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Title: **MINIMIZING WASTE & COST IN DISPOSITION OF LEGACY RESIDUES**

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MINIMIZING WASTE & COST IN DISPOSITION OF LEGACY RESIDUES

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1.0 INTRODUCTION

Research is being conducted at the Los Alamos National Laboratory (LANL) which is directed toward development of a quantitative basis for disposition of actinide-bearing process residues (both legacy residues and residues generated from ongoing programmatic operations). This research is focused in two directions: (1) identifying minimum negative consequence (waste, dose, cost) dispositions working within regulatory safeguards termination criteria, and (2) evaluating logistics/consequences of across-the-board residue discards such as authorized at Rocky Flats under a safeguards termination variance. The first approach emphasizes Laboratory commitments to environmental stewardship, worker safety, and fiscal responsibility. This approach has been described as the Plutonium Disposition Methodology (PDM) in deference to direction provided by DOE Albuquerque.[1, 2] The second approach is born of the need to expedite removal of residues from storage for programmatic and reasons and residue storage safety concerns. Any disposition path selected must preserve the legal distinction between residues as Special Nuclear material (SNM) and discardable materials as waste in order to insure the continuing viability of Laboratory plutonium processing facilities for national security operations.

2.0 WASTE MINIMIZATION AS A DISPOSITION OBJECTIVE

One viewpoint is that the solution to the problem of excess legacy residue inventories is simply to dispose of them as waste. This is essentially the approach that has been initiated at the former Rocky Flats Plant (Rocky Flats has 3 metric tons of plutonium in residue form. A simplistic analysis of a discrete subset of these residues -the one metric ton of plutonium in pyrochemical salts –will serve to illustrate a point that may not be intuitively obvious. Plutonium in pyrochemical salts is readily recovered by aqueous chloride dissolution. Generally, 85 to 95% of the Pu can be recovered in a single dissolution effort. The criticality safety limit for a 208 liter drum is 200 grams of ^{239}Pu, which is to include 2 x radioassay error. Assuming an assay error of 10% (which is unconservative), the drum limit for fissile materials becomes 166 g*Pu [166 + (2 x 10%) = 200]. If the entire metric ton is disposed of as waste, this results in 6024 drums of transuranic waste. On the other hand, if 95% of the plutonium is recovered, the remaining 5% only constitutes 301 drums. Clearly, plutonium recovery dramatically reduces waste. Considering the 25,000 year half-life of ^{239}Pu, unnecessary geologic disposal of plutonium residues may not be a prudent approach to solution of the problem. Hence, a primary focus of the methodology described herein is the identification of a disposition path that minimizes generation of transuranic waste.

3.0 DISPOSITION FUNDAMENTALS: MINIMIZING PROLIFERATION CONCERNS

The most fundamental paradigm of residue processing is that they are attractive for proliferation and terrorism purposes. By definition, plutonium-bearing residues are Attractiveness Level D Special Nuclear Material [3], and as such, residues require safeguarding and security under the Atomic Energy Act of 1954 (as amended), as well as the Nuclear Nonproliferation Treaty. Residues cannot be discarded as waste until they are rendered unattractive for proliferation

purposes. The initial focus of PDM research was the development of a technical rationale for determining when residues could be considered so unattractive. This rationale, referred to as "Attractiveness Level E criteria", is now published as national policy.[3] Attractiveness Level E criteria are those plutonium concentration ceilings for groups of unstabilized residue matrices that represent the mean concentration of plutonium in materials that have been subjected to a single recovery effort using best available recovery technology. A higher (richer) Attractiveness Level E criterion is also established for residues immobilized in monolithic forms such as glass or cement; such immobilization clearly adds significant difficulties for any diversion or recovery effort, hence the higher allowable plutonium concentration. The basis for the PDM research described herein is to identify the plutonium concentration at which residues should be discarded to immobilization in order to minimize the quantifiable negative consequences of residue processing (cost, resource expenditure, waste generation, and radiation dose).

The other approach to reducing proliferation risks to acceptable levels was pioneered at Rocky Flats and involved utilization of a robust inner container as barrier (deterrent) to diversion and blending of richer materials down to < 10 wt*% Pu, and minimizing SNM quantities and time outside of the Protected Area. This disposition path was necessarily supported by a detailed analysis of the entire discard process in a Vulnerability Assessment.

4.0 PDM RESEARCH APPROACH AND INTERIM RESULTS

The basic hypothesis of this research is that an optimum discard point [Pu-concentration below which items will be discarded to (1) solid transuranic (TRU) waste if below the Attractiveness Level E criterion for the matrix, or (2) cement or glass immobilization if above the Attractiveness Level E criterion] can be identified by quantification of the consequences of processing to an iterative range of prospective discard limits. In other words, the consequences of discard over the range of possible discard concentrations from recovering all of the Pu (0 g*Pu/Kg*Net discard limit) to discarding all of the plutonium as waste (1000 g*Pu/Kg*Net discard limit) will be determined by modeling of recovery process and calculating consequences (cost, direct, indirect, & ancillary wastes, dose) of each operation. The logic of this approach is depicted simplistically below.

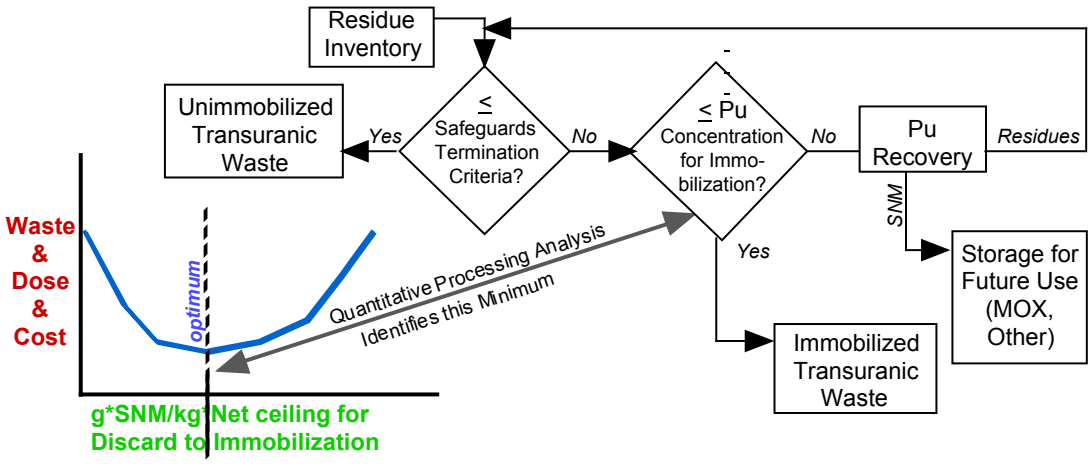


Figure 1: Residue Disposition Rationale

PDM models have been developed which identify minimum negative consequence discard concentrations for hydroxide cakes, SS&C, graphite, filter residues, pyrochemical salts, and glovebox sweepings.[1] The following discussion focuses on the pyrochemical salt model as an example of the process and results obtainable with this analysis approach.

The conceptual pyrochemical salt model (see Figure 2) was designed to quantify the results of extracting the ^{241}Am isotope versus traditional disposal as waste with evaporator bottoms. The reason for this investigation is two-fold: (1) determining is this separation could reduce dose, and (2) preservation of an important national resource. This latter reason bears further discussion. ^{238}Pu is very important to the U.S. for national security as well as science applications. Many of our deep space missions, such as the recent Mars Rover we all watched on television, are entirely dependent on ^{238}Pu for thermal and electrical energy. In the past, ^{238}Pu was produced in reactors at Savannah River, which are now slated for decommissioning. We now have no domestic source for ^{238}Pu and a very limited national inventory. ^{241}Am can be a source for ^{238}Pu via neutron bombardment in accelerators. Thus, extraction of ^{241}Am from pyrochemical salts can be a very important route to meeting a national security and important science need. Note that some pyrochemical plutonium purification operations extract ^{241}Am from plutonium oxides; thus some pyrochemical salts have very high concentrations of ^{241}Am.

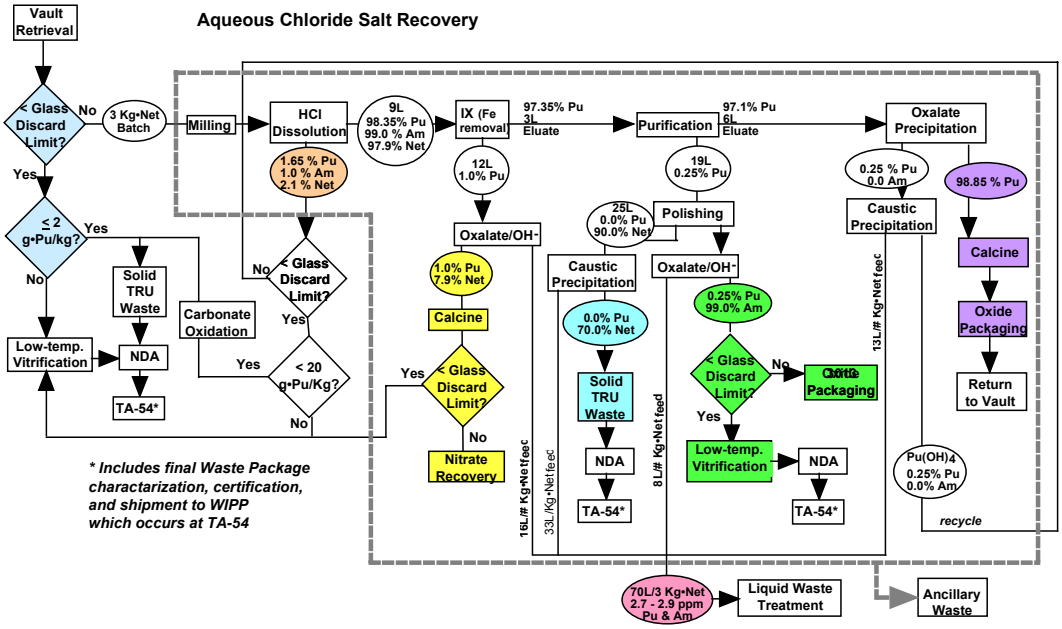


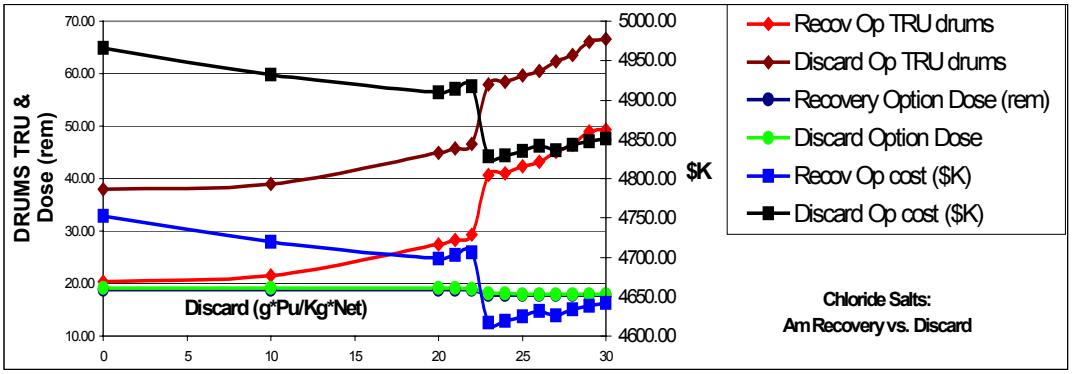
Figure2: Conceptual Pyrochemical Salt Recovery with Am-241 Separation

The aqueous chloride salt model identified minimum operating costs at a discard limit of 23 g*Pu/kg*net for both Am-rich actinide recovery and discard options. However, the results indicate a discard limit of 23 g*Pu/kg*net also coincides with the largest decrease in dose rate and largest increase in waste generation. When evaluating the individual unit operations, it became apparent that this trend was caused by the disposition of the calcined ion exchange batches. For discard limits between 0 and 22 g*Pu/kg*net, the concentration of Pu in these

[1] Dispositions for hydroxide cakes, SS&C, and graphite have been approved by DOE; the graphite disposition has yet to be implemented.

batches (increasing from 19.9 to 22.1 g*Pu/kg*net, respectively) resulted in their being sent to nitrate recovery. Once the discard limit was increased to 23 g*Pu/kg*net, however, the concentration of Pu in the calcined ion exchange batches, about 22.8 g*Pu/kg*net, resulted in their being discarded via low temperature vitrification. It is important to note that continuous (as opposed to discrete event) modeling of this recovery process would not have identified this aspect of the modeled process. Results also indicate that regardless of the discard limit, the recovery of Am-rich actinide results in lower costs and personnel doses than its discard. Note that the difference in dose between the two options is not apparent due to axis scale.

Figure 3: Comparison of Am-241 Separation(from the Pu-bearing substrate) v. No Separation



4.2 UNCERTAINTIES REGARDING COSTS

The operational costs for plutonium processing are well understood at the floor level. The costs of individual operations in plutonium recovery are readily available in terms of manpower, equipment utilization, materials, maintenance, etc. However, the heterogeneous nature of residue inventories, which are composed of materials from research & development and production process upsets, makes broad-brush estimation of costs of plutonium recovery down to a specific discard limit problematic. Depending on factors such as the physical nature of plutonium contamination (surface, embedded, or a constituent of the matrix chemical compound), the degree of the past calcination of the item, and the refractory nature of the substrate, the effort required to achieve a discard concentration may vary considerably among individual items within an inventory subset. In most cases the solubility of both the plutonium and the matrix substrate changes with multiple dissolution events (this is only logical: after the initial leaching/dissolution event, the more refractory materials remain in the solid). In some cases the matrix substrate is more soluble than the plutonium, which results in an item becoming richer (less discardable in terms of plutonium concentration) in plutonium after a recovery effort. Review of historical processing of hydroxide cakes via aqueous nitrate dissolution revealed numerous cases where as many as six dissolution efforts were necessary to achieve the now-obsolete Economic Discard Limits[2]. To properly account for costs (and other processing consequences) of recovery to a specific Pu concentration, the research method described herein analyzes the mass distribution and differing dissolution rates of repeated steps over the cradle-to-grave recovery process.

The real uncertainties in terms of costs are all derivative of the waste management process, which unfortunately continue to evolve as more regulators find careers in the disposal process.

[2] Historically, the residue discard determination was made through equating the cost of production of new plutonium with the cost of recovery through what were called “Economic Discard Limits” or EDL’s. The EDL rationale was declared invalid by DOE after the Strategic Arms Reduction Treaty (START) reduced inventory requirements. [4]

The current approach to this research uses a cost of $12,473.00/drum for transuranic waste which is derived from a peer-reviewed analyses by the Los Alamos National Laboratory performed in 1994.[5] There are two problems with this approach. First, the $12,673.00/drum cost includes a $7000.00/drum cost for disposal at Waste Isolation Pilot Plant (WIPP). This cost estimate comes from DOE Environmental Management Program direction, but a little investigation shows it is derived from the projected WIPP construction cost divided by the projected transuranic waste inventory. A sounder approach to disposal cost estimation would be to divide the WIPP facility life-cycle cost of $8.4B [6] by the design capacity. This results in a disposal cost of $9950 per 208L drum.

Secondly, the cost of processing transuranic waste has escalated since the 1994 analysis and continues to escalate, primarily due to ever-increasing characterization requirements. The estimated cost for generator conditioning of transuranic waste at LANL is now estimated at $5462/drum (or $9322/drum for nonconforming waste such as cement that exceeds wattage limits) and the cost for post-conditioning characterization and certification is $4000/drum. Furthermore, there is an estimated cost of $222/drum for transportation to WIPP (which doubles for cement drums or drums containing heavy pipe components used in discard all scenarios).

Escalation of disposal costs will only drive the optimum discard values derived by the research toward lower plutonium concentrations (i.e., toward more Pu recovery). However, there are model scoping problems discussed in paragraph 4.3 that may also bear on optimum actinide concentration for discard.

4.3 ANALYSIS SCOPING ISSUES

The current analysis scope is depicted in Figure 8. In the current scope, actinides are recovered to oxide form and placed in long-term storage awaiting some decision on future usage. The model is thus limited because the DOE has not yet determined the ultimate disposition of Pu oxides to be recovered from residues that are considered excess to national needs. The apparent possible dispositions are depicted as Paths A (electricity production as MOX), B (direct disposal as a vitreous monolith with fission products –the "can-in-canister" approach), and in an environmentally sound future, Paths A + C (electricity production with reprocessing).

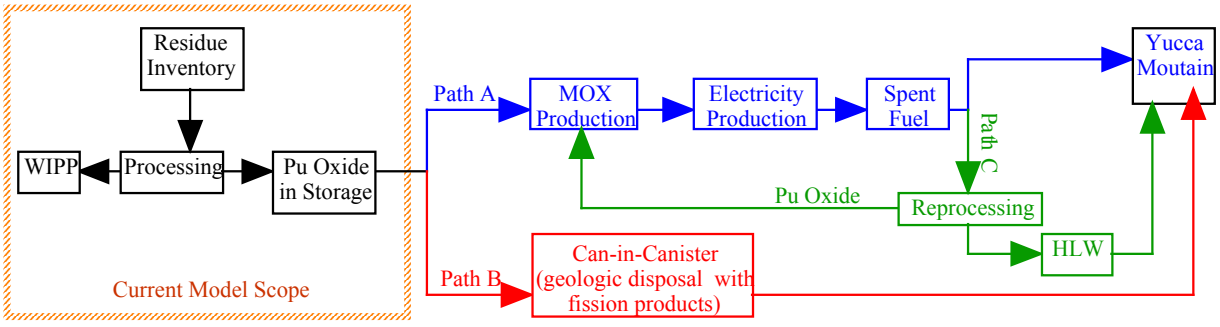


Figure 4: Current and Logical Scope Element Additions

In order to identify a valid minimum negative consequence disposition for residue inventories, models should be extended to incorporate and compare postulated endpoints. This could be done

using historical throughput and cost data from the reprocessing and limited MOX operations in the U.S., and data from ongoing commercial operations in Europe and elsewhere. The DOE Office of Material Disposition would need to perform more detailed cost and dose estimates for the can-in-canister approach than have currently been published to properly incorporate Path B in the disposition model.

It should be noted at this juncture that proponents of the discard-all to WIPP scenario have argued, "why should we pay to recover Pu only to throw it in the ground anyway?" (making the unwarranted assumption that can-in-canister geologic disposal would be the recovered Pu's ultimate disposition). Both economies of scale and environmental stewardship make it intuitively clear that this scenario bears further analysis. The can-in-canister will incorporate 80 Kg*Pu in ceramic discs within a vitreous monolith. Considering again the 1 MT of Pu in Rocky Flats pyrochemical salts, which would constitute a minimum of 6024 drums at a cost of $125.6M[3], this MT would make only 12.5 canisters. The cost of these canisters would have to exceed $10M each before this option becomes more expensive to the taxpayer. Considering the 250,000 years of interest in land disposal of Pu-239 (10 half-lives), certainly disposal of vitreous monoliths containing Pu in ceramic form would be preferable to 6024 drums of unimmobilized Pu salts from an environmental stewardship perspective. If then Path B in Figure 8, which is the least favorable from a resource utilization perspective, is better than the popular dispose-all at WIPP scenario, clearly all options should be rigorously examined cradle-to-grave.

5.0 ANALYSIS OF DISPOSITIONS THAT EXPEDITE RESIDUE DE-INVENTORY

Assessment of legacy residue options for the purpose of expediting de-inventory can be described as the "watershed" (path of least resistance") approach. There are several fundamental constraints and assumptions that bear on such an assessment, including:

- $\geq$ 30 wt*% WG-Pu oxide can be packaged in "3013" cans for disposition by Material Disposition (MD) at Savannah River;
- the SNM-concentration ceiling for Attractiveness Level D residues is 10 wt*% (Attractiveness Level C material is not normally eligible for Discard under a Safeguards termination variance);
- WIPP RCRA Permit characterization requirements will dictate any disposal campaign schedule (residues in "debris" form must be processed first as the Laboratory does not have the analytical capabilities required for "homogenous" wastes and interim storage of residues discarded under a Safeguards termination variance at TA-54 is not an option)
- Logistics constraining NDA and TRUPACT-II loading will be onerous due to anticipated inventory limits and security requirements outside TA-55's Protected Area.

Considering these factors, worst case and best case scenarios can be estimated in terms of waste generation. If the entire legacy residue inventory were discarded as waste, it would comprise approximately 4530 drums (assuming 135 g*Pu/drum where the matrices are not wattage limited, which is what Rocky Flats achieved in their salt blending operations, and assuming the maximum Pu allowed where matrices are wattage limited).
As the ceiling for a safeguards termination variance is 10 wt*%, it is clear that processing of the richest materials to oxide for disposition at Savannah River would be the cost and resource

[3] Using costs developed in Section 5.1 + $1000/drum for the stainless steel inner vessel required by the safeguards termination variance (residue salts, by definition, exceed Attractiveness level E Pu-concentrations).

effective path forward for this inventory subset (because the same effort is required to recover or blend up as to the blend down and divide, and significantly fewer 3013 containers are produced than more expensive waste drums). If the > 10 wt*% ash, filter residues, glovebox sweepings, and pyrochemical salts were blended, and average of >30 wt*% would result, which would meet DOE-STD-3013-2000. (ash, filter residues, and sweepings would simply require calcinations; salts would require aqueous recovery and calcinations). This part of the inventory would become ~90 3013 cans, as opposed to ~ 3000 drums of TRU Waste.

In considering materials to be discarded as waste, it is necessary to evaluate them in categories of debris and homogenous waste due to differing analytical requirements, as previously stated. Homogenous wastes require coring and analysis of VOC's and SVOC's (Volatile & Semi-volatile Organic Compounds). Note that immobilized wastes (cement, glass) are considered homogenous. If such wastes are immobilized (and thus do not require a Safeguards termination variance) they can be stored at TA-54 until the analytical capability becomes available. If such wastes have been or will be subjected to high-temperature processing (e.g., calcinations or vitrification) the VOC/SVOC requirement is eliminated. Given these constraints, the next group of materials that could be similarly processed would include lean (< 10 wt*%) salts, ash, and filter residues, along with various hydrogenous and potentially reactive materials (not reactive in the RCRA-sense).[4] These materials could be packaged in a pipe component under a safeguards termination variance. However, only the ash, salts, and hydrogenous and formerly reactive materials could be sent to WIPP (because of their previous thermal treatment). The other materials would impinge on TA-55's storage capacity, so they really shouldn't be processed until analytical capabilities become available. On the other hand, if these materials were immobilized in a low-temperature vitrification process (developed by the Laboratory for Rocky Flats, but never implemented on a production scale), they would not require a variance or VOC/SVOC analysis. It is important to understand here, that regardless of whether the pipe component or vitrification is employed, the number of drums that would be comprised by this inventory subset would remain the same, ~570. The difference lies in the requirement for a Safeguards termination variance, analytical requirements that cannot currently be met, and very difficult processing logistics.

The following table illustrates some of the consequences of the two alternatives only partially discussed above. These alternatives are by no means all inclusive –other avenues for expedited have been and are being evaluated. The taxpayer costs below include the disposal cost discussed in paragraph 4.1. The DP cost estimates only include waste management operations at TA-55 – costs for processing residues to a form that can be accepted by waste management are not included. The LANL EM costs are for required NDA, analytical characterization, and certification subsequent to TA-55 waste management operations. It should be clear that options involving PU recovery elements are significantly less expensive in terms of waste management/disposal costs and represent a significant waste minimization as well. The costs for Pu recovery, calcinations, immobilization, blending, and packaging have not been estimated thus far, but analysis results using the PDM have already demonstrated that such costs will not exceed the costs of preparation for discard ALL disposal.

[4] Hydrogenous and reactive materials would require calcining to maximize wattage limits and eliminate reactivity.

Waste Generation & Waste Management Cost Comparison				Waste Management Costs		
Alt.	Description	# 3013's	# Drums TRU	Taxpayer Cost ($M)	DP Cost ($M)	LANL EM Costs ($M)
1	Discard ALL	0	**4530**	**90.0**	24.7	18.120
2	≥10 wt*% Pu (salts/ash/filter residues) to 3013's	84	0	0	0	0
	Low-temp vitrify <10 wt*% salts/ash/filt.residues + hydrogenous/reactives	0	568	11.152	3.102	2.272
	Recover 85% of > 10 wt*% MgO	10	55	1.080	0.300	0.220
	Implement existing graphite disposition	0.5	15	0.294	0.082	0.060
	Blend & Calcine ≥ 10 wt*% glovebox sweepings to > 30 wt*% for 3013's (~85% of total Pu)	7	38	0.746	0.208	0.152
	Discard remaining "Debris" in pipe component	0	470	9.332	2.567	1.880
	Total	103.5	**1146**	**22.6**	6.3	4.6

7.0 SUMMARY & FUTURE RESEARCH NEEDS/DIRECTION

It is clear that intelligent analysis of residue inventories and available processing options can be utilized to both minimize wastes and reduce costs. The many constraints on residue processing, particularly at the Laboratory where storage capacity at the plutonium processing facilities is at a premium and where a number of important waste management functions occur outside of the Protected Area, demand a rigorous analysis in terms of resource demands and process bottlenecks if any kind of an expeditious de-inventory of legacy residues is to be accomplished.

The results of the research thus far need to be effectively communicated to Weapons Program decision-makers who tend to favor disposal of residues as waste because (1) residue processing and storage is in competition for personnel/facilities also required for programmatic operations, and (2) it is not fully understood that the personnel who recover Pu are the same personnel that must prepare waste for disposal in the event of such a disposition decision (thus, discard all decisions do not minimize the drain on programmatic resources that residues as a whole constitute).

Disposition rationales for legacy residues must not degrade the ability of national security facilities to process Special Nuclear Material (SNM) in terms of both technical capacities and regulatory constraints, and must provide a valid technical and *legal* basis for ongoing disposition processes. This last point is rather obscure, but very important. Residues are not waste by virtue of their attractiveness for proliferation (they are considered SNM and are not *discardable*; consequently, residues cannot be considered waste until they are rendered unattractive for proliferation purposes). Residues are stored and processed in the same systems/facilities as more strategic forms of plutonium. It is imperative for national security reasons that a valid basis for disposition of residues be followed, as opposed to across-the-board discard decisions such as have occurred at other DOE facilities. When residues become waste by judicial or management *fiat*, the entire inventory becomes subject to environmental laws requiring intrusive and constraining permitting/licensing , which is not an option for essential national security facilities.

Finally, legacy residue disposition is a task that needs serious attention. Storage safety concerns were identified in 1994, and operational needs for storage space at TA-55 have been understood for some years. Yet the resources devoted to residue processing have decreased over this same timeframe, while external pressures to solve residue problems have increased. During the Reagan Administration, the Laboratory recovered more Pu *annually* than exists in our current legacy residue inventory.

REFERENCES

[1] LA-UR 98-4596, *Development & Implementation of Attractiveness Level E Criteria and the Plutonium Disposition Methodology*, D.C. Christensen & M.A. Robinson, Nuclear Materials Technology Division, Los Alamos National Laboratory

[2] Plutonium Disposition Methodology, U.S. Department of Energy, Weapons Quality Division, Albuquerque Operations Office, October 1994

[3] DOE Order O474.1, Safeguards & Security, U.S. Department of Energy, 1999

[4] DP-273, "Plutonium Disposition Plans", C.G. Halsted, Director, Office of Nuclear Weapons Management, U.S. DOE, April 18, 1994

[5] LA-12757-M, Manual, *Los Alamos Waste Management Cost Estimation Model*, L.M. Matysiak and M.L. Burns, Chemical Science Technology Division, Los Alamos Nation Laboratory, 1994

[6] The Baseline Environmental Management Report, Volume III, New Mexico – Wyoming, U.S. Department of Energy, Office of Environmental Management, Office of Strategic Planning and Analysis, June 1996, pp New Mexico 81-82

[7] LA-UR-01-2149, *Modeling Of Plutonium Recovery And Discard Processes For The Purpose Of Selecting Optimum (Minimum Waste, Cost, & Dose) Residue Dispositions*, Mark A. Robinson, Monika B. Kinker, Ronald E. Wieneke, Los Alamos National Laboratory, Los Alamos, New Mexico