution to at least 40 ºC), the Room-60 acid processing capability is expected to be as shown in Figure 4. Whereas the current facility is only capable of treating the maximum rate of TRU waste if the acid was below 4 N, an addition of a cooling system would allow the current RLWTF to process the maximum rate of TRU waste for acid normality up to 8. This is a large increase for the RLWTF as the incoming acid waste from TA-55 has never exceeded 8 N. Even with the recent MOX program at the lab, the acid waste has been usually around 6 N.

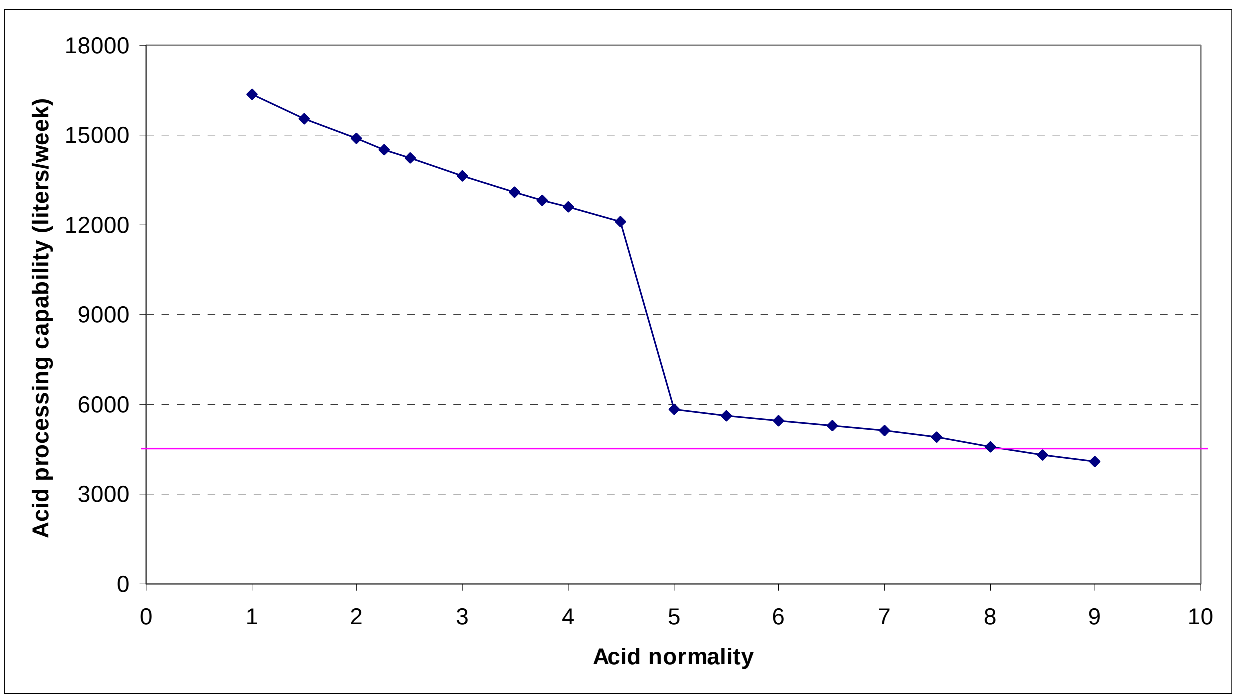

Fig. 4. Projected room-60 capacity with the current design if TK-1 was to be cooled via cooling water at 2.5 gpm (compare with Fig. 2); the solid line indicates the maximum rate of TRU waste that could come from the plutonium processing facility at the lab

Even if the acid waste was to be higher than 8N, the RLWTF would be capable of handling most of the waste from TA-55. This is because the RLWTF rarely gets more than 3,000 liters of acid waste a month from TA-55. Even if one month, the plutonium facility disposes 4,500 liters of acid waste (the maximum rate), it is very unlikely that it will dispose another 4,500 liters of acid waste the following month. Since the WM-66 acid tank can hold up to 15,000 liters of acid waste, the amount of acid that could not be processed could be stored in the WM-66 tank until the following month to be processed.

Recommendations for the New Room-60 Design

In the RLTWF upgrade project (separate from this report), a new Room-60 design has been proposed. This new design was modeled in EXTEND^{TM}.

The new process is fairly simple. After the neutralization takes place, TK-1 content is dumped into a phase separator where it is left for approximately a day. Afterwards, an ultrafilter is used. The generated sludge is sent to the sludge tank and the effluent is sent to the main treatment facility to be further purified.

The TK-1, the phase separator, and the ultrafilter feed tank are specified to be 7,400 liters in size. The sludge tank is designed to hold up to 3,700 liters. In addition, TK-1 is still designed to be cooled just by natural convection. No other cooling system for TK-1 has been considered in the proposed design.

Several problems with the current design exist. First, for acid waste with normality as high as 6 or 7, the neutralization process would cause an enormous amount of heat. Since TK-1 was sized to hold as much as 7,400 liters of waste, it would take several days, if not more than a week, to be cooled. Even the current Room-60 process requires two to three days for TK-1 to cool down to 40 ºC (the model predicts that it would take about 63 hours, see Figure 3). However the new proposed design has an ultrafilter system, which would require its influents to be a lot cooler, even close to the room temperature. Even for a tank just 2,300 liters large, it would take approximately 139 hours or roughly six days for its contents to be cooled down to 27 ºC (see Figure 3). With a tank as large as 7,400 liters, it would just take too much time to cool. Since all the other process in Room-60 depends on the neutralization process and the rate at which it cools, this would be too inefficient.

The second problem with the proposed design is that the tanks are unnecessarily large. Tanks with smaller size could perform the same job as well.

Therefore, rather than continuing to cool TK-1 with natural convection, it is recommended that TK-1 be cooled with cooling water or any other method (as long as TK-1 can be cooled down in less than 24 hours). In addition, it is not necessary for TK-1, the phase separator, and the ultrafilter feed tank to be as large as 7,400 liters (as proposed in the upgrade project). It is found that they only have to be about 2,050 liters large to accommodate all the processing needs.

Assuming that at least one batch of TK-1 can be processed a day, this would allow Room-60 to be capable of handling the maximum incoming rate of TRU waste even if acid normality is as high as 7.

Also to allow the WM-66 tanks to store up to 2 weeks worth of both caustic and acid waste (assuming that TA-55 sends its maximum weekly discharge), it is recommended that WM-66 acid and caustic tank be at least 11,400 liters and 3,800 liters large.

CONCLUSIONS

From this study, it has been demonstrated that the current RLWTF is only capable of treating the maximum rate of TRU waste from the lab's plutonium processing facility if the acid waste is below 4 normal. To enable the current RLWTF to handle acid waste with higher normality, it is strongly recommended that a cooling system be installed for the TK-1 neutralization tank. A simple heat exchanger could cool the neutralization tank in just a few hours, solving this problem.

In addition, the neutralization tank in the future Room-60 design should also be equipped with a cooling system. Without it, Room-60 would not be able to process all the acid waste that could come from the plutonium processing facility. Furthermore, the neutralization tank, the phase separator tank, and the ultrafilter feed tank, could be smaller than what have been proposed in the design and still be capable of handling the maximum rate. The original design planned the three tanks to be capable of holding up to 7,400 liters of waste. However, they need only be 2,050 liters in size.

REFERENCES

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