ORNL
MASTER COPY

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

ORNL-TM-263 (Rev. 1)
COPY NO. - 31
DATE - Jan. 14, 1963

THE USE OF PHOSPHITE AND HYPOPHOSPHITE TO FIX RUTHENIUM FROM HIGH-ACTIVITY WASTES IN SOLID MEDIA

Herschel W. Godbee
Walter E. Clark

To be submitted to Industrial and Engineering Chemistry for publication.

NOTICE

This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report. The information is not to be abstracted, reprinted or otherwise given public dissemination without the approval of the ORNL patent branch, Legal and Information Control Department.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

THE USE OF PHOSPHITE AND HYPOPHOSPHITE TO FIX RUTHENIUM FROM HIGH-ACTIVITY WASTES IN SOLID MEDIA

Herschel W. Godbee
Walter E. Clark

Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee

BRIEF

Fission product ruthenium, normally volatile to the extent of 20 to 60% in evaporation and calcination of simulated high level radioactive wastes to 500-1000°C., can be 99.9% retained in the solid product by addition of 2 moles of phosphite or hypophosphite per liter of waste. As little as 0.1 mole per liter lowered the ruthenium volatility during distillation by factors which varied from 38 for Darex (stainless steel) to 227 for TBP-25 (aluminum-uranium alloy) waste solutions. The final products with Purex (Al-clad natural uranium), TBP-25, and Darex wastes were insoluble glassy solids with densities of 2.4 to 3.8 grams per ml. and represented volume reductions of 2.9 to 8. These volume reductions are essentially the same as those obtained when the waste is calcined without additives.

INTRODUCTION

A process has been developed for converting high-activity wastes to thermally stable solids by evaporation, calcination and melting of the calcined solids. Phosphite or hypophosphite is added to the waste to prevent volatilization of fission product ruthenium and to aid in forming a glass.

Fixation of high activity radioactive wastes in thermally stable, solid, insoluble form represents one of the safest approaches to "ultimate" disposal. Retention of all activity in the solid form is most desirable but is particularly difficult for ruthenium[19]. The latter is highly volatile from the common nitrate wastes under the conditions necessary to produce a vitreous or microcrystalline material with high mechanical strength and thermal conductivity. Volatilization of 100% of the ruthenium appears, however, to be impracticable[4,5].

Phosphite and hypophosphite are excellent agents for fixing ruthenium in stable solid media. Both are powerful reducing agents and are oxidized to phosphate, a glass-former. Calcination-fixation of three types of simulated waste solutions generated from the solvent extraction reprocessing of nuclear reactor fuel elements has been carried out in the laboratory with added phosphite or hypophosphite, both in bench scale batch and semi-continuous experiments, and in larger scale semi-continuous experiments[13,6,7,8,9]. Semi-continuous calcination of all three types without added phosphite or hypophosphite has been demonstrated on both laboratory and unit operations scales[13,15,6,7,8]. Typical compositions of these three waste types are listed in Table 1. For convenience and brevity these compositions are referred to as (1) Purex, a nitric acid waste from aluminum-jacketed uranium fuel elements; (2) TBP-25

an aluminum nitrate-nitric acid waste from enriched uranium-aluminum alloy fuel elements; and (3) Darex, stainless steel nitrates from stainless steel-constituted fuel elements.

In a process developed at Chalk River, Canada[1] for incorporating fission products in glass at about 1350°C., half the ruthenium is volatilized into the off-gas and collected on a bed of siliceous firebrick coated with iron oxide. The loaded bed is melted into a subsequent batch of glass. In a process under development at Harwell, England[11] for incorporating fission products in glass at about 1100°C., the volatile ruthenium is adsorbed on silica gel or ferric oxide beds, and these beds are melted into a subsequent batch of glass. In a process now being developed at the Idaho Chemical Processing Plant[17] for reducing waste solutions to granular solids at about 500°C., 90-99% of the ruthenium may be volatilized and adsorbed on silica gel beds, and the beds regenerated with water. Control of ruthenium volatility would simplify or eliminate recycle systems and side streams to be disposed of, and minimize contamination of process equipment.

No data exist on ruthenium behavior during high temperature (850-1050°C.) calcination of actual high-activity wastes, but nitrite, which is formed from nitrate by radiolysis, suppresses ruthenium volatility somewhat during evaporation of these solutions, as do other mild reducing agents, e.g. nitrogen dioxide and a mixture of tributyl phosphate and hydrocarbon diluent[23], nitric oxide[14,21] and dibutyl and monobutyl phosphoric acids[14]. Since ruthenium volatility is a function of the oxidizing power of the solution, dilution with water and steam stripping of the nitric acid will decrease ruthenium in the off-gas below that obtained by simple distillation.

DESCRIPTION OF POT CALCINATION PROCESS

Waste is fed into an evaporator, where it is combined with nitric acid and water recycled from the pot calciner and rectifier, respectively, to maintain the nitrate and acidity at a level to minimize ruthenium volatility (Figure 1). The evaporator bottoms go to a hold tank where phosphite and hypophosphite are added to control ruthenium volatility and to serve as a source of phosphate for forming glass. Other glass-forming additives (e.g. PbO, NaOH, borax) are also added to the hold tank. The off-gas from the hold tank, mainly nitric oxide (NO) from the nitrate-phosphite-hypophosphite reaction, may be stored for subsequent use in controlling ruthenium volatility in the evaporator or oxidized to NO_2 by addition of oxygen and absorbed in a scrubber downstream from the rectifier. The mixture from the hold tank is pumped to a heated pot where it is evaporated to dryness, calcined, and melted at 850-1050°C. The stainless steel pot would be sealed after filling, and would serve as both the shipping and storage vessel. The recycle of off-gas from the fixation pot will result in a build-up of ruthenium in the evaporator unless the greater part of it can be retained in the solid product.

EXPERIMENTAL

Ruthenium volatility from simulated Purex, TBP-25, and Darex waste solutions (Table 1) was studied during evaporation of the solutions, calcination of the dried solids, and incorporation of the calcined solids in melts with selected additives. The effect of phosphite (HPO_3^{-2}) and hypophosphite ($H_2PO_2^-$) on ruthenium volatility from these waste solutions was investigated both with and without the addition of glass-forming ingredients. All waste solutions studied contained 0.2 mg. per ml. stable

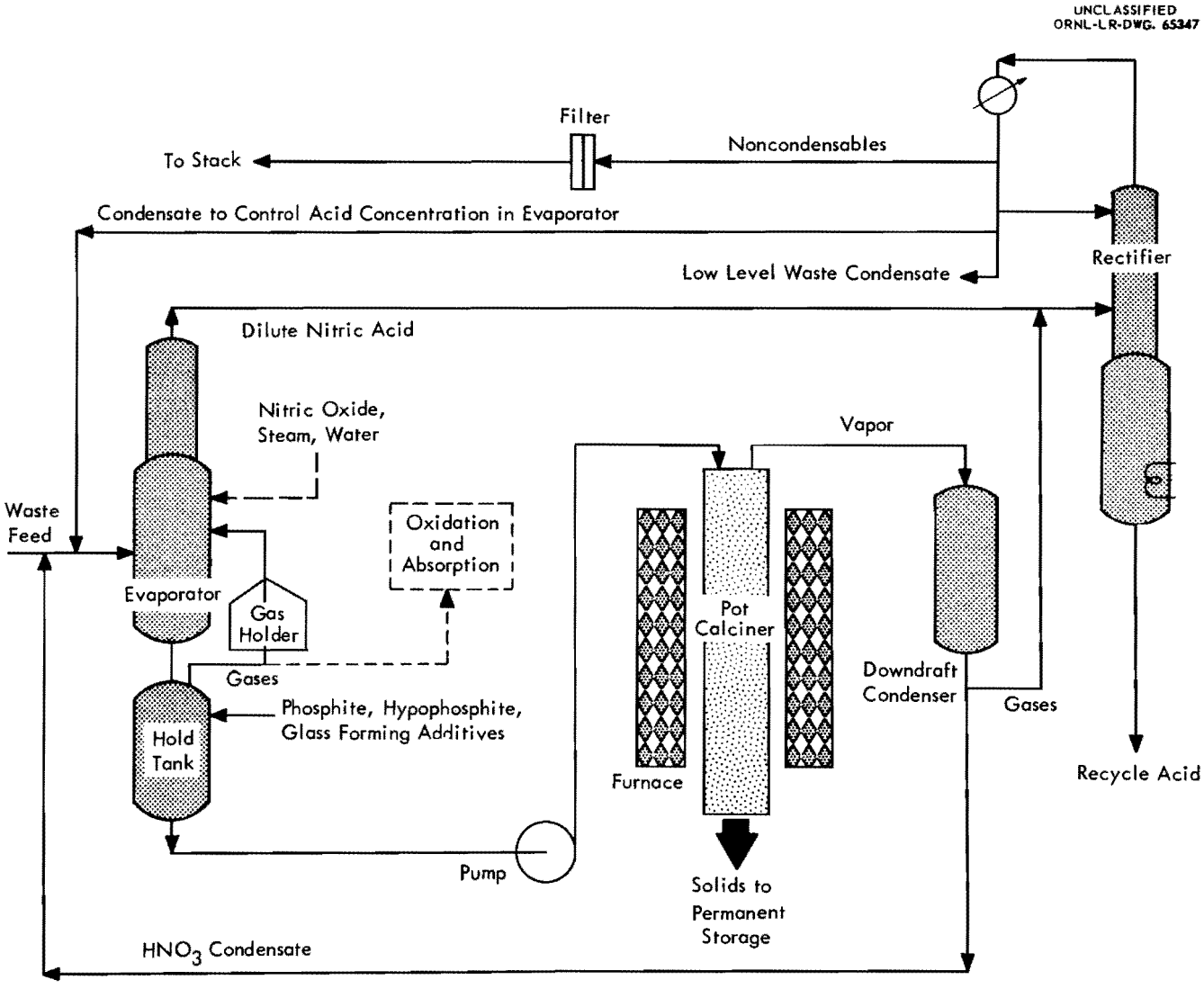

Fig. 1. Pot calcination of high activity wastes. Dashed lines indicate optional operations.

Table 1. Composition of Simulated High-Activity Fuel Processing Waste Solutions Used in Ruthenium Volatility Studies*

Component	Darex (24 gal./kg. U-235 processed)	Purex (40 gal./ton U processed)	TBP-25 (106 gal./kg U-235 processed)
Al^{+3}		0.1 M	1.72 M
Fe^{+3}	1.25 M	0.5 M	0.16 mg./ml.
Cr^{+3}	0.38 M	0.01 M	
Hg^{+2}			4.02 mg./ml.
Ni^{+2}	0.18 M	0.01 M	
Mn^{+2}	0.04 M		
Na^{+}		0.6 M	2.4 mg./ml.
NH_4^{+}			0.05 M
H^{+}	0.75 M	5.6 M	1.26 M
Ru	0.2 mg./ml.	0.2 mg./ml.	0.2 mg./ml.
Cl^{-}**	260 p.p.m.	165 p.p.m.	160 p.p.m.
NO_3^{-}	6.0 M	6.1 M	6.6 M
SO_4^{-2}		1.0 M	
Density, g./ml.	1.33	1.30	1.32

*These compositions are representative of waste compositions which may be expected, although subject to change as improvements in separations technology continue to be made. A brief description of each process follows:

Purex Process: Tributyl phosphate solvent in an inert diluent is contacted with a solution of uranium and fission products in nitric acid. This process is used for the processing of irradiated natural uranium. By far the largest volume of waste now being produced is from the Purex process.

TBP-25 Process: This process is designed to recover enriched uranium from irradiated uranium-aluminum alloy fuel elements. The fuel elements are dissolved in nitric acid using a mercuric nitrate catalyst to give a solution of aluminum nitrate and uranyl nitrate. The extraction solvent is tributyl phosphate in an inert diluent. The waste is primarily aluminum nitrate plus fission products.

Darex Process: Dilute aqua regia is used to dissolve stainless steel-constituted fuels. After removal of chloride ion down to about 100 p.p.m. by nitric acid stripping, the uranium (and plutonium, if present) is recovered by tributyl phosphate extraction.

**Except for Darex waste, which normally contains ~100 p.p.m. Cl^{-}, all Cl^{-} present was added with Ru as $RuCl_3$.

ruthenium and 0.1 microcurie per ml. Ru-106 tracer. This concentration is typical of ruthenium to be expected in a Purex waste solution for a low-burnup reactor and is about ten times that to be expected in a typical TBP-25 waste. Stable and radioactive ruthenium were added as the chlorides. Phosphite was added as phosphorous acid, H_3PO_3. Hypophosphite was usually added as sodium hypophosphite, NaH_2PO_2, and more rarely as hypophosphorous acid, H_3PO_2.

Experiments were carried to about 500°C. when using a 200-ml. borosilicate glass flask as a pot and to about 1000°C. in a quartz outer pot holding either a 200-ml. quartz or stainless steel beaker (Figure 2). Condensates were analyzed for Ru-106 in a gamma scintillation counter. The equipment was decontaminated between experiments by boiling 15 M HNO_3 through it. The residues from the low-temperature experiments were analyzed by gamma counting after dissolution in refluxing 12 M HCl. Residues from the high-temperature experiments were not dissolved in most cases since the dissolution required about a week in refluxing HCl, and the additional information that would have been obtained by closing the material balance did not seem to justify the extra time and effort required.

The volatility of ruthenium from nitric acid solutions during evaporation with and without phosphorous acid (H_3PO_3) addition was investigated by distillation in a Gillespie equilibrium still[12]. Distillation was continued to collection of only one receiver volume (7% of original solution volume) because of the tendency of ruthenium to plate out in the condenser. Consequently the ruthenium volatilities do not represent exact equilibrium conditions. In practice if the condenser was air-cooled, no visible ruthenium deposit was obtained unless the concentration of the acid in the distillate

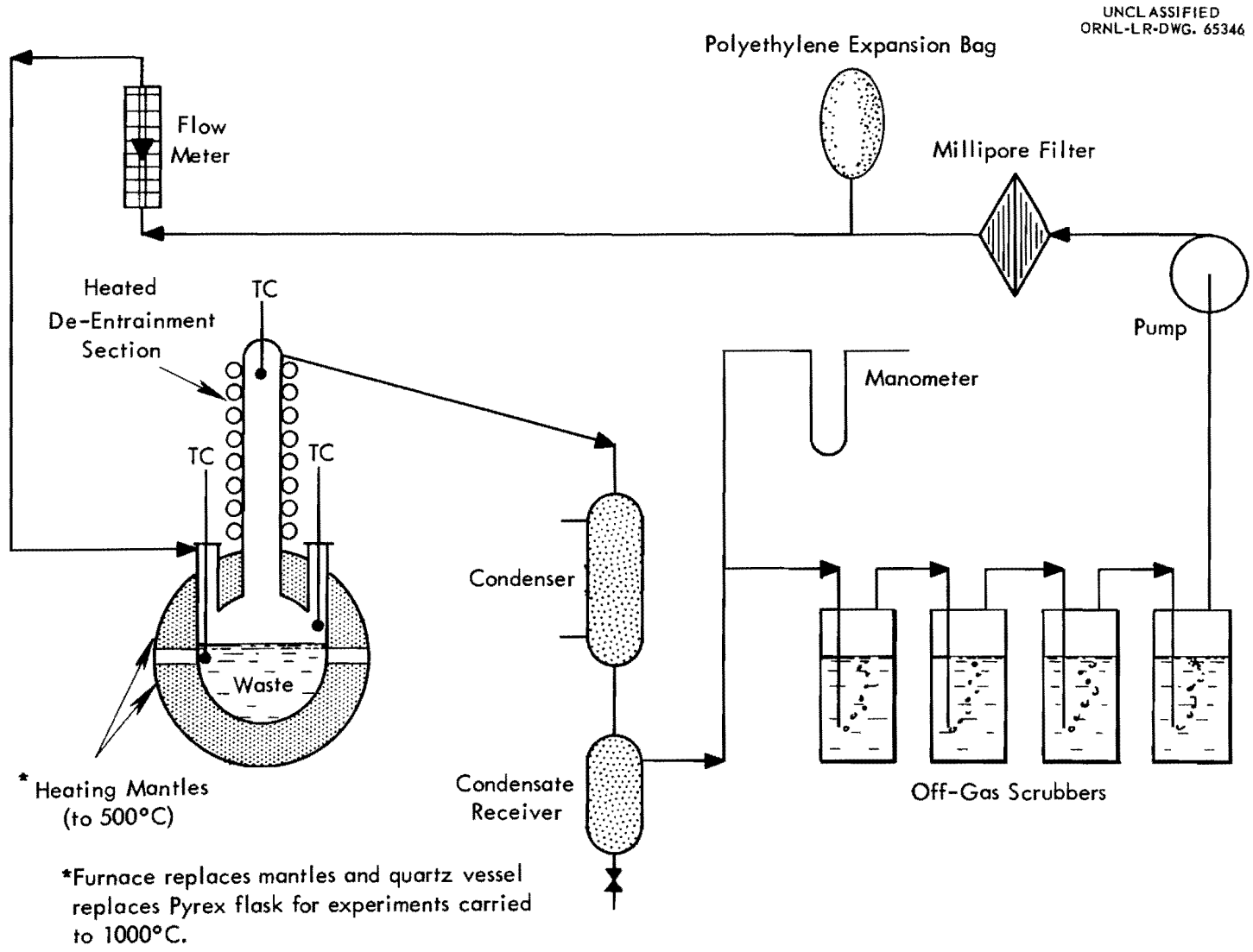

Fig. 2. Apparatus for studying ruthenium volatility during batch evaporation-calcination.

was greater than 8 M. Tracer ruthenium was added as the trichloride, $RuCl_3$. In order to obtain quantitatively reproducible results it was found necessary to distil the solution within a few hours after the addition had been made. On longer aging the relative volatility of the ruthenium decreased. This fact, plus comparison with data obtained with actual waste solutions[23,21,18] leads to the conclusion that the ruthenium volatilities observed in this work represent the maximum to be expected from solutions of this type. The effect of reflux time on ruthenium volatility was not investigated.

A. Calcination-Fixation Experiments

In a series of calcination experiments to 500°C. with Purex waste, ruthenium in the condensate decreased from 28% to 0.03% or less of that originally present in the waste when the phosphorous acid concentration was increased from 0 to 1.5 M (Figure 3). The results of similar series with Darex and TBP-25 wastes were essentially the same. The agreement between gamma counting of Ru-106 and chemical and neutron activation analyses for stable ruthenium indicated that the radioactive and stable ruthenium behaved similarly. When phosphoric acid, H_3PO_4, was added instead of phosphorous acid to give concentrations of 0.75 to 1.5 M, ruthenium volatility from Purex waste was the same as from the waste with no additive. The percentage of original ruthenium in the condensate was not increased by increasing the ruthenium in the feed from 0.2 mg. per ml. to 20 mg. per ml. in the presence of 1.5 M HPO_3^{-2}.

In a series of experiments in which TBP-25 waste was calcined at 1000°C. in a quartz container, ruthenium in the condensate decreased from 60% to 0.05% of the original when the phosphorous acid concentration was increased from 0 to 2.25 M.

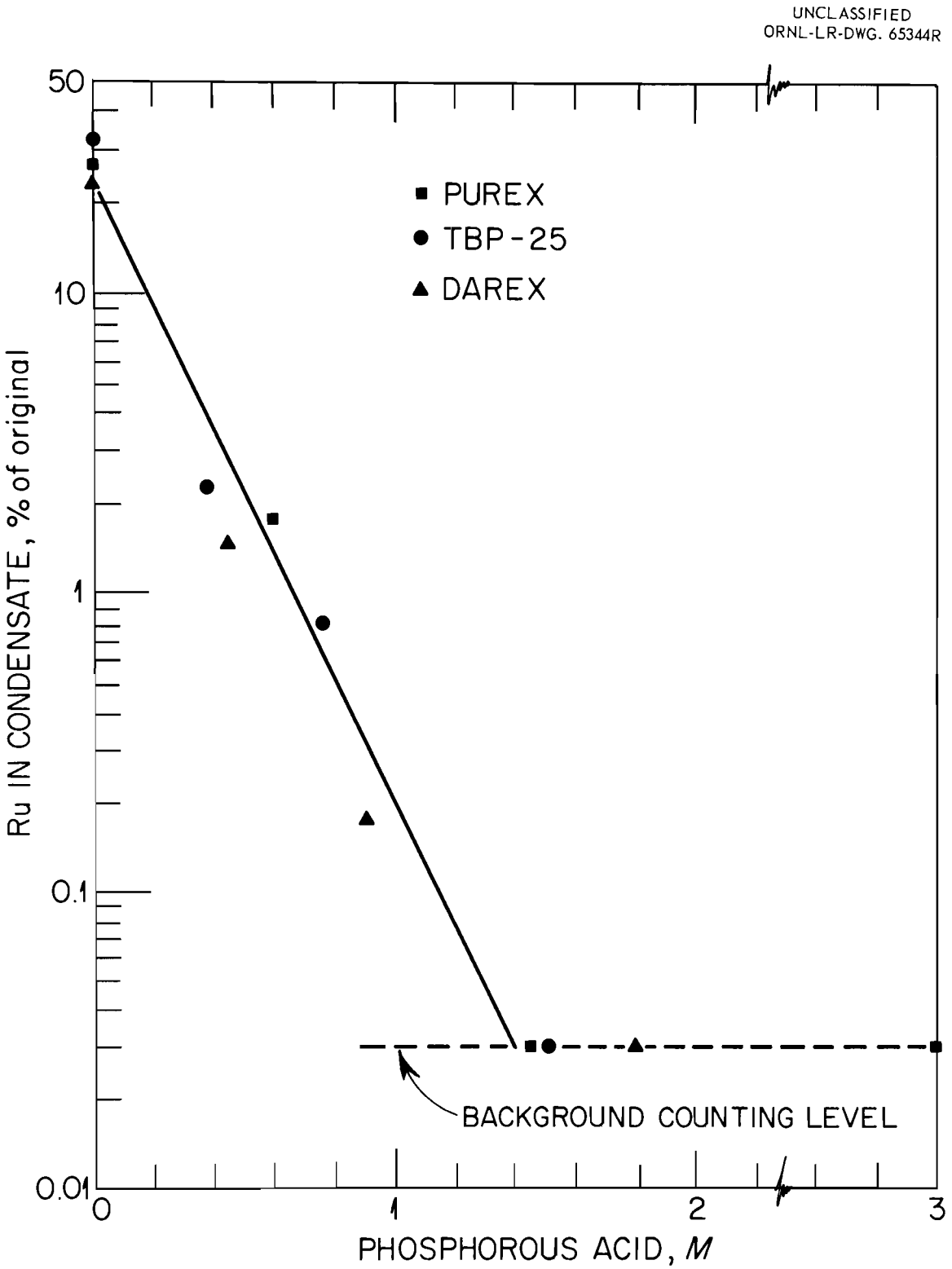

Fig. 3. Effect of phosphorous acid on ruthenium volatility from Purex, Darex, and TBP-25 waste solutions on batch calcination to 500°C. Solutions contained 0.2 mg. Ru per ml. and 0.1 microcurie Ru-106 per ml.

With a stainless steel container the percentages in the condensate were factors of 1.5 to 6.5 less (Figure 4); the difference undoubtedly represents ruthenium that adhered to stainless steel but not to quartz.

Phosphite or hypophosphite not only controls ruthenium volatility but serves as a source of phosphate in forming glasses incorporating waste oxides. A series of glassy products was prepared with Purex waste containing added phosphorous acid, sodium tetraborate, sodium hydroxide, and calcium or magnesium oxide. These had melting points in the range 825° to 925°C. and densities in the range 2.6 to 2.8 grams per ml., incorporated 35 to 45 weight % oxides from waste, and represented waste volume reduction factors of 5.5 to 7.7. There was little if any increase in the volume of the solid product as a result of phosphite addition. One of the more satisfactory products (Figure 5) contained 29, 21.8, 21.5, 10.7, 8.7, and 6.4 weight %, respectively, of P_2O_5, Na_2O, SO_3, Fe_2O_3, MgO, and B_2O_3. The products with Purex waste were glassy only when cooled quickly; when cooled slowly they were rocklike solids. Ruthenium in the condensate from a calcination experiment similar to the one above (i.e. Purex waste to 910°C.) but in stainless steel equipment, was 0.03% or less of the original. Sulfate in the condensate in the experiments with Purex waste varied from about 2% to 12% of the original depending on the additives. Since sulfuric acid in the condensate could cause an additional corrosion problem and would build up in acid recycled to the evaporator (Figure 1), it should be retained in the pot as a stable salt.

Glassy products prepared from TBP-25 waste, sodium hypophosphite, and lead oxide or sodium tetraborate, on heating to 850-1000°C. and cooling by quenching or annealing, had densities ranging from 2.4 to 2.8 grams per ml., contained from 26 to

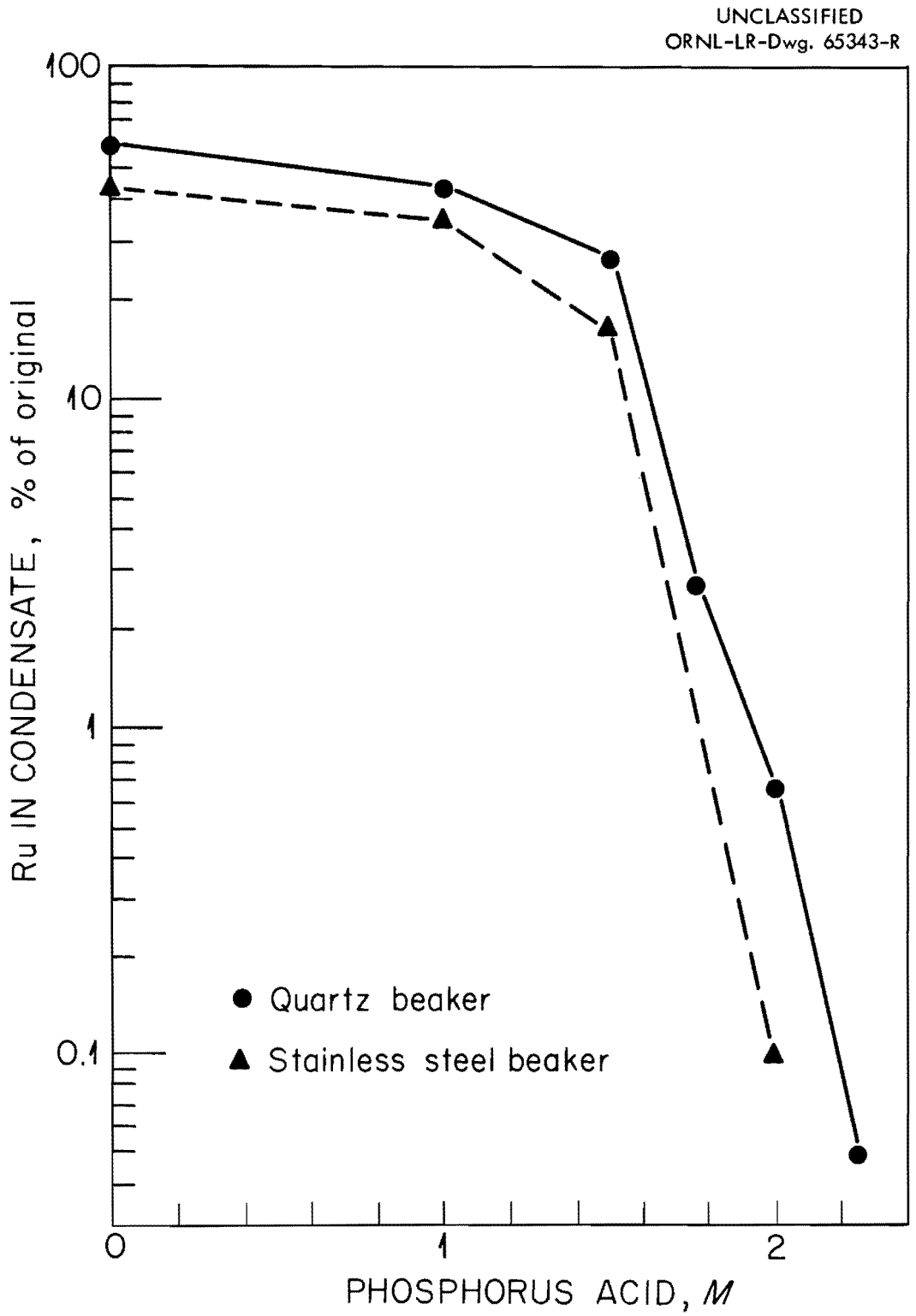

Fig. 4. Phosphorous acid, 2-2.3 M, reduced the volatility of ruthenium to 0.1% or less in batch calcination of TBP-25 waste to 1000°C. Solution contained 0.2 mg. Ru per ml. and 0.1 microcurie Ru-106 per ml.

35 weight % oxides from waste, and represented volume reduction factors of 7.2 to 9.3. Essentially all the mercury was volatilized during calcination and fusion. One of the more satisfactory mixtures from considerations of behavior during heating, softening point, fluidity of melt, and quality of final product, gave a glass with 40.5, 25, 18.6, and 15.9 weight %, respectively, of P_2O_5, Al_2O_3, Na_2O, and PbO (Figure 6). Leaching (dissolution) rates for this glass in distilled water, determined by the leaching of Cs-137 tracer, decreased from 2.1×10^{-6} to 2.5×10^{-7} grams cm^{-2} day^{-1} at the end of the first and fifth weeks, respectively. Ruthenium in the condensate from preparation of this glass was 0.68% of that originally present in the waste.

Glassy products were prepared from Darex waste and sodium hypophosphite, plus aluminum and/or borax. These had softening points which varied between 750° and 1000°C. and densities between 2.7 and 3.8 grams per ml., contained 13.4 to 32.3 weight % waste oxides, and represented volume reductions of 2.9 to 6.6. One of the more satisfactory products contained 37.3, 22.9, 19.7, 10.5, 4.8, 3.0, and 1.4 weight %, respectively, of P_2O_5, Al_2O_3, Na_2O, Fe_2O_3, B_2O_3, Cr_2O_3, and NiO.

The phosphate and boro-phosphate glasses and ceramic products formed from fixation of these simulated wastes were impervious to and relatively insoluble in water as determined by leaching tests on specimens which contained Cs-137. Solubility data are reported elsewhere[10]. Solids produced by calcination with no additives were invariably quite porous.

B. Evaporation Experiments

Ruthenium volatilization from boiling nitric acid solutions containing 0.002 M Ru was such that the log of the distillation factor, $\frac{C_{Ru \text{ in distillate}}}{C_{Ru \text{ in solution}}}$, was a linear function

UNCLASSIFIED
PHOTO 56375

Fig. 5. Purex waste oxides incorporated in phosphate-borate glass. Waste made 1.5 M in H_3PO_3, 0.8 M in MgO, 0.17 M in $Na_2B_4O_7$, and 1.7 M in NaOH, evaporated to dryness and heated to 850°C. Density 2.7 g/ml; product contained 39 wt % waste oxides; volume reduction factor, 5.5.

UNCLASSIFIED
PHOTO 55772

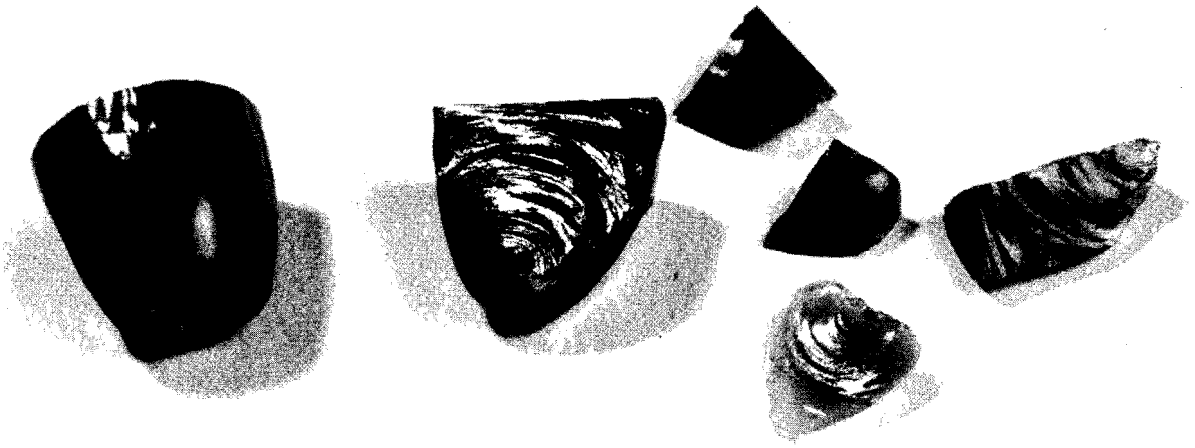

Fig. 6. TBP-25 waste oxides incorporated in phosphate-lead glass. Waste made 2 M in NaH_2PO_2 and 0.25 M in PbO, evaporated to dryness and heated to 1000°C. Density 2.8 g/ml; 26 wt % waste oxides; volume reduction factor, 8.

of the log of the concentration of nitric acid in the distillate until the latter reached a concentration of about 8 M. At higher acidities (corresponding to 10 M HNO_3 in the still pot) the relative volatility of the ruthenium increased sharply (Figure 7). This effect is generally consistent with observations made elsewhere[23]. Ruthenium volatility is known to be a function of the species present in solution, particularly of the oxidation state. These experiments are not necessarily directly applicable to actual waste solutions in which the ruthenium species are unknown. They do show that under carefully controlled conditions ruthenium volatilizes in a predictable manner. Apparently a different volatilization mechanism is involved at higher acid concentrations. The distillate from 12 M HNO_3 formed in two layers. The concentration of ruthenium in the upper layer (orange) was six times that in the lower (brown) (8×10^{-4} vs 4.9×10^{-3} M). The layers were miscible on stirring. A black deposit, presumably RuO_2, was noted in the cooler parts of the condenser.

Boiling HNO_3 solutions, 1.7 M in $Al(NO_3)_3$, behaved like pure HNO_3 solutions as long as the free HNO_3 concentration in the distillate was less than 0.1 M. At higher distillate acid concentrations the ruthenium volatility increased rapidly (Figure 7), apparently because of the higher boiling temperatures of the salt solutions. When the boiling points were lowered by reducing the pressure from 748 to 570 mm. of Hg, the ruthenium volatilities approximated those from HNO_3 alone (Figure 7).

The effectiveness of relatively small concentrations of phosphite in suppressing ruthenium volatility is shown by the fact that 0.1 M H_3PO_3 added to nitric acid and synthetic waste solutions prior to Gillespie still distillation lowered ruthenium volatility by factors varying from 420 for 12 M HNO_3 to 38.4 for Darex waste solution and increased nitric acid separation factors correspondingly (Table II).

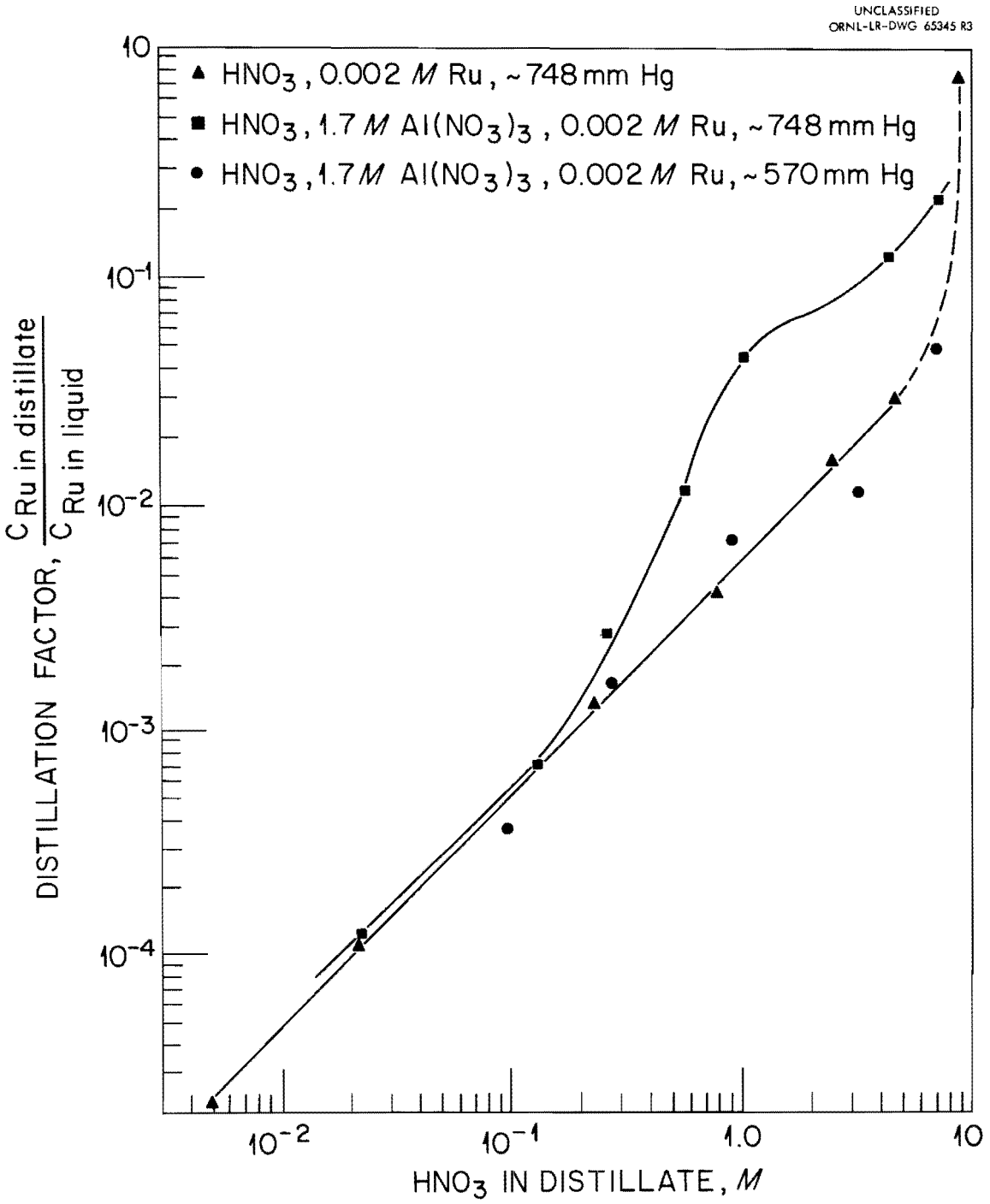

Fig. 7. Volatilization of ruthenium from acid nitrate solutions.

Table II. Effect of 0.1 M H_3PO_3 on Ruthenium Volatility during Distillation of Nitrate Solutions

Solution	Activity of Ru in Distillate, counts/min·ml		Reduction Factor	Separation Factor* HNO_3 from Ru	
	No H_3PO_3	0.1 M H_3PO_3		No H_3PO_3	0.1 M H_3PO_3
12 M HNO_3	2.78×10^5	662	420	0.956	378
6 M HNO_3**	3.76×10^5	145	2593	0.126	364
1.7 M $Al(NO_3)_3$-2 M HNO_3	1.77×10^4	85	208	11.1	2055
Snythetic waste solutions***					
TBP-25	9.08×10^3	40	227	13.8	4920
Purex	3.17×10^4	650	48.8	2.22	117
Darex	9.64×10^4	2.51×10^3	38.4	1.50	57.5

* Defined as $\dfrac{C_{HNO_3(V)}/C_{HNO_3(L)}}{C_{Ru(V)}/C_{Ru(L)}}$.

** Ru added as nitrosyl hydroxide.

*** Compositions listed in Table I.

C. Reaction Mechanisms

Chemical reduction of H_3PO_3 is reported to take place through the symmetrical, undissociated form of the acid, $P(OH)_3$[20]. Increasing the concentration of hydrogen ion in the solution favors formation of $P(OH)_3$:

$$HP(O)OOH^- + H^+ \rightleftharpoons HPO(OH)_2 \rightleftharpoons P(OH)_3.$$

In preliminary work on the stoichiometry of the phosphite-nitrate reaction, visible reaction between 1.0 M H_3PO_3 and 6 M HNO_3 began and was apparently complete between 110 and 115°C. when no salt nitrate was present. In solutions containing salt nitrate (e.g., 1 M H_3PO_3, 2 M $Al(NO_3)_3$), the reaction was still incomplete at 250°C. Small concentrations of Ag^+, Hg^{++}, Pd, and oxides of nitrogen aid in initiating the reaction[2,3,16]. The phosphite is oxidized to phosphate, while the nitrate is reduced to a mixture of NO and NO_2 with smaller amounts of N_2O and N_2. The proportions of the products vary with the conditions of the experiment, but the principal product is normally NO[22].

CONCLUSIONS

Relatively small concentrations of phosphite and/or hypophosphite are very effective in preventing volatilization of ruthenium from Purex, TBP-25, and Darex waste solutions during evaporation. Ruthenium volatility was decreased to less than 0.1% during high-temperature (~1000°C.) calcination of Purex and TBP-25 wastes made approximately 2 M in phosphite or hypophosphite.

The use of phosphite or hypophosphite to decrease or eliminate ruthenium volatility will minimize activity in the off-gas from waste fixation processes. Since both phosphite and hypophosphite are oxidized to phosphate, which is a glass-former, they

aid in producing a solid product with low void space and low solubility. Dense phosphate products with relatively low (850-1050°C.) melting points can be made from Purex, TBP-25, and Darex wastes. Operation at these temperatures allows the use of stainless steel instead of ceramic containers.

ACKNOWLEDGMENT

The authors are indebted to G. D. Davis, W. E. Shockley, K. L. Servis, and F. R. Clayton for assistance with the laboratory work.

LITERATURE CITED

1. Bancroft, A. R., Watson, L. C., and Hoelke, C. W., "The Volatilization and Collection of Ruthenium and Cesium in a System for Incorporating Fission Products into Glass," Atomic Energy of Canada, Ltd., CRCE-1004 (Jan. 20, 1961).
2. Blaser, B., Z. Physik Chem. A166 64 (1933).
3. Blaser, B. and Matei, I., Ber. 64B 228-9 (1931).
4. Callis, C. F., "Deposition of Ruthenium Activity on Oxidizer During Head-End Treatment," Hanford Atomic Products Operation, HW-19391 (Nov. 16, 1950).
5. Callis, C. F., and Moore, R. L., U. S. Patent 2,903,332 (Sept. 8, 1959).
6. Clark, W. E., and Godbee, H. W., "Waste Treatment and Disposal Progress Report for October and November 1961," Oak Ridge National Laboratory, ORNL-TM-133, pp. 16-25 (Mar. 13, 1962).
7. Ibid., December 1961 and January 1962, ORNL-TM-169, pp. 10-14 (June 15, 1962).
8. Ibid., February and March 1962, ORNL-TM-252, pp. 22-29 (Sept. 10, 1962).
9. Ibid., November 1962-January 1963, in preparation.
10. Clark, W. E. and Godbee, H. W., SM-31/12, "Proceedings of the Symposium on the Treatment and Storage of High Level Radioactive Wastes," Vienna (Oct. 8-13, 1962).
11. Elliott, M. N., Grover, J. R., Hardwick, W. H., and Johnson, K.D.B., "The Disposal of Fission Product Wastes by Incorporation into Glass," United Kingdom Atomic Energy Authority, AERE-R 3610 (January 1961).
12. Gillespie, D. T., Anal. Ed., Ind. Eng. Chem. 18 575-577 (1946).

13. Godbee, H. W., and Clark, W. E., Oak Ridge National Laboratory, unpublished results.

14. Godbee, H. W., and Roberts, J. T., "Laboratory Development of a Pot Calcination Process for Converting Liquid Wastes to Solids," Oak Ridge National Laboratory, ORNL-2986 (Aug. 30, 1961).

15. Hancher, C. W. and Suddath, J. C., "Pot Calcination of Simulated High-Level Waste with Continuous Evaporation," Oak Ridge National Laboratory, ORNL-TM-117 (Dec. 26, 1961).

16. Krause, A., Ber. 71B 1033-40 (1933).

17. MacQueen, D. K. and Stevens, J. I., "Design Bases for ICPP Waste Calcination Facility," Idaho Chemical Processing Plant, National Reactor Test Site, IDO-14462 (Apr. 22, 1959).

18. Mansfield, R. G., Oak Ridge National Laboratory, unpublished work, 1953.

19. May, C. E., Newby, B. J., Rohde, K. L., and Withers, B. D., "Ruthenium Behavior in a Nitric Acid Scrubber," Idaho Chemical Processing Plant, National Reactor Test Site, IDO-14448 (Sept. 29, 1958).

20. Mitchell, A. D., J. Chem. Soc. 123 1031 (1924).

21. Rohde, K. L., May, C. E., Newby, B. J., and Withers, B. D., Ind. Eng. Chem. 51 68-70 (1959).

22. Servis, K. L., Oak Ridge National Laboratory, unpublished work (1961).

23. Wilson, A. S., "Ruthenium Behavior in Nitric Acid Distillation," Hanford Atomic Products Operation, HW-45620 (Sept. 1, 1956).

DISTRIBUTION

1. F. L. Culler
2-7. R. E. Blanco
8. J. C. Bresee
9. K. B. Brown
10-14. W. E. Clark
15. D. E. Ferguson
16-20. H. W. Godbee
21. H. B. Graham
22. H. E. Goeller
23. A. T. Gresky
24-25. E. J. Murphy
26. M. J. Skinner
27. M. E. Whatley
28-29. Central Research Library
30. Document Reference Section (Y-12)
31. Laboratory Records (RC)
32-33. Laboratory Records
34. Research and Development (ORO)
35-50. DTIE