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Title:

THE IMPACT OF A VARIETY OF REPROCESSING OPTIONS ON THE PURITY, WASTE GENERATION, AND PERSONNEL EXPOSURE

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The Impact of a Variety of Reprocessing Options on the Purity, Waste Generation, and Personnel Exposure

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ABSTRACT

A study of reprocessing options for recycled plutonium to be cast into metal ingots of specified purity is presented. The options include: nitrate and chloride aqueous processes, molten salt extraction, molten salt electro refining, direct oxide reduction, and multiple-cycle direct oxide reduction. A preliminary examination of a variety of unit operations determined that two viable options exist for purification of plutonium:

- a combination flow sheet which uses hydrometallurgical and pyrometallurgical processes and

- a flow sheet which uses primarily a hydrometallurgical processes.

The data presented are of the impacts on product purity, waste generation, personnel radiation exposure, and plutonium utilization. The simulation used in this study was performed with ProMoS, a software package developed at the Los Alamos National Laboratory written in the LISP programming language. The simulation was started with 1000kg of impure plutonium metal and operated until the feed was consumed. ProMoS provided the data upon which the comparisons for product purity, waste generation, personnel exposure and plutonium utilization were made.

BACKGROUND

Simulation models are used in the TSA-7 group at the Los Alamos National Laboratory (LANL) to provide data critical to the assessment of complete systems such as a manufacturing enterprise with linked inventory control for feed and intermediate and final product items[1]. The simulation tools developed in TSA-7 are provided to allow the examination of a production process before implementation to assure that the capital and operating costs are efficiently utilized[2]. The simulation also estimate personnel exposure, scheduling impacts, equipment availability, production efficiency, operating personnel requirements, and inventory behavior prior to operation[3]. This allows those responsible for the process start-up to be forewarned of any potential problem areas.

The tool used in this study--ProMoS (Process Modeling System)--was developed to provide the ability to follow the progress of "items" (discrete parts or batches of material) through a complex processing flow sheet to determine how the attributes of those "items" change throughout the process. The attributes typically observed are mass, impurity levels, state, concentration, age, and residence time. Items can be feed items or materials, product parts, product material, in-process residues, or waste materials and packages.

INITIAL FLOW SHEET OPTION SCREENING

The ProMoS simulator was used to provide an initial screen of potential processing options to treat impure plutonium--required for the metal to meet ingot casting specifications. The result produced two viable flow sheet options: the first, or baseline, consists of the collection of hydrometallurgical and pyrometallurgical unit operations currently being used at Los Alamos to purify plutonium metal (Figure 1). The second option is based upon a hydrometallurgical circuit (Figure 2).

Both options use a molten salt extraction (MSE) process to remove americium before each of the baseline and alternate purification processes. The Americium removal is performed up front to reduce the operating personnel exposure to acceptable levels. Americium contamination in aged plutonium metal which is such that handling this material would expose the operators to potentially harmful levels of gamma radiation. The americium "grows" into the plutonium due to ^{241}Pu decay.

SIMULATION OF TWO ALTERNATIVES

The first alternative analyzed was the baseline flow sheet. This option was the result of evolutionary development in plutonium processing at Los Alamos over 50 years and is the current processing flow sheet used in the Nuclear Materials Technology (NMT) division at LANL to produce pure plutonium metal. This flow sheet is displayed in Figure 1. As can be seen, americium removal is the first step. This is done by adding chlorine gas to molten plutonium metal to oxidize the americium into the salt produced at the surface of the metal. The metal--cleaned of americium--is then purified using a combination of pyrometallurgical and hydrometallurgical unit operations.

Much of this flow sheet consists of processes which extract and purify plutonium from in-process residues produced by operations such as ER (electrorefining), aqueous chloride and nitrate recovery operations, and oxide roasting of casting skulls and oxide. This combination was developed to maximize plutonium recovery.

The second option--designated as the Alternative Flow sheet--eliminates ER, salt oxidation and chlorination, salt distillation and aqueous chloride processing. As can be seen in Figure 2, the alternative is a much simpler flow sheet with 70% of the steps, one-third of the residue types and one-half of the waste types. This options appeared to offer many advantages before the detailed analysis was completed.

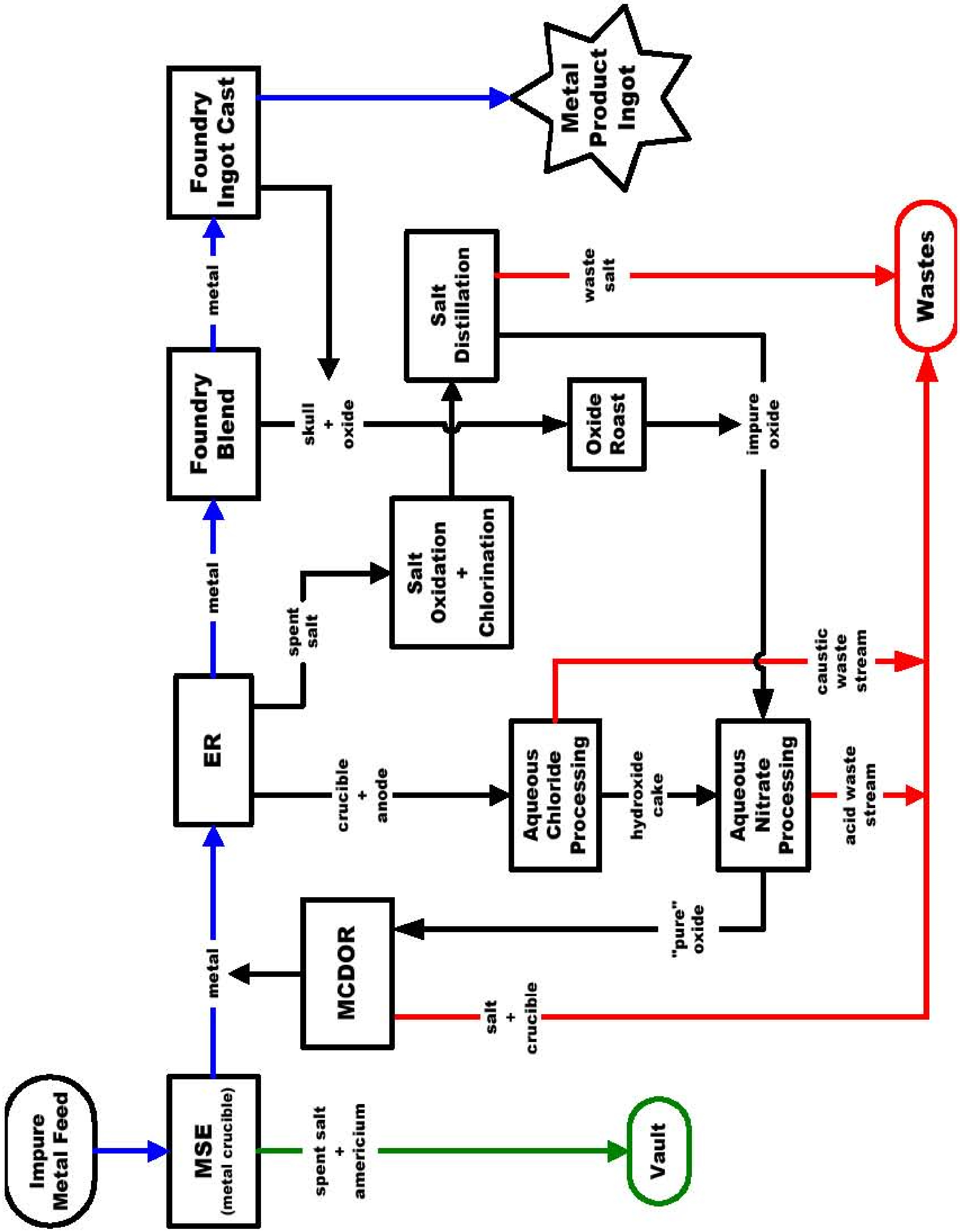

Figure 1. Baseline flow sheet for plutonium processing

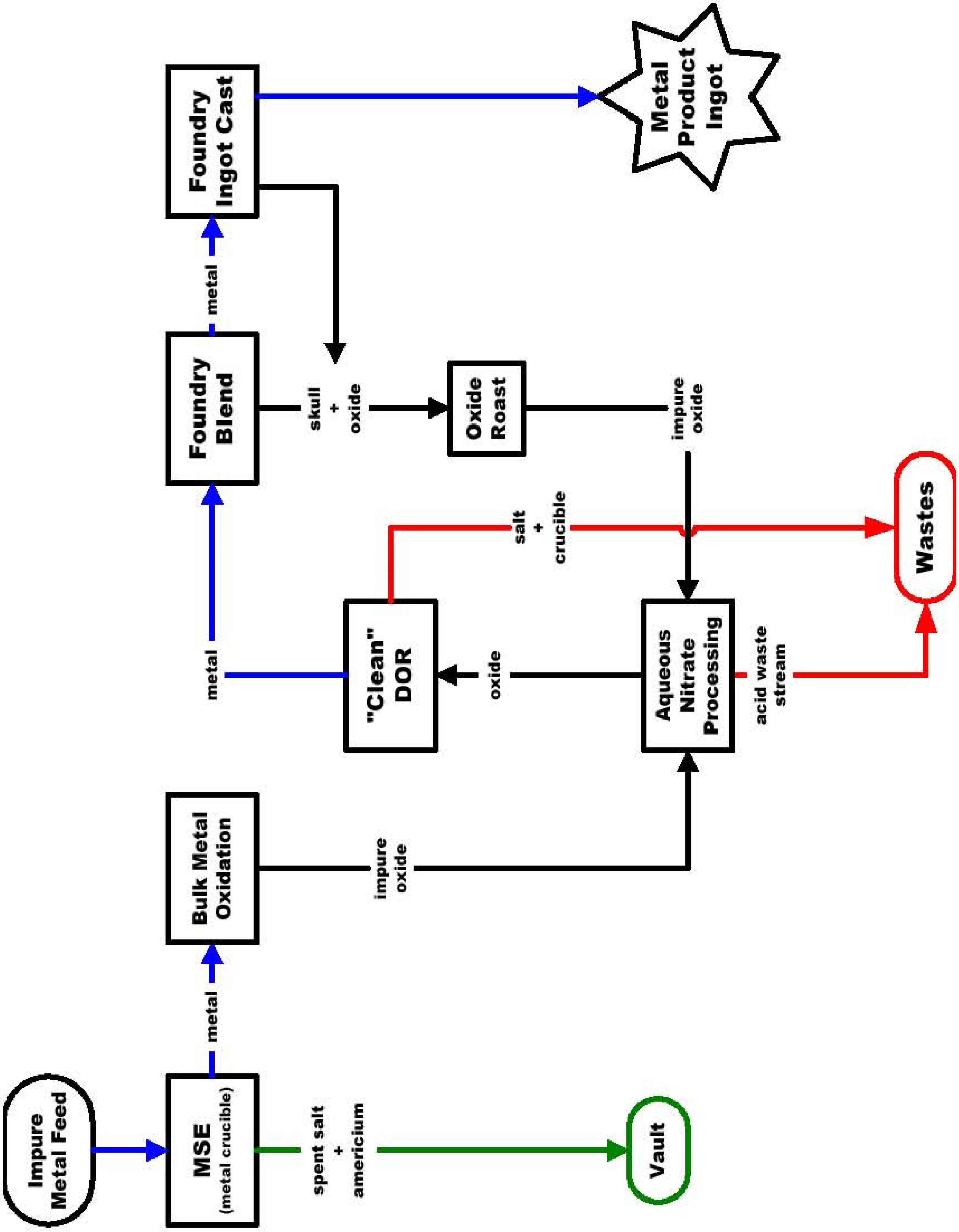

Figure 2. Alternate flow sheet for plutonium processing

SUMMARY AND CONCLUSIONS

The detailed simulation of the two options determined that the original flow sheet was better is nearly every category. The data presented in Table 1 show the distribution of plutonium as a result of the various processing steps. The metal purity is essentially the same for both flow sheet options, with a very slight advantage to the alternative flow sheet. However, in every other category, the baseline flow sheet out-performed the alternative. The Summary information shown in Table 2, clearly shows that the baseline process loses 46% more plutonium to the waste stream and yields 3% more plutonium to the product ingot.

The radiation exposure data in Figure 3 shows that the radiation exposure to baseline process operating personnel is 31% of the exposure of the personnel operating the alternative process. The difference is particularly dramatic for the aqueous nitrate processing operation which is understandable due to the increase of material flow through this step in the alternative, as is shown in Table 1.

The data in Figure 4 indicates that the baseline option requires 56% of the personnel compared to the alternative flow sheet. This is supported by the data on the number of "parts" or batches of material handled by personnel in each of the two options. As can be seen in Figure 5, the baseline personnel handle 67% fewer parts. This supports both the personnel loading and radiations exposure data.

The bottom line: even though the alternative process seemed superior from a flow sheet simplicity standpoint, it did not provide the advantages that are essential to an actinide processing operation over all. The radiation exposure and personnel resource loading are critically important contributors to the operating costs. Therefore, the simulation has confirmed what experience has developed over the years--the baseline flow sheet offers advantages in efficiency, exposure, resource utilization and product flow.

REFERENCES

1. ***Methodology for Developing Decision Support Tools Focused on Highly-Constrained Operational Facilities*, Boerigter, ST. Los Alamos National Laboratory Technical Report LA-UR-00-911, 24 February 2000.**

2. ***Simulation-Based Analysis of a Pit Manufacturing Facility and Model Granularity*, Boerigter, ST, Rising, TL Los Alamos National Laboratory Technical Report LA-UR-00-1729, 4 April 2000.**

3. ***Alternatives for increasing the nuclear materials processing space at Los Alamos for future missions*, Kornreich, DE, DeMuth, NS, Zerkle, CE, Stark, WA, Kubat-Martin, KA, Richardson, DH, Haarman, RA, Boerigter, ST. Los Alamos National Laboratory Technical Report LA-UR-97-1000, 30 March 1997.**

Table 1. Comparison of simulation output for Baseline and Alternate flow sheets

Material Flow (as fraction of input material)	Flowsheet 1				Flowsheet 2			
Location	Pu	Americium	Iron	Total Mass	Pu	Americium	Iron	Total Mass
Foundry Blend	98%	0.05%	0.00%	1,236 kg	95%	0.00%	0.01%	1,337 kg
Foundr Ingot Cast	94%	0.05%	0.00%	1,337 kg	92%	0.00%	0.01%	1,444 kg
Product Ingot	91%	0.05%	0.00%	918 kg	88%	0.00%	0.01%	1,015 kg
Vault Storage	5%	97.00%	5.00%	930 kg	5%	97.00%	5.00%	930 kg
Chloride Waste Stream	0%	0.00%	2.20%	4,603 kg	0%	0.00%	0.00%	482 kg
Nitrate Waste stream	0%	0.02%	0.52%	263,091 kg	0%	0.03%	0.88%	666,839 kg
Wastes	1%	2.93%	91.74%	586,866 kg	3%	2.96%	93.84%	1,430,516 kg

Table 2. Summary performance data for Baseline and Alternate flow sheets

Material Destiny (kgs)	Flowsheet 1		Flowsheet 2	
Location	Americium	Pu	Americium	Pu
Sample Archive	0.00	10.79	0.00	21.68
Vault Storage	48.50	50.00	48.50	50.00
Product Ingot	0.02	908.31	0.00	879.20
Waste	1.46	10.79	1.48	30.48
In Process Remaining	0.01	20.11	0.01	18.64
Grand Total	50.00	1,000.00	50.00	1,000.00

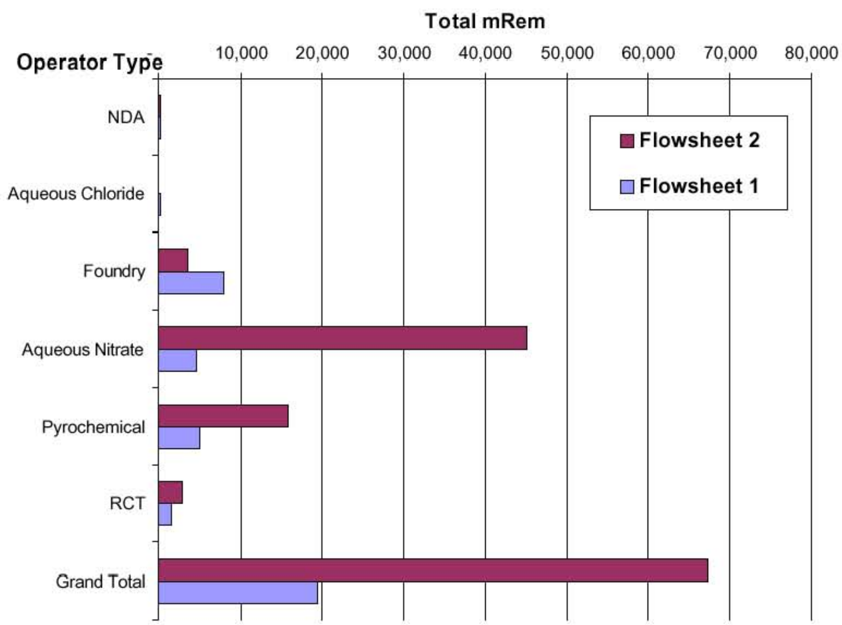

Figure 3. Personnel radiation exposure for the Baseline and Alternate flow sheets

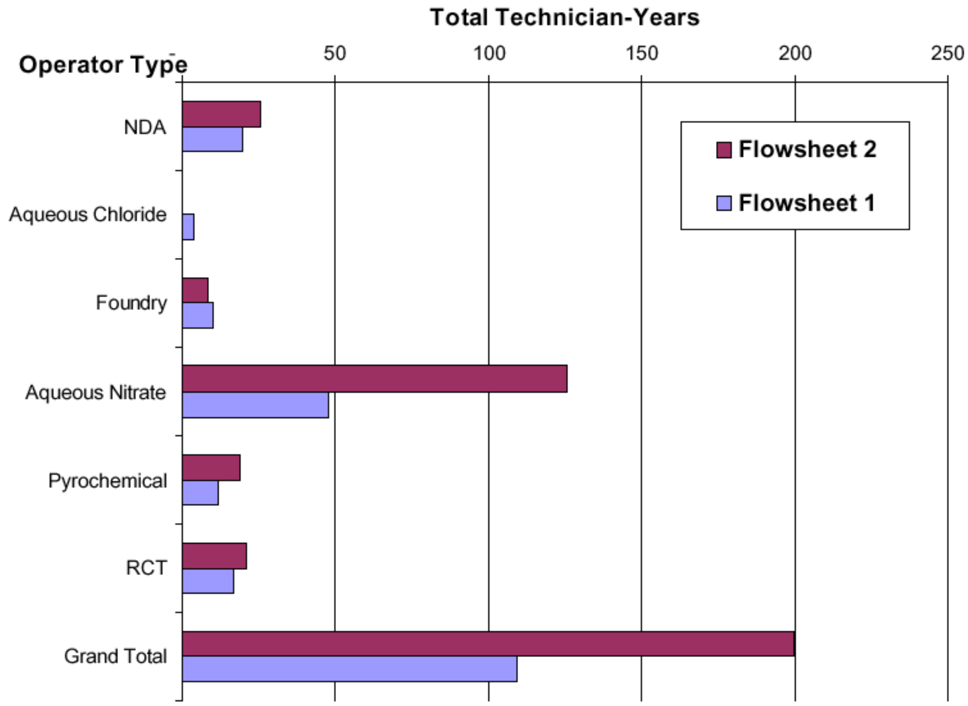

Figure 4. Operating personnel requirements for Baseline and Alternate flow sheets

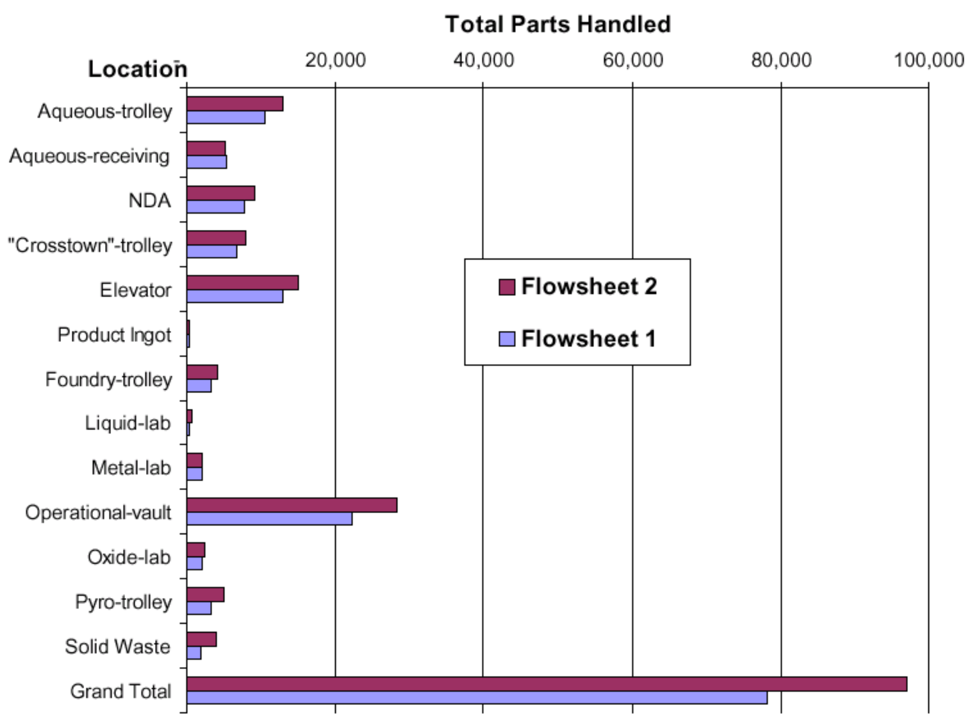

Figure 5. Parts handling requirements for Baseline and Alternate flow sheets