OFFICIAL

BNWL-B-277

UNCLASSIFIED

HIGH TEMPERATURE METHODS
FOR DISPOSAL OF
CONTAMINATED METAL EQUIPMENT

JULY 1973

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

*This report is intended primarily for internal use
by the sponsoring organization and Battelle.*

Prepared for the U.S. Atomic Energy
Commission under Contract AT(45-1):1830

MASTER

BNWL-B-277

UNCLASSIFIED

OFFICIAL USE ONLY

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PATENT STATUS

This document copy, since it is transmitted in advance of patent clearances, is made available in confidence solely for use in performance of work under contracts with the U.S. Atomic Energy Commission. This document is not to be published nor its contents otherwise disseminated or used for purposes other than specified above before patent approval for such release or use has been secured, upon request, from the Chief, Chicago Patent Group, U.S. Atomic Energy Commission, 9800 So. Cass Avenue, Argonne, Illinois, 60439.

PRELIMINARY REPORT

This report contains information of a preliminary nature prepared in the course of work under Atomic Energy Commission Contract AT(45-1)-1830. This information is subject to correction or modification upon the collection and evaluation of additional data.

NOTICE

The report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1)-1830

HIGH TEMPERATURE METHODS FOR
DISPOSAL OF CONTAMINATED METAL EQUIPMENT

By

H. T. Fullam

Chemical Technology Department

July, 1973

This report is based on work sponsored by the
Atlantic Richfield Hanford Company under United
States Atomic Energy Commission Contract AT(45-1)-2130

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON

MASTER
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

SUMMARY

Three high temperature processes for simplifying the disposal of contaminated metal equipment were investigated: (1) fused salt decontamination; (2) metal meltdown under a fused inorganic slag; and (3) metal meltdown under an inert atmosphere.

It was found that contaminated stainless steel could be effectively decontaminated using oxidizing fused salts. Decontamination factors (DF's) of 10^6 or better were obtained using fused sodium nitrate at 800°C and several stages of fused salt-metal contact. The process generates a large volume of contaminated salt, however, which severely limits its practical application. Even using maximum salt recycle, a salt-to-metal weight ratio of 3 to 1 or greater is required to give a DF of 10^6 .

By melting down contaminated metal under a fused inorganic slag it was hoped that the radioactivity would be transferred to the slag phase, leaving a clean metal phase which could be cast into an ingot for uncontrolled reuse. It was found upon melting the metal that only a fraction of the activity was transferred to the fused slag, with the major portion remaining in the metal phase. When the metal phase solidified, the residual activity was dispersed throughout the metal in a nonleachable and nonsmearable form. The process simplifies equipment disposal by reducing the metal bulk volume to a minimum and effectively sealing the residual activity in the solidified metal. The process has the disadvantage of generating a substantial volume of contaminated inorganic slag which must be disposed of.

By melting contaminated metal under an inert atmosphere it was hoped that the activity could be dispersed and fixed in the solidified metal. Experiments showed that, upon melting the metal, the bulk of the activity was

retained in the metal phase while the remainder was vaporized or transferred to the crucible. Upon solidification of the metal, the residual activity was fixed in the metal phase in nonleachable form. Leaching tests with water and acids showed that the rate of pickup of activity by the leach solution corresponded approximately to the rate of metal corrosion. Thus metal meltdown under an inert atmosphere would simplify equipment disposal by consolidating the metal in a minimum volume and effectively sealing the residual activity in the metal phase.

CONTENTS

	Page
SUMMARY	i
INTRODUCTION	1
EXPERIMENTAL PROGRAM	2
METAL SAMPLES	2
ANALYTICAL PROCEDURES	5
EXPERIMENTAL PROCEDURES	6
Fused Salt Decontamination	7
Fused Slag-Metal Meltdown	8
Metal Meltdown	12
RESULTS AND DISCUSSION	13
FUSED SALT DECONTAMINATION	14
FUSED SLAG-METAL MELTDOWN	23
METAL MELTDOWN	32
CONCLUSIONS	37
FUTURE WORK	39
ACKNOWLEDGEMENTS	41

ILLUSTRATIONS

	Page
FIGURE 1. Globar Furnace Used in Meltdown Tests	11
FIGURE 2. Metal Corrosion and NaNO_3 Loss Versus Reaction Temperature	17
FIGURE 3. Leaching of Stainless Steel in Water, After Meltdown Under a Fused Slag	30
FIGURE 4. Leaching of Stainless Steel in 2M HNO_3 , After Meltdown	31
FIGURE 5. Leaching Rate for Beta-Gamma Contaminated Stainless Steel in Water, After Metal Meltdown	35
FIGURE 6. Leaching Rate for Alpha and Beta-Gamma Contaminated Stainless Steel in Water, After Metal Meltdown	36

CONTENTS (contd)TABLES

	Page
TABLE I. Typical Gamma Energy Analysis of Metal Samples	4
TABLE II. Effect of NaNO_3 Reuse on Metal Decontamination	18
TABLE III. Typical Gamma Energy Analysis Data for Metal Samples Contacted with Fused Sodium Nitrate	20
TABLE IV. Decontamination with Fe_2O_3 Containing Salts	22
TABLE V. Gamma Energy Analysis of Metal Samples Melted Under an Inorganic Slag	26
TABLE VI. Effect of Remelting on Removal of Radionuclides from Stainless Steel	27
TABLE VII. Effect of Molten Slag-Metal Contact Prior to Metal Meltdown on Metal Decontamination	28
TABLE VIII. Typical Results from Metal Meltdown Experiments Using Stainless Steel	34

INTRODUCTION

The disposal of failed or excess contaminated metal equipment represents a serious problem to the nuclear industry. This is especially true for bulky or complex items such as tanks, reactors, or piping, which have a large surface-to-weight ratio. Ultimate disposal of such equipment requires either:

1. thorough decontamination of the equipment followed by normal disposal or,
2. controlled disposal of the equipment to insure a minimum loss of radioactivity to the environment.

Either approach to equipment disposal represents a substantial operating cost to the nuclear industry.

At the request of the Atlantic Richfield Hanford Company (ARHCO), Battelle-Northwest performed a study to evaluate the feasibility of certain high temperature processes for simplifying the disposal of contaminated metal equipment. Three different processes were investigated:

1. Metal decontamination using a fused salt.
2. Metal meltdown under a fused inorganic slag with subsequent casting of the molten metal into an ingot.
3. Metal meltdown under an inert atmosphere with subsequent casting of the molten metal into an ingot.

The objective of the first approach was to decontaminate the surface of the metal by contact with the fused salt. In the second process the desired objective was to take up the radioactive contamination in the slag phase leaving a clean molten metal phase which could be cast into an ingot.

The third process was studied as the result of information developed while investigating the second process. Its objectives were to consolidate the

contaminated metal in ingot form, and disperse and fix the radioactivity throughout the metal phase in a nonleachable form.

All three processes were evaluated on a laboratory scale using contaminated metal specimens weighing up to 50 grams each. The tests were carried out using stainless steel samples contaminated with alpha and/or beta-gamma activity. The experimental procedures used, the results obtained, and the evaluation of each process based on the experimental results are discussed in the following sections.

EXPERIMENTAL PROGRAM

METAL SAMPLES

All of the decontamination and meltdown tests were performed using contaminated samples of stainless steel. The samples were obtained from several sources and were contaminated with beta-gamma and/or alpha activity. The samples used, their source, and the radionuclides present were as follows:

- A. Simulated samples - Contaminated samples of 304 stainless steel were prepared by contacting the metal with Hanford high-level Purex Acid Waste (PAW) solution. The stainless steel samples were sectioned from one-inch-diameter rod. Each piece was about 3/8-inch thick and weighed approximately 40 grams. The samples were contacted with approximately two liters of three-year-old PAW solution which was heated to boiling during the day and allowed to cool overnight. The operation was repeated each day for several days with water being added periodically to maintain the volume. After repeated boilings the samples were removed from the solution,

washed with water and dried. Two separate batches of samples were prepared. Representative samples from each batch were analyzed. Samples from one batch showed an average of 15-20 microcuries of beta-gamma activity, while the other batch averaged 35-40 microcuries. The radionuclides detected on the metal samples were ^{90}Sr , ^{106}Ru , $^{110\text{m}}\text{Ag}$, ^{125}Sb , ^{134}Cs , ^{137}Cs , $^{144}\text{Ce-Pr}$ and possibly ^{90}Zr . A typical gamma energy analysis (GEA) of the samples is given in Column 2 of Table I.

- B. Pump impeller samples - A partially decontaminated pump impeller (from B-plant) was obtained from ARHCO. It was sectioned into pieces weighing about 40 grams each. Radioactivity associated with the samples was all beta-gamma. The amount of radioactivity varied somewhat from sample to sample depending on the sample shape and exposed surface area but averaged about ten microcuries total activity each. Radionuclides present were ^{60}Co , ^{90}Sr , ^{106}Ru , $^{110\text{m}}\text{Ag}$, ^{125}Sb , and $^{144}\text{Ce-Pr}$. A typical GEA is shown in Column 3 of Table I.
- C. Dip-leg samples - A section of a dip-leg was received from ARHCO and sectioned into pieces weighing approximately 40 grams each. The previous history of the dip-leg was unknown other than that it originated in the Purex plant. Analysis of several samples indicated an average beta-gamma activity of about 20-60 microcuries per sample together with a much lower level of alpha activity (~ 0.25 microcuries each). The principal beta-gamma emitters present included ^{90}Sr , ^{106}Ru , ^{125}Sb , ^{137}Cs and $^{144}\text{Ce-Pr}$ with lesser amounts of ^{60}Co and $^{110\text{m}}\text{Ag}$. A representative GEA is given in Column 4 of Table I.

TABLE I. Typical Gamma Energy Analysis of Metal Samples

Radionuclide	Counts per Five Minute Period*			
	Simulated Samples	Pump Impeller Samples	Dip-Leg Samples	Alpha-Contaminated Samples
^{60}Co	300	200	200	200
^{106}Ru	5,400	4,000	2,000	1,000
$^{110\text{m}}\text{Ag}$	600	1,000	50	80
^{125}Sb	58,000	5,000	2,500	750
^{134}Cs	800			
^{137}Cs	6,200		20,000	10,000
$^{144}\text{Ce-Pr}$	20,500	60,000	150,000	60,000

* Samples counted on a gamma energy analyzer using a lithium drifted diode or sodium iodide crystal.

D. Alpha-contaminated samples - A section of 1/2-inch 304L stainless steel pipe was obtained from ARHCO and sectioned into pieces about one inch long (approximately 32 grams each). Direct surface measurements indicated an average alpha count of 5×10^6 d/min (about two microcuries) per sample. Beta-gamma activity averaged twenty microcuries per sample, and consisted of ^{60}Co , ^{90}Sr , ^{106}Ru , $^{110\text{m}}\text{Ag}$, ^{125}Sb , ^{137}Cs and $^{144}\text{Ce-Pr}$ (see Column 5 Table I for a typical GEA).

When the program was initiated it was hoped that a wide variety of contaminated metal specimens could be obtained from various plant sources. However, we were unable to obtain the variety of samples desired and testing was limited to the samples described above.

ANALYTICAL PROCEDURES

To determine the effectiveness of a particular decontamination process it is necessary to know the level of radioactivity associated with a metal specimen before and after treatment. Determination of the total activity (alpha and/or beta-gamma) associated with a given metal sample generally requires dissolution of the sample and careful analysis of the resultant solution. Since this is impossible for metal samples which are to be used in decontamination tests, it was necessary to select representative samples from each group of metal specimens and analyze them for total activity. Other samples in the same group were then assumed to have the same approximate initial level of activity. Overall decontamination factors (DF's) obtained with a given decontamination treatment were calculated based on the assumed initial level of activity.

For gamma-emitting nuclides a gamma energy analyzer (GEA) was used to

determine relative amounts of activity associated with a particular metal sample before and after treatment. Decontamination factors for specific nuclides were then obtained from the gamma energy analysis. For samples which had been decontaminated to very low levels the GEA could not be used. Instead it was necessary to dissolve the sample and use beta counting to determine the total activity associated with the sample.

Two different energy analyzers were used. One utilized a sodium iodide well crystal, while the second used a lithium drifted diode. Considerably different counting rates were obtained for similar samples on the two instruments. This accounts for the different counting rates given for similar samples in subsequent sections of the report.

Direct alpha counting can be used to approximate the alpha activity associated with a specific sample. However, this does not provide a reliable measure of the alpha activity because of shielding problems. To determine the decontamination factors for treated alpha-contaminated samples the procedure used was to assume an initial alpha activity (based on analysis of dissolved representative samples) and then dissolve and analyze the sample after treatment.

EXPERIMENTAL PROCEDURES

The procedures used in evaluating the three metal treating processes were relatively simple and straightforward. They consisted basically of analyzing the metal specimens for contained activity prior to treatment, treating the metal specimens under a prescribed set of conditions, and analyzing the treated specimens for residual activity.

The effectiveness of a particular decontamination treatment is defined by the decontamination factor (DF) achieved (the DF is defined as the ratio

of the initial activity to the residual activity). In the metal meltdown tests two factors are important: the amount of the initial activity retained in the solidified metal ingot and the rate at which the activity can be leached from the metal phase. In each of the test procedures the principal problems encountered were related to obtaining an accurate measure of the radioactivity associated with a particular metal specimen.

Fused Salt Decontamination

The fused salt decontamination tests were carried out in a small muffle furnace. The salt and metal specimens were contained in either vycor or impervious alumina crucibles. Test temperatures varied from 400 to 900°C, depending on the salt systems used, with the bulk of tests being carried out at 800°C. Several salt systems were evaluated. Oxidizing fused salts containing sodium nitrate or ferric oxide were the most effective; and most of the tests were made using oxidizing salts. In carrying out the fused salt decontamination tests the following procedure was used.

1. The metal specimen was weighed and analyzed by GEA to determine the initial gamma activity level.
2. The sample was placed in a vycor or alumina crucible (Coors AD99) and covered with a layer of the inorganic salt(s) of known weight.
3. The crucible plus salt and sample were analyzed by GEA.
4. The crucible was placed in the muffle furnace which had been preheated to the desired test temperature (above the melting point of the salt).
5. The crucible was maintained at the test temperature for the required time.
6. The crucible was removed from the furnace and cooled to room temperature.

7. The crucible and its contents were analyzed by GEA.
8. The sample and salt were removed from the crucible.
9. The sample was separated from the salt, washed with distilled water, dried and weighed.
10. The sample, salt and crucible were analyzed individually by GEA to determine the residual gamma activity. Residual alpha activity on the metal specimen was determined by dissolving the sample in aqua regia and analyzing the resultant solution. If the residual gamma activity of the treated sample was very low, it was also determined by analyzing the acid solution.
11. In some experiments several stages of fused salt-metal contact were used. In such cases the treated metal sample, after weighing and counting, was placed in a new crucible and steps 2-10 repeated. Depending on the information desired, either new salt or salt from the previous test was used in the repeat test.

With many of the salts used, specifically those containing nitrates, salt decomposition and/or vaporization at the test temperature was a problem. Therefore the time and temperature at which the salt-metal contact was carried out was generally limited. In most cases the contact time at temperature was held to four hours or less, and the temperature to less than 900°C.

Fused Slag-Metal Meltdown

The generalized procedure which was used for the combination fused slag-metal meltdown tests was as follows.

1. The metal specimen was weighed and the activity level determined by gamma energy analysis.
2. The sample was placed in an impervious alumina crucible (Coors AD99) and covered with a layer of inorganic slag material.

3. The crucible plus sample and slag were analyzed by gamma energy analysis.
4. The crucible and its contents were placed in an impervious closed end alumina tube (Coors AD99).
5. The tube was placed in a Globar furnace in a vertical position and purged with high-purity argon for 30-60 minutes to remove the air.
6. The furnace was then heated to 1500°C and held for one hour under an argon flow. At 1500°C both the slag and metal were molten. The metal, being of higher density, settled to the bottom of the crucible and was covered by the molten slag. Crucible temperature was measured by means of a calibrated optical pyrometer.
7. The crucible was then cooled to room temperature under argon and removed from the furnace.
8. The crucible and its contents were analyzed by gamma energy analysis.
9. The crucible was broken and the metal sample recovered.
10. Residual slag was removed from the metal surface and the sample was weighed.
11. The metal sample was analyzed by gamma energy analysis.
12. The slag was separated as well as possible from the crucible and each was analyzed by gamma energy analysis.
13. If no further testing of the metal sample was planned it was dissolved in aqua regia and the resultant solution was analyzed for total activity.
14. If leaching tests were planned for the sample the following procedure was used. The sample was placed in a stoppered flask with a known volume of leach solution (usually distilled water or dilute nitric acid). The solution was stirred with a magnetic stirrer, and sampled periodically to determine the activity level of the leach solution. The leach tests lasted up to ninety days. For the first few hours of the leach test samples were taken every 1-2

hours. As the tests continued samples were taken on a daily or weekly basis. Because of the very low level of beta-gamma activity in the leach solution, beta counting was used to measure the activity level. Alpha activity was determined by alpha counting. At the conclusion of a leach test the metal sample was dissolved in aqua regia and the resultant solution analyzed for total activity.

The combination of fused slag, molten metal and high temperature represents a very corrosive medium. Considerable effort was expended evaluating various crucible-slag combinations to find systems which would retain their integrity for the required test period at 1500°C. Several different crucible materials were tested including fire clay, graphite, mullite, zirconia, magnesia and alumina. Various slag compositions were prepared and tested using various metal oxides, sulfates, borates, silicates, phosphates and fluorides. The only satisfactory crucible material was an impervious alumina, Coors AD99, and it could be used with only a limited number of slag compositions. Typical slags which could be contained in the fused alumina were

- a. $\text{SiO}_2\text{-B}_2\text{O}_3$
- b. CaO-MgO-SiO_2
- c. $\text{CaO-SiO}_2\text{-B}_2\text{O}_3$
- d. $\text{CaO-Fe}_2\text{O}_3$

Other slags tested were either too corrosive to the alumina or were too volatile.

It was found that a melt temperature of 1500°C was required to obtain good separation between the molten slag and the molten metal. This represents a difficult temperature regime in which to work because it is above the temperature limit for wire-wound resistance furnaces, and is at about the operating limit for silicon carbide resistance furnaces. A resistance furnace was constructed using Globar silicon carbide heaters as shown in Figure 1.

FIGURE 1 GLOBAR FURNACE USED IN METAL MELTDOWN TESTS

The furnace used two sets of four Globars in series. Each set of four heaters was powered by a 110 volt-20 ampere variable transformer. A temperature of 1500°C could be maintained in the furnace with a power input of 3600 watts. The furnace normally required about four hours to reach 1500°C. The furnace temperature was measured using a calibrated optical pyrometer, and was controlled manually using one or both variable transformers.

The problems encountered in finding suitable crucible-slag systems are due primarily to the laboratory scale of the meltdown tests. For a large-scale operation one could go to cold wall containers to hold the molten slag and metal. This would allow a much wider variety of slags and container materials to be used.

Metal Meltdown

The meltdown tests in which the metal specimens were melted under an inert atmosphere were carried out in the following manner:

1. The metal sample was weighed and the activity level determined by gamma energy analysis.
2. The sample was placed in an alumina crucible inside an alumina tube.
3. The alumina tube was placed in the Globar furnace and purged with argon for 30-60 minutes.
4. While maintaining the argon flow, the temperature of the furnace was raised to 1500°C and held there for one hour to melt the metal sample.
5. The sample was then cooled to room temperature under argon and removed from the crucible.
6. The sample was again weighed and the activity level determined by gamma energy analysis.

7. If no further testing was planned for the sample, it was dissolved in aqua regia and analyzed for residual activity by alpha, beta and gamma counting.
8. If leach tests were to be made on the sample they were carried out as described in Item 14 under Fused Slag-Metal Meltdown.

The alumina crucibles, alumina tubes, furnace, and related equipment used in the meltdown tests were the same as those used in fused slag-metal meltdown tests.

RESULTS AND DISCUSSION

The effectiveness of a specific decontamination process is defined by the decontamination factor obtained. When counting techniques (GEA-alpha counting) are used to determine the activity present on a metal specimen a number of factors can influence the results obtained. Sample shape, sample position, self-shielding, etc., will affect the results significantly. For example, when flat metal specimens (1 in. diam x 3/8 in. thick) were counted using a GEA, then turned over and recounted, the following results were obtained.

Sample	Metal Analyzed	GEA - Counts per Five-Minute Period				
		⁶⁰ Co	¹⁰⁶ Ru	^{110m} Ag	¹²⁵ Sb	¹⁴⁴ Ce-Pr
1	Side A Up	128	4,520	683	3,146	7,827
	Side A Down	120	4,747	731	3,280	6,572
2	Side A Up	403	5,738	1,267	4,762	121,372
	Side A Down	291	5,160	1,324	4,916	114,372

Such variations made it difficult to obtain reproducible DF's for individual radionuclides by gamma energy analysis. More reliable data were obtained by determining overall DF's based on dissolution and analysis of the metal

samples after treatment. Because of these variations in the GEA data the results presented in the following sections represent the most conservative data obtained. In most tests the individual DF's obtained, based on GEA data, were higher than those reported. Overall DF's reported are based on analysis of dissolved metal samples and are typical of the results obtained.

In general the metal samples obtained from the various sources behaved quite similarly in their response to a given treatment. In each case effective decontamination required dissolution of the contaminated metal surface indicating diffusion of radionuclides into the metal. Decontamination methods that effectively removed beta-gamma activity were equally effective in removing alpha activity.

FUSED SALT DECONTAMINATION

The experimental results show that stainless steel can be decontaminated to low levels using fused salts. To be effective, however, an oxidizing fused salt must be used which corrodes a substantial fraction of the metal phase. Fused sodium nitrate and salt systems containing ferric oxide were the most effective decontaminating agents tested.

There appeared to be an equilibrium established between the contaminants in the metal phase and those in the fused salt which limited the decontamination obtained in a single contact regardless of the amount of metal dissolved. Hence, for a single-stage contact, the overall decontamination factor (DF) obtained depended to a large extent on the salt-to-metal ratio used in the test.

To decontaminate a metal specimen to very low activity levels several stages of salt-metal contact were required with fresh salt being used for each stage. An additional problem was that the overall DF obtained per stage

decreased as the activity level of the metal specimen decreased. Individual DF's for the specific radioisotopes varied significantly.

Typically, for a process using sodium nitrate at 800°C, metal samples contaminated with 15-20 microcuries of beta-gamma activity, and an initial salt-to-metal weight ratio of 3:1, an overall DF of about 1000 was obtained in the first contact stage. In the second and third contact stages (using a 3:1 weight ratio of fresh salt for each stage) the overall DF's obtained were about 100 and 10 respectively. Residual activity associated with the metal specimen was less than 3×10^{-5} microcuries as determined by beta counting of the dissolved sample. Therefore, to obtain an overall DF of 10^6 at least three stages of salt-metal contact were required, and the weight of salt required was at least nine times the weight of metal processed. By partial recycling of used salt the salt requirement could probably be cut by a factor of 2-3.

Samples contaminated with alpha activity were readily decontaminated with an oxidizing fused salt such as sodium nitrate. The DF's obtained per stage of salt-metal contact were similar to those obtained with beta-gamma contaminated samples.

Sodium nitrate melts at 308°C, and decomposes above the melting point with the liberation of oxygen and oxides of nitrogen. At temperatures above about 600°C the oxygen and/or oxides of nitrogen formed react with the metal phase to form oxides. The oxides formed, in turn, dissolve in the remaining fused salt. At 600°C the metal was attacked very slowly, while at 800°C the metal was corroded at a rate of approximately 3 mils per hour. At temperatures above 800°C the metal was corroded at an even faster rate, but the rates of nitrate decomposition and salt evaporation were also greatly

accelerated. The optimum reaction temperature appeared to be between 750 and 800°C. Data on the rates of metal corrosion and weight loss due to salt decomposition and vaporization as a function of reaction temperature are shown in Figure 2. The corrosion rate data obtained for a specific reaction temperature were very erratic as can be seen by referring to Figure 2. This indicated some unknown and uncontrolled variable affected the rate of metal attack. Unfortunately it was not possible to investigate what this variable might be.

When a metal sample, which had been contacted previously with NaNO_3 , was recontacted with the same salt little or no decontamination was obtained in the second contact (see Table II). If, however, the used salt from a first-stage contact was contacted with an untreated metal sample a good overall DF was obtained. Similarly, used salt from a second-stage contact could be used to successfully decontaminate metal from a first-stage contact. In this way contaminated salt could be reused several times without affecting the DF's obtained significantly. Based on the laboratory scale tests, it appears that, in order to obtain an overall DF of 10^6 in three contact stages at 800°C using maximum salt recycle, a NaNO_3 -to-metal ratio of at least 3:1 is required. On a large scale a countercurrent contactor, if one could be designed, would probably be the optimum design.

The contact time required for effective decontamination with fused sodium nitrate is difficult to define precisely based on the laboratory data. Effective decontamination requires dissolution of the metal surface, and results presented earlier showed the erratic corrosion rate data obtained. Since the data are so erratic it is hard to specify a contact time. It appears, however, that at 800°C a contact time of 1-2 hours is sufficient. Longer contact times led to excessive metal dissolution without increasing

FIGURE 2 METAL CORROSION AND NaNO_3 LOSS VERSUS REACTION TEMPERATURE

TABLE II. Effect of NaNO_3 Reuse on Metal Decontamination

Test Conditions

Temperature - 800°C
Contact Time - 1 hour
Initial NaNO_3 -to-Metal Weigh Ratio $\sim 3:1$

<u>Run 1</u>	<u>Overall DF</u>
1 st Contact - New Salt, New Metal	1120
2 nd Contact - Same Salt, Same Metal	1
3 rd Contact - Same Salt, Same Metal	1
 <u>Run 2</u>	 <u>Overall DF</u>
1 st Contact - New Salt, New Metal	1040
2 nd Contact - Same Salt, New Metal	1025
 <u>Run 3</u>	 <u>Overall DF</u>
1 st Contact - New Salt, New Metal	985
2 nd Contact - Same Salt, New Metal	1070
3 rd Contact - Same Salt, New Metal	915
4 th Contact - Same Salt, New Metal	960

the DF significantly, while shorter contact times gave much poorer DF's.

The individual radionuclides present in the metal phase responded differently to the decontamination treatment, although all could be reduced to very low levels by contact with sodium nitrate. In general, the higher the initial level of activity of a specific radionuclide the more effectively it was removed. For example, with most metal specimens the $^{144}\text{Ce-Pr}$ content was about ten times greater than the ^{106}Ru , yet the $^{144}\text{Ce-Pr}$ could normally be reduced to a nondetectable level (by GEA) in two contact stages while three stages were usually required to reduce the ^{106}Ru to the same level. Similarly, ^{60}Co , which was initially present at very low levels in all samples, required at least three stages to be reduced to a nondetectable level (by GEA). Typical GEA data for metal samples treated with NaNO_3 are given in Table III. The data are for metal specimens from three different sources. At low activity levels (after the first decontamination stage) it is difficult to obtain an accurate measure of the residual activity by GEA. This made it almost impossible to determine the individual DF's obtained in the various contact stages. Instead, it is necessary to dissolve the sample after the final treatment and rely on beta counting to obtain an overall DF.

A variety of other fused salt systems were evaluated, but most were relatively ineffective in comparison to sodium nitrate. Only those systems containing ferric oxide or magnetite approached sodium nitrate in effectiveness. The iron-oxide-containing systems tested included:

- a. $\text{KCl-NaCl-Fe}_2\text{O}_3$
- b. $\text{KCl-LiCl-Fe}_2\text{O}_3$
- c. $\text{NaCl-LiCl-Fe}_2\text{O}_3$

TABLE III. Typical Gamma Energy Analysis Data for Metal Samples
Contacted with Fused Sodium Nitrate

Test Conditions

Reaction Temperature - 800°C
Contact Time - 1-2 hours
Initial Salt-to-Metal Ratio ~3

Run No.	Contact Stage	GEA - Counts per Five-Minute Period					
		⁶⁰ Co	¹⁰⁶ Ru	^{110m} Ag	¹²⁵ Sb	¹³⁷ Cs	¹⁴⁴ Ce-Pr
1	Initial	206	4215	1031	4671		63,457
	1	15	105	35	8		26
	2	3	7	4	0		0
	3	0	0	1	0		0
2	Initial	330	6138	710	53,740	6139	21,342
	1	12	56	41	12	0	71
	2	4	3	0	1	0	0
	3	1	0	0	2	0	0
3	Initial	116	1984	41	2140	23,410	147,200
	1	16	175	8	34	121	332
	2	3	13	0	0	0	0
	3	0	1	0	0	0	1

- d. $\text{KCl-Fe}_2\text{O}_3$
- e. $\text{NaCl-Fe}_2\text{O}_3$
- f. $\text{NaCl-KCl-Fe}_3\text{O}_4$

Table IV shows typical gamma energy analysis data for metal samples before and after treatment with salts containing Fe_2O_3 . Typically, using a salt-to-metal weight ratio of 3:1 per stage, three contact stages, and a reaction temperature of 800°C , ferric oxide-containing salts will give an overall DF of 10^5 to 10^6 from beta-gamma and alpha activity.

A comparison of the data presented in Table IV with GEA data obtained with sodium nitrate (Table III) indicates that the system containing Fe_2O_3 is the more effective decontaminating agent. However, the counting rates were obtained with different instruments and a direct comparison of the results should not be made. Overall results obtained by dissolution and analysis of treated metal specimens showed NaNO_3 to be more effective than ferric oxide containing salts in decontaminating stainless steel.

Salts containing Fe_2O_3 or Fe_3O_4 were quite corrosive to the stainless steel samples, but the rate of attack was less than with NaNO_3 . At 800°C the corrosion rate was about one mil per hour for all Fe_2O_3 -containing systems tested.

Other salt systems tested included:

a. $\text{K}_2\text{S}_2\text{O}_7$	g. $\text{B}_2\text{O}_3\text{-MnO}_2$
b. KCl-LiCl	h. $\text{B}_2\text{O}_3\text{-Bi}_2\text{O}_3$
c. KCl-NaCl	i. $\text{V}_2\text{O}_5\text{-CaO}$
d. NaCl-LiCl	j. NaCl-NaOH
e. $\text{CaO-P}_2\text{O}_5$	k. KCl-KOH
f. $\text{P}_2\text{O}_5\text{-B}_2\text{O}_3$	l. NaCl-MnO_2

TABLE IV. Decontamination with Fe_2O_3 Containing SaltsTest Conditions

Reaction Temperature - 800°C
 Reaction Time - 2 hours
 Salt-to-Metal Ratio - 3:1 (weight)
 Number of Contacts - One

Salt Composition (mole %)			Gamma Energy Analysis, Counts per 30 Min.					
KCl	LiCl	Fe_2O_3		^{60}Co	^{106}Ru	$^{110\text{m}}\text{Ag}$	^{125}Sb	^{144}Ce
85		15	Before	84	654	4250	2640	46,746
			After	2	5	18	34	91
85		15	Before	120	828	9924	6480	68,614
			After	0	2	14	12	42
55	35	10	Before	420	1890	6568	7946	104,324
			After	0	0	12	6	20
55	35	10	Before	318	1436	10,828	8126	75,520
			After	0	2	10	6	12
			After 2nd Contact	0	0	0	1	0

None of the systems gave an overall DF greater than 100 in the first contact stage, and in the second stage the DF's were 10 or less.

In summary, fused salts can be used effectively to decontaminate stainless steel. However, to obtain DF's of 10^6 or greater at least three stages of salt-metal contact are required. Using maximum salt recycle a salt-to-metal weight ratio of at least 3:1 is required for a DF of 10^6 . The contaminated salt would present a serious disposal problem and it is doubtful if fused salt decontamination has any practical application. It might be possible to decontaminate metal with fused salts to the point where it could be released for uncontrolled use, but the cost would make the process uneconomical.

FUSED SLAG-METAL MELTDOWN

The original objective of melting contaminated metal under a fused inorganic slag was to produce a molten metal phase which could be cast into a clean ingot. Ideally, the radioactivity initially associated with the metal would be transferred to the fused slag leaving a metal phase free from activity. Unfortunately, it was impossible to achieve a metal phase free from activity, regardless of the fused slag that was used. In every case a substantial portion of the initial activity associated with the metal specimen remained with the molten metal. The residual activity was found to be dispersed throughout the metal ingot. Even when the metal was remelted several times, using fresh slag for each melt, it was impossible to obtain a clean metal phase. The overall DF obtained with multiple meltings never exceeded about 1000 and was usually much less. Maximum decontamination was achieved when the slag was melted and allowed to react with the solid metal before the metal was melted. Even so, the DF obtained was relatively low.

Examination of the solidified metal ingot, after separation from the slag, showed the sample to be covered with a small amount of contaminated loose material. When the ingot was washed with water, the loose contamination was removed and, after drying, the ingot was free of smearable contamination. The residual contamination dispersed in the metal phase was found to be uniformly distributed throughout the metal and could not be removed except by dissolving the metal. Leaching tests using water, nitric acid and aqua regia indicated that the residual radioactivity was leached from the metal at approximately the same rate as the metal was corroded by the leach solution. When water was the leach solution the metal corrosion rate was extremely slow and the pickup of activity by the leach solution was correspondingly low.

Many slag-crucible combinations were evaluated. It was difficult to find suitable combinations which could be used at 1500°C, the temperature that was required to give good slag-metal separation. Salts which were used in the fused salt decontamination work were far too volatile to be used at 1500°C. Other slag systems were too corrosive and dissolved the metal or could not be adequately contained. Only four slag-crucible-metal systems were found that were sufficiently compatible at 1500°C to be useful in the meltdown tests. They were:

<u>Fused Slag Composition</u>	<u>Crucible</u>	<u>Molten Metal</u>
SiO ₂ -B ₂ O ₃	Al ₂ O ₃	304L SS
CaO-MgO-SiO ₂	Al ₂ O ₃	304L SS
CaO-SiO ₂ -B ₂ O ₃	Al ₂ O ₃	304L SS
CaO-Fe ₂ O ₃	Al ₂ O ₃	304L SS

Other usable slag-crucible combinations could undoubtedly be found, but time did not permit the effort required for their development. (As mentioned

earlier, for a large-scale operation, slag-metal containment would not be a major problem because cold wall containers could be used.)

When the slag and metal were heated directly to 1500°C, and held at that temperature, poor decontamination was achieved regardless of the slag used. Table V shows typical gamma energy analysis data for metal samples before and after melting under various slag compositions. The metal samples were sections of the pump impeller described earlier. In every case the overall DF obtained was low as were the individual DF's. The molten slags dissolved a significant amount of metal and the longer the system was held at 1500°C the more metal was dissolved. Slag systems containing ferric oxide were especially bad in this regard. The DF's obtained, however, did not appear to depend on the amount of metal dissolved. In tests where the slag and metal were held at temperature for periods up to four hours the metal dissolved sometimes exceeded 50%. Analysis of the residual metal indicated the activity level was almost identical, on a weight basis, to metal which had been held at temperature for one hour.

When a metal specimen, which had previously been melted under a slag, was remelted under a fresh slag, little or no decontamination was achieved. Table VI shows gamma energy analysis data for samples which were remelted several times using fresh slag for each melt. The remelting obviously had little effect on the activity content of the metal phase.

Metal decontamination can be improved somewhat if the metal is contacted with the molten slag for 1-2 hours before the metal is melted (see Table VII). This apparently permits the surface layer of metal to dissolve in the slag together with most of the activity. After the metal melts the diffusion of activity back into the metal phase is apparently quite slow.

TABLE V. Gamma Energy Analysis of Metal Samples
Melted Under an Inorganic Slag

Test Conditions

Reaction Temperature - 1500°C

Time at Temperature - 1 hour

Slag-to-Metal Weight Ratio - ~2.5:1

Slag Composition, mole %	Metal Analysis	GEA - Counts per 5 Minutes					Metal Dissolved (Wt%)
		^{60}Co	^{106}Ru	$^{110\text{m}}\text{Ag}$	^{125}Sb	$^{144}\text{Ce-Pr}$	
50 SiO ₂ - 25 MgO- 25 CaO	Initial	430	6,258	2,016	7,652	161,081	5.5
	Final	39	1,924	198	1,727	2,975	
50 SiO ₂ - 25 MgO- 25 CaO	Initial	268	5,157	1,292	5,201	106,700	5.1
	Final	16	926	36	558	1,440	
23 CaO- 73 Fe ₂ O ₃	Initial	316	5,938	2,073	4,792	101,741	19.0
	Final	102	61	174	671	1,048	
60 SiO ₂ - 40 B ₂ O ₃	Initial	403	4,738	4,009	6,776	97,316	3.1
	Final	60	730	224	816	607	
60 SiO ₂ - 40 B ₂ O ₃	Initial	377	6,361	1,955	7,780	109,638	2.9
	Final	23	409	363	309	1,266	
50 SiO ₂ - 25 CaO- 25 B ₂ O ₃	Initial	89	3,281	719	2,878	23,918	5.7
	Final	38	2,834	275	1,654	3,454	
50 SiO ₂ - 25 CaO- 25 B ₂ O ₃	Initial	379	5,962	1,344	7,943	115,615	8.2
	Final	61	888	247	791	791	

TABLE VI. Effect of Remelting on Removal of Radionuclides from Stainless Steel

Test Conditions

Reaction Temperature - 1500°C
 Time at Temperature - 1-1.5 hours
 Slag-to-Metal Ratio - ~2.5:1

Slag Composition (mole %)	Metal Analysis	GEA Counts per 5 Minutes			Metal Dissolved (Wt%)
		¹⁰⁶ Ru	¹²⁵ Sb	¹⁴⁴ Ce-Pr	
60 SiO ₂ - 40 B ₂ O ₃	Initial	1,859	1,380	17,354	
	1st Melt	1,557	895	0	4.0
	2nd Melt	1,454	847	0	1.3
	3rd Melt	1,510	921	0	0.9
	4th Melt	1,396	816	0	1.7
60 SiO ₂ - 40 B ₂ O ₃	Initial	8,158	8,117	150,300	
	1st Melt	290	290	671	2.6
	2nd Melt	138	194	270	3.0
	3rd Melt	154	185	263	1.2
50 SiO ₂ - 25 CaO- 25 MgO	Initial	5,962	7,943	115,615	
	1st Melt	888	791	791	8.2
	2nd Melt	817	452	703	3.7
	3rd Melt	762	512	546	4.1
	4th Melt	597	493	607	2.5

TABLE VII. Effect of Molten Slag-Metal Contact
Prior to Metal Meltdown on Metal
Decontamination

Test Conditions

Temperature - One hour at 1300°C (slag melted)
Then one hour at 1500°C

Slag-to-Metal Ratio - ~2.5:1

Slag Composition (mole %)	Metal Analysis	GEA - Counts/5 Minutes			Metal Dissolved (Wt%)
		^{106}Ru	^{125}Sb	$^{144}\text{Ce-Pr}$	
50 SiO ₂ - 25 CaO- 25 MgO	Initial	5,374	3,967	87,412	
	Final	62	31	102	7.8
60 SiO ₂ - 40 B ₂ O ₃	Initial	5,901	5,796	101,312	
	Final	116	58	179	5.3
50 SiO ₂ - 25 CaO- 25 B ₂ O ₃	Initial	6,327	5,783	123,450	
	Final	71	103	116	9.7
50 SiO ₂ - 25 CaO- 25 B ₂ O ₃	Initial	4,773	52,617	28,312	
	1st Melt	102	76	31	9.1
	2nd Melt	109	53	52	3.5

Using this technique overall DF's approaching 1000 can be obtained. However, additional decontamination by remelting is not possible.

Leaching tests were run on a number of metal samples after meltdown. Leach solutions used included distilled water, 2M HNO_3 and aqua regia. The metal was corroded at a fairly rapid rate when aqua regia was used. Analysis of the aqua regia solution for radioactivity and weight measurements of the metal samples showed that the pickup of activity by the leach solution corresponded, within experimental error, to the rate of corrosion of the metal.

When water was used as the leach solution the corrosion rate of the metal was very slow. Pickup of activity by the water was correspondingly low as can be seen by referring to Figure 3. Activity in the leach solution increased rapidly for the first 24-48 hours, and remained relatively constant thereafter. The data are scattered, however, and it is difficult to tell if there was a gradual increase in activity over long time periods. When nitric acid was used as the leach solution activity pickup was somewhat more rapid for the first 48 hours and appeared to increase gradually with time thereafter (Figure 4).

In summary, it was not possible to produce a clean metal phase by melting the metal under a fused slag. The solidified metal always contained substantial amounts of radioactivity. The contamination was dispersed throughout the metal phase, and after a water wash of the sample, the contained activity was nonsmearable and essentially nonleachable.

Meltdown consolidates the metal into a minimum volume and thus reduces the storage space required. The fact that the contamination is nonleachable also simplifies the disposal problem. However, the process does

FIGURE 3 LEACHING OF STAINLESS STEEL IN WATER, AFTER MELTDOWN UNDER A FUSED SLAG

FIGURE 4 LEACHING OF STAINLESS STEEL IN 2 M HNO_3 , AFTER MELTDOWN

generate a fused slag which is contaminated with radioactivity, and which will present a disposal problem. The limited work done indicates the weight of slag produced would probably about equal the weight of metal processed, assuming maximum use of the slag. The overall result of the process would be an increase in disposal costs.

METAL MELTDOWN

The results of the fused slag-metal meltdown tests suggested the possibility of simplifying contaminated metal disposal by melting the metal and casting it into ingots. The metal volume would be reduced to a minimum and the contained activity might be nonleachable. Subsequent experiments showed that when stainless steel was melted, under an inert atmosphere, the bulk of the radioactivity associated with the metal was dispersed throughout the molten metal. When the metal solidified the radioactivity was fixed in the metal phase and was essentially nonsmearable and nonleachable. Experimental results show that 20-35% of the activity was lost during melting by vaporization or transfer to the alumina crucible. The remaining activity was retained in the metal ingot. Controlled dissolution of the solidified metal in aqua regia indicated the residual radioactivity was uniformly dispersed in the metal phase. When the metal ingot was removed from the alumina crucible there was a small amount of loose contaminated material present. A water or acid wash removed the loose material, however, and the washed ingot was then free of smearable contamination. When the washed ingot was leached in water or acid the uptake of contamination by the leach solution was very slow and corresponded to about the rate at which the metal corroded. The leaching data confirmed the results obtained using metal which had been melted under a slag.

Typical results from a metal meltdown test using stainless steel are shown in Table VIII. The sample was washed three times with 2M nitric acid prior to meltdown and each wash solution was analyzed by GEA. After meltdown the sample was washed with water, then washed twice in 2M HNO_3 , and partially dissolved in aqua regia. Prior to meltdown a substantial amount of radioactivity was removed from the sample by the HNO_3 washes. After meltdown, however, very little radioactivity could be removed from the sample by washing with nitric acid. Only by dissolving the metal was it possible to transfer a significant amount of activity to the acid solution.

Leaching tests were run on a large number of metal samples after meltdown using both water and nitric acid as the leach solution. The leach tests lasted for up to ninety days. Typical water leaching data are shown in Figures 5 and 6. In every case the pickup of activity by the leach solution was very slow. In most cases the activity in the solution reached a maximum in about 24-48 hours and remained relatively constant thereafter. If a solidified sample, which had been previously leached in water, was placed in fresh leach solution the pickup of activity by the new solution was much less than by the original leach solution.

All of the results obtained indicate that the radioactivity is leached from the solidified metal specimens at approximately the same rate as the metal is corroded by the leach solution. When water is the leach solution and stainless steel is the metal phase the corrosion rate is extremely low, and the leaching of activity from the metal is correspondingly low.

Measurements on the solidified metal samples indicate the bulk density of the samples approaches the theoretical density of stainless steel ($\sim 8.0 \text{ g/cm}^3$). Therefore, meltdown serves to consolidate the contaminated

TABLE VIII. Typical Results from Metal Meltdown
Experiments Using Stainless Steel

Step	Analysis Of:	Metal Dissolved	GEA Counts/5 Min.	
			^{106}Ru	^{125}Sb
1	Original metal sample		14,448	130,878
2	1 st HNO_3 wash solution	None	2,111	1,199
3	Metal after 1 st HNO_3 wash		10,251	121,194
4	2 nd HNO_3 wash solution	None	279	3,283
5	Metal after 2 nd HNO_3 wash		8,989	122,435
6	3 rd HNO_3 wash solution	None	148	1,702
7	Metal after 3 rd HNO_3 wash		8,578	115,606
8	Metal after melting at 1500°C		7,799	87,304
9	Water wash after melting		110	246
10	1 st HNO_3 wash after melting	None	2	13
11	Metal after HNO_3 wash		8,004	87,208
12	2 nd HNO_3 wash after melting	None	1	3
13	Metal after HNO_3 wash		7,995	87,934
14	1 st aqua regia leach solution	4 wt%	244	1,549
15	Metal after aqua regia leach		7,558	85,404
16	2 nd aqua regia leach solution	30 wt%	1,017	7,136
17	Metal after 2 nd aqua regia leach		4,738	52,416

FIGURE 5 LEACHING RATE FOR BETA-GAMMA CONTAMINATED STAINLESS STEEL IN WATER, AFTER METAL MELTDOWN

FIGURE 6 LEACHING RATE FOR ALPHA-BETA-GAMMA CONTAMINATED STAINLESS STEEL IN WATER, AFTER METAL MELTDOWN

metal into a minimum volume, in addition to fixing the activity in the metal phase. This reduction in metal volume reduces the ultimate storage space required for metal disposal.

In summary, when contaminated stainless steel is melted under an inert atmosphere the bulk of the activity is dispersed in the metal phase. When the metal solidifies the activity is fixed in the metal and is nonleachable. The metal bulk volume is reduced to a minimum and ultimate disposal is greatly simplified.

CONCLUSIONS

Based on the results of the laboratory tests it is possible to draw a number of conclusions regarding the technical and economic potential of the three processes which were studied. Additional testing on a pilot scale would permit a more thorough process evaluation but would probably not change the conclusions which were reached and which are discussed below.

The fused salt decontamination process does not appear to be practical for the large-scale decontamination of metal equipment. Using an oxidizing fused salt, such as sodium nitrate, and several stages of salt-metal contact it is possible to obtain decontamination factors (DF's) of greater than 10^6 from beta-gamma and alpha activities. However, the process generates substantial volumes of contaminated salt waste, which in itself represents a serious disposal problem. Even using maximum salt recycle, the salt-to-metal ratio required to give a nominal DF of 10^6 would be at least 3:1 on a weight basis. In addition, effective decontamination requires the dissolution of a substantial portion of the metal phase in the fused salt. The cost of disposing of the contaminated salt would probably approach or exceed the cost of disposal of the contaminated metal.

The meltdown of contaminated metal under a fused inorganic slag failed to produce a clean metal phase. Only a portion of the activity was transferred to the slag phase, with much of the activity remaining in the metal phase. Even repeated melting of a metal specimen, using fresh slag for each meltdown, failed to reduce the activity in the metal phase by a factor of more than about 1000. Analysis of solidified metal ingots showed the residual activity was dispersed and fixed in the metal phase. After an initial water wash, the residual activity in the metal phase was essentially nonsmearable and nonleachable. Extended leaching tests with water and mineral acids showed that the pickup of activity by the leach solution corresponded approximately to the corrosion rate of the metal. The fused slag meltdown process serves to simplify the disposal of metal equipment since the metal is effectively consolidated and the activity in the metal is nonleachable. However, the process has the distinct disadvantage of generating a substantial volume of contaminated slag which must be disposed of. Depending on the degree of metal decontamination desired, the amount of contaminated slag generated could approach or exceed the metal processed on a weight basis.

The meltdown of contaminated metal under an inert atmosphere produces a metal ingot in which the radioactivity is dispersed and effectively fixed in the metal phase. In the process the bulk volume of the metal is reduced to an absolute minimum. A large fraction of the radioactivity originally present is fixed in the metal phase in nonleachable form. As a result, final storage of the consolidated metal is greatly simplified. The radioactivity which volatilizes during meltdown represents a secondary handling problem, but this could be taken care of through the use of absolute filters in the off-gas lines. The retort used to melt the metal would absorb some

radioactivity which would build up with time. Disposal of items such as the retort liner (refractory brick?) would present an additional disposal problem.

Of the three processes evaluated, metal meltdown under an inert atmosphere seems to offer the most practical method of disposing of contaminated metal equipment. However, additional work is needed to fully evaluate the potential value of the meltdown process. A pilot scale test, using several kilograms of metal, is needed to demonstrate that the radionuclides can be fixed in the metal phase on a large scale. Additional factors to be considered, which will require additional testing, include: the meltdown of metals other than stainless steels and the effects of nonmetallics, which may be present with the metal, on the meltdown process.

FUTURE WORK

Laboratory-scale tests have shown that the metal meltdown process offers a simple and effective method for disposing of contaminated metal equipment. However, additional pilot-scale tests are required to demonstrate its practical application.

The laboratory melts were limited to stainless steel samples weighing up to 50 grams. In the tests it was demonstrated that the bulk of the radioactivity ($\geq 65\%$) was retained in the metal phase in nonleachable and nonsmearable form. Pilot-scale tests using several kilograms of metal are needed to demonstrate that similar results can be achieved on a practical scale. It is possible that large-scale tests would show a greater amount of radioactivity is retained in the metal phase than the laboratory-scale tests showed due to the reduced surface-to-volume ratio of the molten metal in the large-scale tests.

Several other factors must also be considered which will require experimental evaluation. All of the meltdown tests were carried out using stainless steel. Since it may be impractical to segregate metals according to composition, tests should be run to determine if a variety of metals can be melted and cast concurrently without adversely affecting the process. Low-melting metals such as lead, magnesium, aluminum, copper, etc., could represent a serious problem. High-melting metals such as zirconium, titanium, tantalum, etc., could also present a severe operating problem. Tests would be necessary to determine the extent of alloying that could be achieved at reasonable operating temperatures with mixtures of metals. The bulk of the tests could be carried out using nonradioactive materials which would reduce the cost of the tests substantially.

The presence of organic or inorganic materials such as insulation, gaskets, etc., with the metals could present severe processing problems. Segregation of the nonmetallics is possible but could be quite expensive. Determining the effects of these materials on meltdown on a laboratory scale would be extremely difficult. Pilot-scale tests would be necessary to fully evaluate the effects of nonmetallics. The bulk of the tests could be carried out using noncontaminated materials. Both inorganic and organic materials would be tested. Tests with contaminated materials would be limited to the minimum required to verify the results obtained with noncontaminated materials.

For the pilot-scale tests the melter should have the capability of handling at least 10 kilograms of metal. An induction-heated unit would probably be the most economical melter to use for the tests.

ACKNOWLEDGEMENTS

Most of the experimental work reported was performed by R. J. Elovich and A. D. Peoples. M. J. Szulinski and T. R. McKenzie of ARHCO provided information and technical guidance and assisted in obtaining needed metal samples.

DISTRIBUTION

U.S. Atomic Energy Commission, Washington Division
of Production

F. P. Baranowski

U.S. Atomic Energy Commission, Richland
Operations Office

O. J. Elgert
P. G. Holsted
W. E. Lotz
T. A. Nemzek

Atlantic Richfield Hanford Company

G. E. Backman	L. M. Knights
G. E. Benedict	C. W. Malody
G. L. Borsheim	T. R. McKenzie
D. E. Braden	B. J. McMurray
D. J. Brown	G. A. Nicholson
L. E. Bruns	G. C. Oberg
M. H. Campbell	L. M. Richards
R. P. Corlew	R. E. Roberts
J. Faulhaber	H. P. Shaw
H. H. Hopkins	H. E. Smith
W. P. Ingalls	P. W. Smith
R. E. Isaacson	M. J. Szulinski
R. D. Jensen	J. H. Warren

Battelle-Northwest

J. W. Bartlett
R. E. Burns
H. T. Fullam (10)
V. L. Hammond
R. E. Nightingale
J. A. Partridge
H. H. Van Tuyl
Technical Information Files (2)