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**Volume Reduction of Low-Level
Contaminated Metal Waste by Melting—
Selection of Method and Conceptual Plan**

G. L. Copeland
R. L. Heestand
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OAK RIDGE NATIONAL LABORATORY
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METALS AND CERAMICS DIVISION

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SELECTION OF METHOD AND CONCEPTUAL PLAN**

G. L. Copeland, R. L. Heestand, and R. S. Mateer

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VOLUME REDUCTION OF LOW-LEVEL CONTAMINATED METAL WASTE BY MELTING—SELECTION OF METHOD AND CONCEPTUAL PLAN

G. L. Copeland, R. L. Heestand, and R. S. Mateer*

ABSTRACT

A review of the literature and prior experience led to selection of induction melting as the most promising method for volume reduction of low-level transuranic contaminated metal waste. The literature indicates that melting with the appropriate slags significantly lowers the total contamination level of the metals by preferentially concentrating contaminants in the smaller volume of slag. Surface contamination not removed to the slag is diluted in the ingot and is contained uniformly in the metal. This dilution and decontamination offers the potential of lower cost disposal such as shallow burial rather than placement in a national repository. A processing plan is proposed as a model for economic analysis of the collection and volume reduction of contaminated metals. Further development is required to demonstrate feasibility of the plan.

INTRODUCTION

Current disposal methods for contaminated metallic wastes are undergoing a review to determine if improved methods are needed. Current Department of Energy (DOE) guidelines require retrievable storage for all DOE waste containing transuranic elements above a miniscule amount. The high cost of this retrievable storage and ultimate disposal in a repository is a strong incentive to reduce the volume requiring storage. The project at the Oak Ridge National Laboratory (ORNL) on "Volume Reduction and Resource Methods for Contaminated Metal Wastes," began in FY 1977 to investigate methods of volume reduction and resource recovery for contaminated metal wastes.

The initial emphasis in the program was to clean the bulk of metals to the point that they could be recycled either within DOE or as commercial scrap. The contaminants would be concentrated in a relatively small volume of slag for reclamation or permanent disposal. Metals that are not decontaminated sufficiently for recycle would be reduced in volume as ingots which are easily handled for burial or storage. The current emphasis is primarily on the technical and economic feasibility of melting for volume reduction alone.

Prior work in this field has shown that melting for volume reduction is a technically feasible approach to consolidating metal waste and that melting effectively removes uranium contamination from many metals. The removal of transuranic elements appears feasible also but has not been extensively demonstrated. The literature that we reviewed and our experience have indicated that induction melting is the most promising method. For a preliminary assessment of the economic feasibility of melting for volume reduction, the following three classes of materials are considered:

1. materials destined for retrievable storage regardless of treatment,
2. materials suitable for shallow landfill burial without treatment;
3. materials destined for retrievable storage which by melting treatment could be made suitable either for shallow landfill burial or reuse.

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Preliminary indications are that melting for volume reduction simply to reduce space required for disposal will be economically justified for material required to be placed in retrievable storage. For material suitable under current guidelines for shallow landfill disposal with no further treatment, volume reduction will probably not be economically sound. More detailed analysis is required to determine whether melting for decontamination and reuse is economically advantageous for material suitable for landfill burial under present guidelines. Melting for decontamination will permit a large economic gain since metals destined for retrievable storage can instead be buried in a landfill or reused after melting.

Programs at other sites involving decommissioning and decontamination are related to this program. For example, the remote disassembly and size reduction program at Atlantic Richfield Hanford Company¹ and the electropolishing work at Battelle Northwest Laboratory² will furnish data on methods of size reduction and disassembly of contaminated equipment that will be directly applicable to equipment that must be reduced and partially disassembled for melting. Electropolishing as a decontamination method has some advantages and disadvantages compared with melting. Where tools and equipment can be reused in their original condition after decontamination by electropolishing, the advantage is obvious. Where the material after treatment is ultimately placed in retrievable storage, buried in a landfill, or recycled as scrap metal, the decision as to which method to use will depend on the economics which are not completely determined at this time. Factors to consider are the capital costs, volume of secondary wastes generated, and operating costs. Electropolishing will require more pretreatment of equipment such as degreasing and paint removal. Electropolishing is probably more suited to a continuous production line operation and would not be as affected by irregular feed-through rates. Electropolishing does not provide any volume reduction, and many pieces will have irregular shape and contain cracks, welds, etc., which will make verification of low nuclide content difficult for landfill burial or recycle as scrap. Ingots resulting from volume reduction will be uniform and easily checked for radioactive nuclide content by gamma energy assay of the entire ingot.

MELTING FOR METAL RECOVERY

Thermodynamic Basis for Removal of Contaminants by Melting

The following calculations are an attempt to provide a thermodynamic basis for assessing the potential for removal of transuranic (TRU) contaminants from a metal by melting. This treatment assumes that the contaminant can combine with oxygen and enter a slag which acts as an inert solvent for the oxides of the contaminant and the base metal. Generalized expressions are developed for the equilibrium constant and partition ratio of the contaminant between the melt and the slag. The generalized expression for formation of the oxide of contaminant metal R is

The expression for the formation of the oxide from a melt of the pure metal M is

1. G. A. Beitel, "Remote Disassembly of Radioactively Contaminated Vessels by Means of an Arc Saw," ARH-SA-289 (June 1977).

2. R. P. Allen, H. W. Arrowsmith, and W. C. Budke, "Electropolishing as a Large Scale Decontamination Technique," presented at the annual AIChE Meeting, November 14-18, 1977, New York.

Subtracting Eq. (2) from Eq. (1),

Equation (3) essentially represents the reduction of the base melt oxide from the slag by the contaminant R with the contaminant oxide being formed and dissolving in the slag. We have the equilibrium constant K_{eq} for Eq. (3) in terms of the activities of the components:

$$K_{\text{eq}} = (a_{R_D O_E})^{1/E} (a_M)^{B/C} / (a_R)^{D/E} (a_{M_B O_C})^{1/C} \quad (4)$$

Solving for the activity of the contaminant in the melt,

$$(a_R)^{D/E} = (1/K_{\text{eq}}) (a_{R_D O_E})^{1/E} (a_M)^{B/C} / (a_{M_B O_C})^{1/C} \quad (5)$$

$$a_R = (1/K_{\text{eq}}^{E/D}) (a_{R_D O_E})^{1/D} (a_M)^{BE/CD} / (a_{M_B O_C})^{E/CD}$$

According to Henry's law, the activity of a substance in small concentrations in a solvent can be represented as proportional to its concentration,

$$a = k \cdot \text{concentration.}$$

Thus, the activity of the contaminant oxide in the slag is

$$a_{R_D O_E} = k_{R_D O_E} \cdot \text{molar fraction}_{R_D O_E}$$

$$a_{R_D O_E} \approx k_{R_D O_E} \cdot \frac{\text{weight } R_D O_E}{\text{total slag weight}}$$

The activity of the metal contaminant dissolved in the melt can similarly be written

$$a_R \approx k_R \cdot \frac{\text{weight } R}{\text{weight melt}}$$

If we define the partition coefficient as the amount of contaminant in the slag divided by the amount of contaminant in the melt, then the partition ratio, λ , is approximately

$$\lambda \approx \frac{a_{R_D O_E} / k_{R_D O_E}}{a_R / k_R} \cdot \frac{\text{weight of slag}}{\text{weight of melt}} \quad (6)$$

The ratio of the activities, $a_{R_D O_E} / a_R$, can be obtained from Eq. (5):

$$\frac{a_{R_D O_E}}{a_R} = K_{\text{eq}}^{E/D} \cdot \frac{(a_{M_B O_C})^{E/CD} (a_{R_D O_E})^{1-1/D}}{(a_M)^{BE/CD}} \quad (7)$$

Inserting Eq. (7) in Eq. (6),

$$\lambda \approx \frac{k_R}{k_{R_D O_E}} \cdot K_{\text{eq}}^{E/D} \cdot \frac{(a_{M_B O_C})^{E/CD} (a_{R_D O_E})^{1-1/D}}{(a_M)^{BE/CD}} \cdot \frac{\text{weight of slag}}{\text{weight of melt}} \quad (8)$$

The activity of the base metal in the melt, a_M , should approach unity at equilibrium and can be neglected. To make some very approximate calculations, assume the following conditions:

weight of slag/weight of melt = 0.1;

a_{MBOC} = activity of base metal oxide in slag = 0.1;

a_{RDOE} = activity of contaminant oxide in slag = 0.01.

Then the partition ratio, λ , is

$$\lambda = (0.1)(K_{eq})^{E/D} (0.1)^{E/CD} (0.01)^{1-1/D} (k_R/k_{RDOE}) \quad (9)$$

$$\lambda = K_{eq}^{E/D} (10)^{-3+E/CD-1/D} (k_R/k_{RDOE})$$

Also, the equilibrium constant, K_{eq} , can be calculated from the relationship

$$\Delta F_{\text{reaction}}^{\circ} = -R T \ln K_{eq} \quad (10)$$

$$\text{or } K_{eq} = e^{\frac{-\Delta F_{\text{reaction}}^{\circ}}{RT}} \quad (11)$$

where $\Delta F_{\text{reaction}}^{\circ}$ is in cal/g atom oxygen and is calculated for Eq. (3), $R = 2$, and T = temperature in Kelvin.

We now have a calculated value or an assumed value for everything needed to calculate λ except for the constants from Henry's law, k_R and k_{RDOE} . For the moment, assume these constants are unity (ideal solutions) for a sample calculation for the removal of uranium from an iron melt at 1900 K. The oxide reaction considered is

Comparing Eq. (12) with Eq. (3), we see that $B = 1$, $C = 1$, $D = 1$, and $E = 2$. From Glassner³ we obtain $\Delta F_{\text{FeO}} = -1.36 \times 10^5$ J/mol ($-32,500$ cal/g atom) and $\Delta F_{\text{UO}_2} = -3.81 \times 10^5$ J/mol ($-91,100$ cal/g atom). The free energy change for Eq. (12) is -2.45×10^5 J/mol ($-58,600$ cal/g atom).

$$\begin{aligned} K_{eq} &= \exp \frac{-\Delta F_{\text{reaction}}}{RT} \\ &= \exp \frac{-(-58,600)}{(2)(1900)} \\ &= 5 \times 10^6 \end{aligned}$$

From Eq. (9),

$$\begin{aligned} \lambda &= K_{eq}^{E/D} (10)^{-3+E/CD-1/D} \\ &= (5 \times 10^6)^2 (10)^{-3+2-1} \\ &= 5 \times 10^{10} \end{aligned}$$

Starkey, Quigley, and McElvey⁴ report data from melting 6×10^6 kg of uranium-contaminated steel which permit calculating λ from actual results obtained. The 231 kg of slag (8.5% of melt weight)

3. Alvan Glassner, "The Thermochemical Properties of the Oxides, Fluorides, and Chlorides to 2500°K," ANI-5750 (1957).

4. R. H. Starkey, et al., "Health Aspects of the Commercial Melting of Uranium-Contaminated Ferrous Metal Scrap," *Ind. Hyg. J.* 21, 178-81 (April 1960).

contained 2.12% uranium or 5 kg. The ingots contained < 3 ppm uranium or 0.008 kg. (The contaminated steel had originally contained 2000 ppm uranium.)

$$\lambda = \frac{\text{weight uranium in slag}}{\text{weight uranium in ingot}}$$

$$= \frac{5}{0.008} = 6 \times 10^2$$

This actual λ of 6×10^2 is obviously quite different from the calculated value of 5×10^{10} . There are several sources of potential experimental errors and calculational or assumptional errors. The most likely source of error of this magnitude is in the assumption that the Henry's law constants are unity. These constants can differ greatly from unity. For example, the constant for titanium in nickel has been shown to be about 10^{-5} (Ref. 5).

In spite of the potential for large error, partition ratios were calculated using this approach for several base melts with oxide slags and several contaminants using ΔF values from Glassner³ (Table 1). As shown above, these partition ratios are at best several order-of-magnitude approximations. However, they agree in order of sequence with the available data presented below. Kinetic factors are also not considered, and a marginal partition ratio (i.e. about 10^{-8} to 10^8) suggests that slag composition, interfacial conditions, and presence of other impurities may control for these systems in full-scale operations. Calculations can readily be made for other temperatures and base metal contaminant systems of interest. The primary purpose of these calculations and presentation of Table 1 is to aid in the interpretation of experimental results later in the program and in understanding previous results listed below. Determination of the Henry's law constants during the experimental program will allow much more accurate extension of calculated partition ratios to conditions not determined experimentally.

5. D. J. Bradley, "The Thermodynamics of Carbon in Nickel-Based Multicomponent Solid Solutions," ORNL/TM-6282 (April 1978), p. 144.

Table 1. Partition Ratios for Oxide Slags for Several Contaminants and Melts^a

Contaminant	Base Melt and Temperature							
	Tin 600 K	Lead 600 K	Aluminum 1000 K	Copper 1400 K	Iron 1900 K	Nickel 1900 K	Chromium 1900 K	Zirconium 2100 K
Thorium	$10^{5.5}$	$10^{6.8}$	10^4	$10^{3.1}$	$10^{1.4}$	$10^{1.7}$	10^9	10^0
Hafnium	$10^{4.4}$	$10^{5.7}$	10^{-3}	$10^{2.6}$	$10^{1.0}$	$10^{1.2}$	10^5	10^{-3}
Uranium	$10^{4.2}$	$10^{5.6}$	10^{-3}	$10^{2.4}$	$10^{1.1}$	$10^{1.4}$	10^6	10^{-3}
Neptunium	$10^{3.8}$	$10^{5.1}$	10^{-6}	$10^{2.4}$	10^9	$10^{1.3}$	10^4	10^{-4}
Plutonium	$10^{3.3}$	$10^{4.1}$	10^{-3}	$10^{2.0}$	10^8	$10^{1.1}$	10^6	10^{-2}
Titanium	$10^{2.9}$	$10^{4.5}$	$10^{-1.0}$	$10^{2.1}$	10^7	$10^{1.0}$	10^1	10^{-7}
Wolfram	10^{-2}	$10^{1.2}$	$10^{-2.9}$	10^8	10^{-2}	10^1	10^{-7}	$10^{-1.4}$
Cobalt	10^{-7}	10^0	$10^{-1.9}$	10^0	10^{-5}	10^{-3}	10^{-8}	$10^{-1.1}$
Technetium	$10^{-2.8}$	10^4	$10^{-2.9}$	10^3	10^{-8}	10^{-3}	$10^{-1.6}$	$10^{-2.5}$

^aPartition ratio is defined as the amount of contaminant in the slag divided by the amount of contaminant in melt at equilibrium. Values in this table are approximations calculated using Eqs. (9) and (11) in the text and assuming the Henry's law constants are unity. Values > 1 indicate contaminant will concentrate in slag; < 1 indicates contaminant will primarily stay in melt.

Previous Work

Successful decontamination of uranium from several metals has been accomplished on a laboratory and commercial scale. Mautz et al.⁶ have reviewed the literature and compiled the results of work done before 1975. Blatz⁷ reported the results of laboratory and pilot plant melting of uranium-contaminated steel. Decontamination factors of from 10 to 40 were observed with mean uranium concentration in the resulting ingot ranging from 0.5 to 1.5 ppm. Klevin and Harris⁸ reported the results of laboratory melts of uranium-contaminated nickel, nickel and stainless steel, stainless steel, aluminum, and copper. The uranium content of all but the aluminum was reduced to around 1 ppm. The aluminum was not successfully cleaned by this oxidation treatment due to preferential combination of aluminum with oxygen. The uranium content of the aluminum ingots was reduced 30 to 50%. Fullam⁹ reported less success using a variety of slags to decontaminate stainless steel. He found residual contamination uniformly dispersed throughout the metal even after several melts with a slag cover. This result is not surprising, however, since one of the radioactive contaminants in the stainless steel melt was cobalt. Table I shows an unfavorable partition ratio for cobalt. We have no data on kinetic factors, but probably very little cobalt would be removed to the slag until essentially all the chromium in the steel was oxidized. Decontamination factors up to 1000 were measured when the metal was washed with the molten slag prior to melt down. Production-scale work has been reported by several DOE facilities, including the Fuel Materials Production Center^{4,10} operated by National Lead Company at Fernald, Ohio, the Y-12 Plant¹¹ at Oak Ridge, Tennessee, and ORNL¹² (both operated by the Union Carbide Corporation, Nuclear Division). The ORNL report involved 7.7×10^4 kg of stainless steel yielding ingots ranging from less than 0.02 to 0.04 ppm uranium and 1×10^5 kg of copper yielding less than 0.02 to 3.10 ppm uranium. The Y-12 work recovered 2.7×10^7 kg of uranium-contaminated common steel scrap resulting in ingots containing 0.4 to 0.6 ppm uranium. The Y-12 melts also included 2.2×10^6 kg of stainless steel ingots with an average uranium content of 0.6 ppm. Nickel and nickel alloys resulted in 0.01 to 4 ppm uranium, and copper and brass alloys ranged from 0.01 to 2.5 ppm uranium. The FMPC work involved 6×10^6 kg of steel contaminated with uranium, and 3.5×10^5 kg of steel contaminated with radium. The resulting ingots contained less than 3 ppm uranium, and the radium content was reduced to an average of 9.0×10^{-5} ppm.

Argonne National Laboratory¹³ is completing a program to show the feasibility of removing plutonium contamination from scrap steel. The objective of their work was to clean the metal of TRUs to below the 10 nCi/g level required for DOE waste disposal in nonretrievable burial. Their laboratory results indicate that this degree of decontamination was achieved except for a minor increase in

6. E. W. Mautz, et al., *Uranium Decontamination of Common Metals by Smelting—A Review*, NLCO-1113 (February 1975).

7. Hansen Blatz, "Sale of Uranium Contaminated Steel Scrap Recommended," *Iron Age* 170, 125-7 (September 25, 1952).

8. P. B. Klevin and W. B. Harris, "Remelting May Permit Reclaiming Uranium-Contaminated Metals," *Nucleonics* 14, 93-96 (April 1956).

9. H. T. Fullam, *High-Temperature Methods for Disposal of Contaminated Metal Equipment*, BNWL-B-277 (July 1973).

10. R. H. Starkey, J. A. Quigley, and J. W. McKelvey, "Health Aspects of the Commercial Melting of Radium Contaminated Ferrous Metal Scrap," *Ind. Hyg. J.* 22, 489-93 (December 1961).

11. M. Sanders and J. D. McLendon, Union Carbide Corporation, Nuclear Division, Y-12 Plant, internal correspondence, circa 1958-1960.

12. D. M. Davis, J. C. Hart, and A. D. Warden, "Hazard Control in Processing Stainless Steel and Copper Contaminated with Uranium," *Ind. Hyg. Quart.* 18, 235-41 (September 1957).

13. T. J. Gerding, M. G. Seitz, and M. J. Steindler, "Salvage of Plutonium and Americium-Contaminated Metals," presented at the annual AIChE Meeting, November 14-18, 1977, New York.

contamination on the skin of some ingots. Decontamination factors as high as 10^6 were measured. A second melting may be required in some instances to overcome the slight concentration in the skin of the ingot which raises the local concentration to above the limit for disposal.

Current Programs

There are several programs in progress at the present time producing information of interest to this task. FMPC¹⁴ is proposing a portable smelter that would be moved to a given location to process the accumulated metal scrap over a period of several months and then be moved to another location. The Paducah Gaseous Diffusion Plant¹⁵ is presently operating a nickel smelter for noncontaminated scrap, and the processing of contaminated scrap is planned. The possibility of using the Paducah facility for ferrous scrap is being considered. The Oak Ridge Gaseous Diffusion Plant has proposed an arc furnace for processing contaminated ferrous scrap from the diffusion plants.¹⁶ Other efforts such as the work to process high-level waste at Hanford are not directly applicable but are producing information on dismantling and cutting methods and melting techniques which are helpful to the low-level programs. Other programs such as fuel cladding hull consolidation provide data on melt-crucible interactions and decontamination factors.

Selection of Melting Method

Several melting methods have previously been considered for smelting of contaminated scrap metals including electric induction, electric arc, and fossil fuel fired. FMPC¹⁴ reviewed and listed the advantages and disadvantages of each of these types in their feasibility study for a portable smelter. We selected a medium frequency electric induction furnace for our study for many of the same reasons it was selected by FMPC: high melting rate, simple collection of dust and fumes, ability to melt all scrap under consideration, good control of melting conditions, controlled agitation, and good control and minimum volume of slag. BNWL¹⁷ has investigated Inductoslag melting for metals such as zirconium which require good atmospheres and cold crucible techniques. The method is promising for special applications where size reduction is easily accomplished and the metals are being recycled. However, Inductoslag melting is not justified for general volume reduction.

COLLECTION, TREATING, AND STORAGE PLAN

Types of Scrap Considered

This program is primarily considering the routine day-to-day low-level contaminated scrap consisting mostly of equipment such as glove boxes, hand tools, pumps, motors, electronics racks, and piping. With the present collection system in the Oak Ridge area, it is difficult to estimate the quantity of this metal scrap generated. However, a reasonable approximation may be a half ton per day for the three plants in Oak Ridge. A melter about 1 m diam by 1 m deep to accept reasonably sized pieces of scrap will only need to operate several shifts per week to accommodate this scrap generation rate on a routine basis. Several options are available for utilizing the facility the remainder of the time:

1. Utilize the melter personnel for steam decontamination, disassembly, size reduction, and sorting. The melter would only be operated when a sufficient quantity of sized scrap is available for the desired number of melting runs.

14. J. H. Cavendish (ed.), *Feasibility Study of a Portable Smelter for Scrap Metals*, NLCO-1132 (June 1976).

15. T. L. Ashwood, *Processing of Uranium-Contaminated Ferrous Scrap in the C-746-A Facility*, KY-G-259 (May 1974).

16. *Engineering Feasibility Report for a Smelting Facility for Processing Uranium Contaminated Ferrous Scrap*, K-D-3374 (November 1974).

17. R. G. Nelson and B. Griggs, *Chop-Leach Fuel Bundle Residues: Densification by Melting*, BNWL-SA-5704 (1976).

2. Utilize the melter for campaigns to process large volumes of scrap of a particular type such as that resulting from a plant's decommissioning or rebuilding (such as the Capacity Expansion Program at the gaseous diffusion enrichment plants). Specific compositions could be processed separately in these campaigns to maximize the value of the scrap in the event of recycle.
3. Enlarge the geographic area served by the melter to increase the volume. Economics and other transportation considerations will be important in evaluating this option to determine whether another melter should be built or the waste transported to a central facility.
4. Melt some higher level radiation-contaminated scrap in addition to the low-level scrap. Important considerations are the costs of modifying or adding a facility for size reduction of the scrap and the possibility of needing biological shielding for the melter and slag handling facilities. The melted metal could be handled separately or mixed with the lower level scrap depending on the degree of cleanliness achieved and the disposition of the metal.
5. Melt some noncontaminated scrap in addition to or along with the low-level scrap. This, of course, will primarily be an economic decision based on the cost of melting and the value of the scrap before and after melting. Dilution of the melt (after an initial slagging operation) with noncontaminated scrap may enable the melted metal to reach even lower contaminant levels. From consideration of the partition factor developed above, the clean scrap would have to be added after the initial slag was removed with the majority of the contaminants to be effective in lowering the contaminant level in the melt.

The relative merits of these options are dependent on several unknowns at this point. The melter should be sized for handling of reasonable sized pieces of scrap. This size will be more than adequate for handling the expected volume of low-level contaminated metal scrap in a typical region such as the Oak Ridge area.

Nature and Degree of Contamination

The nature of the contamination depends on the origin of the scrap. The melting system with normal slagging operations will concentrate the thorium, uranium, and the TRUs into slag; however, isotopes such as technetium and cobalt will not generally be transferred to the slag. Scrap containing radioactive isotopes not concentrated in the slag should be kept segregated from the other scrap.

For purposes of estimating handling and melting costs, the scrap will be assumed to require no biological shielding for disassembly, sorting, and premelting size reduction. For some scrap, this level of contamination may require precleaning either at the generation site or at the melting facility. This precleaning presents no problem since scrap equipment is normally decontaminated to the maximum extent practical before disposal.

Collection System

The present ORNL radioactive waste handling system bundles contaminated scrap—mostly paper and plastics—into plastic bags and into metal garbage cans for shallow burial. Commercial sites like the one at Maxey Flats, Kentucky, have operated on a similar basis for contaminated trash from DOE and commercial sources. A Nuclear Regulatory Commission (NRC) study¹⁸ concluded that existing commercial burial sites are sufficient to handle the expected waste through 1990. They recommended

18. U.S. Nuclear Regulatory Commission, "NRC Task Force Report on Review of the Federal/State Program for Regulation of the Commercial Low-Level Radioactive Waste Burial Grounds," (January 1977).

that further licensing of burial sites be suspended until the possibility of using improved methods has been thoroughly investigated. One plan being investigated is to incinerate the burnables and recover the valuable nuclides. The metals would be melted for recycle, high density burial, or storage. The recent closing of the burial site at Maxey Flats¹⁹ accentuates the need to find acceptable long-range solutions for the disposal of radioactive wastes.

ORNL is studying a system of separate bins for burnable and inert contaminated trash. If this system is successful, scrap will be available in more manageable form. In our conceptual plan, contaminated scrap will be steam decontaminated, disassembled, and visually sorted into bins, for example, copper coils, iron armatures, and aluminum housing of a motor will go separate routes. Steam, rather than chemical or electrolytic decontamination, is recommended because it avoids chemical wastes. The particulates will be recovered by centrifuge and filter. Centrifuge sludge will be continuously monitored and segregated for disposal on the basis of level of contamination. Filters will be processed along with the burnable trash.

Although commercial scrap processors make little attempt to disassemble and segregate metals, we feel that DOE has a responsibility to attempt to produce quality grades of metallurgical remelt stock, perhaps to show that such conservation measures can be economically attractive to the commercial processors. Typical examples of low-quality scrap are steel containing tin and lead which loosen furnace refractories, and copper in Fe-Ni-Cr matrix which would increase corrosion of stainless steel. Of more fundamental importance is the loss of copper as a recycle tramp element and the loss of chromium to waste slag in the manufacture of mild steel. With this extra effort for scrap separation, the product of the melter would be standard grades of scrap in pig or shot form suitable for remelt. If current policy, regulations, or economics prohibit recycle or reuse, the scrap will be surface stored or shallowly buried in such a manner that it could eventually be reclaimed if economics and need justify it.

When many alloys are heated in an oxidizing atmosphere, the naturally occurring oxide layer thickens. In many cases such as that of aluminum, the oxide skin actually contains the molten alloy for some time after the melting point is reached. When the alloy becomes molten, it eventually breaks through the skin and runs out. Since most radioactive contamination is on the surface, much of the contamination may stay with the oxide skin or dross and provide a high degree of separation of the radioactive nuclides. Conversely, if the metals were placed in a molten bath, the contaminants would dissolve uniformly in the melt, and a slagging operation would be the only means of concentration.

Consolidation

The scrap metals as partially disassembled will be separated according to melting point in a gradient furnace. The moving hearth furnace (or a series of individual furnaces) will be resistance heated to minimize air currents which would burden the offgas contamination filter system. Ceramic pallets with funnel bottoms will be hand loaded, each with presorted nonferrous scrap of one visual type. On passing through the furnace, the pallets will be heated from room temperature to 1100°C. During the early stages of this passage, paint and fragments of plastic will burn off. The various alloys will melt in succession as the temperature reaches their melting ranges. The molten alloys will drain through the funnel bottoms into clay-graphite crucibles in individual resistance furnace enclosures. After collection, the melt in the crucibles will be equilibrated with slag and cast into molds. The sequence of alloys generated will follow the sequence Pb-Sn, Al-Zn-Mg, Cu-Sn-Ni with the unmelted portions consisting mostly of stainless steel and super alloys (Fe-Cr-Ni-Co). These metals which melt at higher temperatures will be collected into crucible charges for the induction melting furnace.

¹⁹ *The Courier-Journal*, Louisville, Ky., December 21, 1977 page B-4; December 31, 1977 p. A-4.

This concept may be applicable to continuous feed of various metals but would probably be most efficient with prior metal separation with the melting conducted in batches.

The magnetic stream, sorted out earlier, consisting of steel, will be preheated in a separate hooded resistance furnace and melted as separate lots in the induction furnace. The dross skins from the drip-melt and preheat furnaces will be collected by vacuum cleaner arrangement for burial or nuclide recovery.

There will be some burnoff of fabric wrappings, rubber bushings, and other materials from the disassembled equipment in the preheat and drip-melt furnaces. A proper offgas filtering system will be required. This initial burnoff will minimize fume problems in the induction melter. If the metal consolidation system were located near an incinerator for burnable scrap, a joint offgas system could probably be utilized.

The induction melting furnace will be a tilt pour type with an offgas collection system. The furnace will be sized for convenience in handling the scrap rather than on the anticipated tonnage per day of scrap to be processed. It is anticipated that a single refractory lining and slag system will serve for all the types of scrap to be handled. However, the feasibility studies may show a need for different slags for various nuclides or metals. The metal product will be controlled only within very wide ranges of composition and thus will be suitable only for remelt stock. The metal could be cast as pigs or shotted depending primarily on the ease and reliability of sampling techniques for alloy content and nuclide content. The slags will be collected either for nuclide recovery or disposal.

Analysis for Radioactive Nuclide Content

The gamma energy assay method is considered the best method for characterizing the nuclide content of the melted metal and the slags and dross. To demonstrate the sensitivity of this equipment, a sample of 206 g aluminum and 0.103 g of depleted uranium was induction melted and cast (nominal 500 ppm uranium in aluminum). The sample was smeared and checked by the area Health Physics technicians with their instruments and determined to be clean. This sample was counted by the Analytical Chemistry group using the gamma energy assay method for an arbitrary 350 s. The results were 0.63 counts/s at 92.95 keV and 2.26 counts/s at 98.72 keV from ^{238}U x-rays and 0.264 counts/s at 186.01 keV from ^{235}U gammas. The sample was later analyzed by the classical wet chemistry technique as 254 ppm uranium at the top (including some dross), 88 ppm at the center, and 94 ppm at the bottom. The results of the gamma energy assay counter can be converted to nCi/g by calibrating the readout with standards for a specific size and shape sample. The product of the melter can be a standard shape suitable for total gamma energy assay. Thus, the gamma energy assay method appears to be a most promising technique for radioactive nuclide analysis of the total melter output.

Chemical Analysis

X-ray fluorescent analysis appears to be the best technique for chemical analysis of the melted metal. This technique is generally limited to elements heavier than aluminum and is primarily a qualitative tool. However, with long counting times and uniform sample geometry such as pigs or shot, it can be calibrated and corrected for interferences to yield quantitative results. In the melter, it can be used to assay the final product and give rough percentages of the major elements and also to detect any unexpected elements. Portable units of this type are available for qualitative use in scrap sorting and could be used as an aid in initial separation of the scrap.

In instances where x-ray fluorescence may not give consistent analyses, the full resources of the ORNL Analytical Chemistry group will be employed to use, among other procedures, emission spectroscopy for impurities detection and standard bulk methods for specific elements. These services will be essential in initial calibration of the x-ray fluorescence unit.

Disposition of Products

The melted metal product will be in the form of cast pigs or drums of shot divided into lots of known approximate composition. If recycle or reuse is not allowed due to regulatory restraints, these lots containing less than 10 nCi/g TRUs or the then current guideline) will be eligible for shallow burial in a landfill. With some minimal additional expense these burials could be categorized so that the value of the scrap would not be lessened if recycle should later become feasible.

If regulations allow sale and recycle of scrap containing *de minimus* quantities (ie., a defined level below which the material is considered to be free of contaminant) of ^{235}U and TRUs, obtaining optimum value from the scrap will probably involve cooperation with one or more premium scrap dealers. The metallurgical remelt industry prizes most highly those grades of scrap where the analysis is known and in which the content is similar to the composition of standard products in regard to major alloying elements with only small amounts of elements known to be detrimental to the class of alloys being made. Specialty alloy producers stockpile scrap purchases with their analysis in a computerized inventory. When a particular alloy grade is to be produced, the computer surveys the inventory and dictates the amounts of various scrap lots to be used in making the melt.

It is inevitable, particularly in the early stages of operation of the melter, that some batches will be of such composition that they cannot be blended into anything acceptable to the remelt industry, for example, the superalloy industry can use Fe-Cr scrap containing either cobalt or nickel, but not both. Lots of this mixed character material will have to be recycled further back into the extraction process. Similarly, some of the Pb-Sn would be diverted to the primary refiners rather than to the solder and battery lead remelters. This need for additional refining lowers the value of the scrap.

By contrast, if the aluminum alloys can be cleaned sufficiently, they will be readily marketable. The aluminum foundry industry can make good use of the strengthening effects of the copper and magnesium which are common elements in aluminum scrap. Aluminum casting alloys can also tolerate relatively high levels of iron and other base metals likely to be introduced in the melting operation. The basic problem with aluminum is how to get the radioactivity levels low enough.

The metal lots that are higher than allowed in nuclide content may be put through the melting operation a second time or placed in a repository for disposal depending on the success rate of cleaning the particular metal of the contaminants present. If burial in a repository is required, the pigs are a minimum volume form and, for most metals, a relatively stable containment for the nuclides.

The slags and drosses will contain most of the nuclides from the incoming contaminated scrap. The slags may be placed in a repository or processed for further nuclide concentration or recycle. The processes and relative merits of this recycling are outside the scope of this study except that the types of slags and relative nuclide contents may be indicated by the feasibility studies. In any event, it will be desirable to minimize the volume of slag generated.

MODELS FOR ECONOMIC ANALYSIS

Economic analysis of the cost and benefits of the volume reduction will be based on the general flow sheet in Fig. 1 using scrap generated at three Oak Ridge plants as an example. These include a gaseous diffusion plant, a weapons production plant, and a research laboratory. This combination should yield a representative cross section of the types of material which would be encountered in a waste metal volume reduction facility. The analysis assumes that the burnable contaminated waste has been previously separated from the nonburnable, preferably through collection methods. The nonburnable waste will be steam cleaned to remove loose contamination. The cleaning sludge will be collected for processing or burial or other storage. The low-level contaminated waste considered for this melter should then be

Fig. 1. Flow Sheet for Economic Analysis of Volume Reduction of Contaminated Metal Waste by Melting.

amenable to hands-on sorting and disassembly/sizing operations with only minimal personnel protection such as protective clothing and a respiratory mask. At this point, the required operations vary, depending on the ultimate disposal or use of the metal. Three options will be considered: (1) simple volume reduction for burial with no reuse or recycle, (2) reuse of the metal within DOE, and (3) sale of metal scrap as remelt stock in the commercial metal scrap market.

Volume Reduction Only

If the metal is not to be reused or recycled in any way, the sorting operation could be simplified. Some sorting would still need to be done to remove insulation, cement, and other materials from the metal. Mixing all the metal scrap together would decrease its worth if reclaiming ever became practical in the future. Sorting and gradient melting should be included along with segregation in burial for this eventuality. Sorting will also improve the melting operation by lowering fumes and lengthening refractory life.

The economic gain from melting for volume reduction is twofold. Retrievable storage is costly both in terms of money and utilization of natural resources. Many items of process equipment are very bulky and not easily amenable to disassembly and compaction. Melting these items and casting as ingots will

minimize the storage volume. Second, the concentration of nuclides in the slag during melting lowers the contamination level of the metal. The metal ingots may then be suitable for near-surface burial in an unsupervised landfill. Only minimal segregation and cataloging would then allow future reclamation should it be desired. The uniformity and density of the ingots will allow gamma assay for nuclide content to verify suitability for burial which might not have been possible before melting due to bulk, geometry, crevices, etc. Thus, melting for volume reduction will reduce the volume requiring burial and will also allow much cheaper burial methods for much of the metal. The nuclides will be concentrated in the slags which will be a relatively small portion of the melter load. The slags will require retrievable storage or reprocessing to reclaim the nuclides. For this analysis, the slags will be assumed to be stored and the question of nuclide recovery will not be addressed.

The following cursory look at the economics shows a decided economic advantage if disposal in a permanent repository can be minimized. The cost of a permanent repository will of course depend on the type, location, and other variables not decided at this time. However, for an approximation, the costs estimated for one of the options evaluated by the Savannah River Plant²⁰ for disposing of high-level wastes as a solid are used. The option considered involves the storage of 3.78 m³ containers of fused salt in offsite geologic storage. For their study, the costs of temporary storage, transportation, and geologic storage added up to approximately \$10,000/m³. Steel has a nominal volume of 1.1×10^{-4} m³/kg. An optimistic estimate of the minimum volume which could be obtained by disassembly, shredding, and compacting is about 2.2×10^{-4} m³/kg. Thus, simply melting 1000 kg of steel scrap to obtain maximum density would avoid storage for 1.1×10^{-4} m³ with a cost avoidance of about \$1200. If the 1000 kg of metal were decontaminated to a level at which it could be shallowly buried at \$141/m³ with 0.014 m³ of slag placed in geologic storage, the cost avoidance would be about \$2200.

The melting cost of steel has been estimated as about \$0.1/kg for the portable melter proposed by National Lead¹⁴ and as about \$0.38/kg for the arc melter proposed by Union Carbide.¹⁶ Melting for volume reduction is obviously economically attractive if geologic storage is required for the waste metal. Melting for volume reduction alone is not economically justifiable for metal which could be shallowly buried at near current costs of about \$141/m³.

Reuse of Metal Within DOE

If we assume that regulations prohibit recycle of materials containing *de minimus* quantities of enriched uranium and TRUs, there are several potential applications where the metal could be reused within DOE facilities. For instance, metal reclaimed from low-level wastes could be used as a matrix or as containers for long-term storage of high-level wastes. The cost and resource benefits of such uses will be compared to the additional melting and quality assurance costs required to reuse the metal. The scope of this program will not permit feasibility studies for reuse.

As a rough approximation, economics of this case are similar to volume reduction alone. Some additional costs will be incurred in closer control of melt chemistry, ingot quality, and quality control provisions for chemical content and radioactive nuclide content. The value of the metal which would have been purchased for the use can be added as a cost benefit. Careful analysis of the fabrication steps required will be necessary to ensure economic feasibility. The volumes involved may well make it economically attractive to purchase parts commercially rather than fabricate them within DOE even with "free" metal.

20. Energy Research and Development Administration, *Alternatives for Long-Term Management of Defense High-Level Radioactive Waste. Savannah River Plant, Aiken, South Carolina*, vol. 1, ERDA 77-42/1, E. I. Du Pont de Nemours & Co., Aiken, S. C. (May 1977).

Commercial Recycle of Scrap Metal

The NRC is now considering an amendment to 10 CFR Part 70 that will define a trivial or *de minimus* quantity of enriched uranium and TRUs that would permit commercial recycle of metals containing less than these amounts. The product of the melter will be ingots (or pigs) or shot that will permit accurate gamma assay for nuclide content and permit easy approximate analysis for alloy content by x-ray emission spectroscopy. The additional costs would be for alloy analysis and quality assurance measures and some additional sorting and segregation to obtain high-quality remelt stock.

The economic considerations are similar to the above cases. However, since the melter product would be remelt stock for the commercial market, its value and utility would be straightforward. The economic benefit of melting for recycle will be marginal as compared with shallow burial. Previous analyses of the large volume of well characterized scrap from the cascade improvement program at the gaseous diffusion plants have shown either a slight economic incentive¹⁴ or slight economic deficit.¹⁶

FEASIBILITY STUDIES PLANNED

The concentration of uranium in the slag and resultant decontamination of the metal has been demonstrated several times for steel, stainless steel, nickel, copper, and brass. Results of aluminum melting show that uranium is removed far less effectively. We plan to determine the decontamination factors for the metals we expect to be encountered in scrap processing. We hope to show that a single crucible-slag combination will suffice for all the metals requiring induction melting. Several methods of decontaminating the aluminum will be tried including drip melting, cryolite as a slag, and degassing agents containing chlorine. If practical, the demonstration melts will be extended to actual scrap mixtures containing TRU elements:

CONCLUSIONS

Several conclusions and observations may be made at this stage of our program. Melting of contaminated metal scrap for volume reduction is technically feasible. Melting for volume reduction alone will undoubtedly be economically justified in cases where the scrap will be placed in a national repository such as geologic storage. Melting for volume reduction alone will probably not be economically justified for scrap that may be shallowly buried under current guidelines.

Assuming that regulations are passed allowing sale of scrap containing *de minimus* quantities of enriched uranium and transuranics, the economics of melting for recycle appear to be marginal as compared with those of shallow burial. Melting of many metals under a slag decontaminates the metal by concentrating the radioactive nuclides in the smaller volume of slag. This decontamination of the metal may allow either much cheaper shallow burial rather than geologic storage or even reuse of the metals. Experiments are under way to quantify these partition ratios for nuclides and metals of interest. Methods of analyzing the resulting metal ingots for chemical content and nuclide content are available without further development.

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