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

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### 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 approaching equilibrium by factors which varied from 38 for Darex to 225 for TBP-25 (aluminum) waste solutions. The final products with Purex, TBP-25, and Darex (stainless steel) wastes were insoluble glassy solids with densities of 2.4 to 3.8 g/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.

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## 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<sup>(1)</sup>. 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<sup>(2,3)</sup>.

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 synthetic wastes from the Purex<sup>(4)</sup> and TBP-25 (aluminum)<sup>(5,6,7)</sup> solvent extraction processes 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 semicontinuous experiments. Semicontinuous calcination of synthetic Purex, TBP-25, and Darex (stainless steel) wastes without added phosphite or hypophosphite has been demonstrated on both laboratory and unit operation scales<sup>(8,9)</sup>.

In a process developed at Chalk River Canada<sup>(10)</sup> 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<sup>(11)</sup> 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<sup>(12)</sup> for reducing waste solutions to granular solids at about 500°C, 90-99% of the ruthenium may be volatilized and ad-



sorbed 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 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<sup>(13)</sup>, nitric oxide<sup>(8,14)</sup> and dibutyl and monobutyl phosphoric acids<sup>(8)</sup>. 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 (Fig. 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<sub>2</sub> 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.

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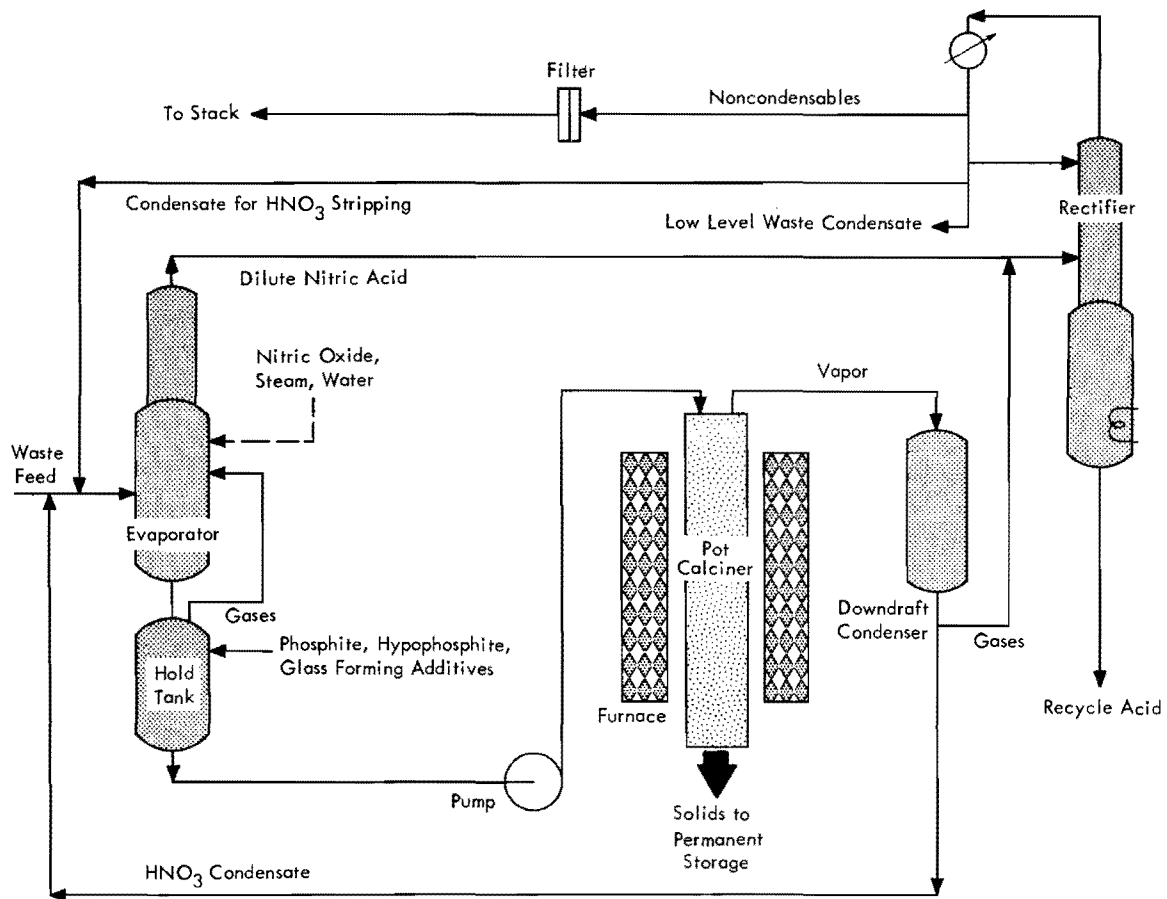


Fig. 1. High activity wastes are converted to solids by pot calcination. The dotted line indicates an optional operation.

## 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 ( $\text{HPO}_3^-$ ) and hypophosphite ( $\text{H}_2\text{PO}_2^-$ ) 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/ml stable ruthenium\* and 0.1  $\mu\text{c/ml}$  Ru-106 tracer. Stable and radioactive ruthenium were added as the chlorides. Phosphite was added as phosphorous acid,  $\text{H}_3\text{PO}_3$ . Hypophosphite was usually added as sodium hypophosphite,  $\text{NaH}_2\text{PO}_2$ , and more rarely as hypophosphorous acid,  $\text{H}_3\text{PO}_2$ .

Experiments were carried to about 500°C when using a 200 ml Pyrex flask as a pot and to about 1000°C in a quartz outer pot holding either a 200 ml quartz or stainless steel beaker (Fig. 2). Condensates were analyzed for Ru-106 in a gamma scintillation counter. The equipment was decontaminated between experiments by boiling 15 M  $\text{HNO}_3$  through it. The residues from the low-temperature experiments were analyzed by gamma counting after dissolution in refluxing 12 M  $\text{HCl}$ . Residues from the high-temperature experiments were not dissolved in most cases since the dissolution required about a week in refluxing  $\text{HCl}$ , 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 ( $\text{H}_3\text{PO}_3$ ) addition was investigated by distillation in a Gillespie equilibrium still <sup>(15)</sup>. 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. The effect of reflux time on ruthenium volatility was not investigated. In practice it was found that if the condenser was air-cooled, no visible ruthenium deposit was obtained unless the concentration of the acid in the distillate was greater than 8 M.

\* 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.

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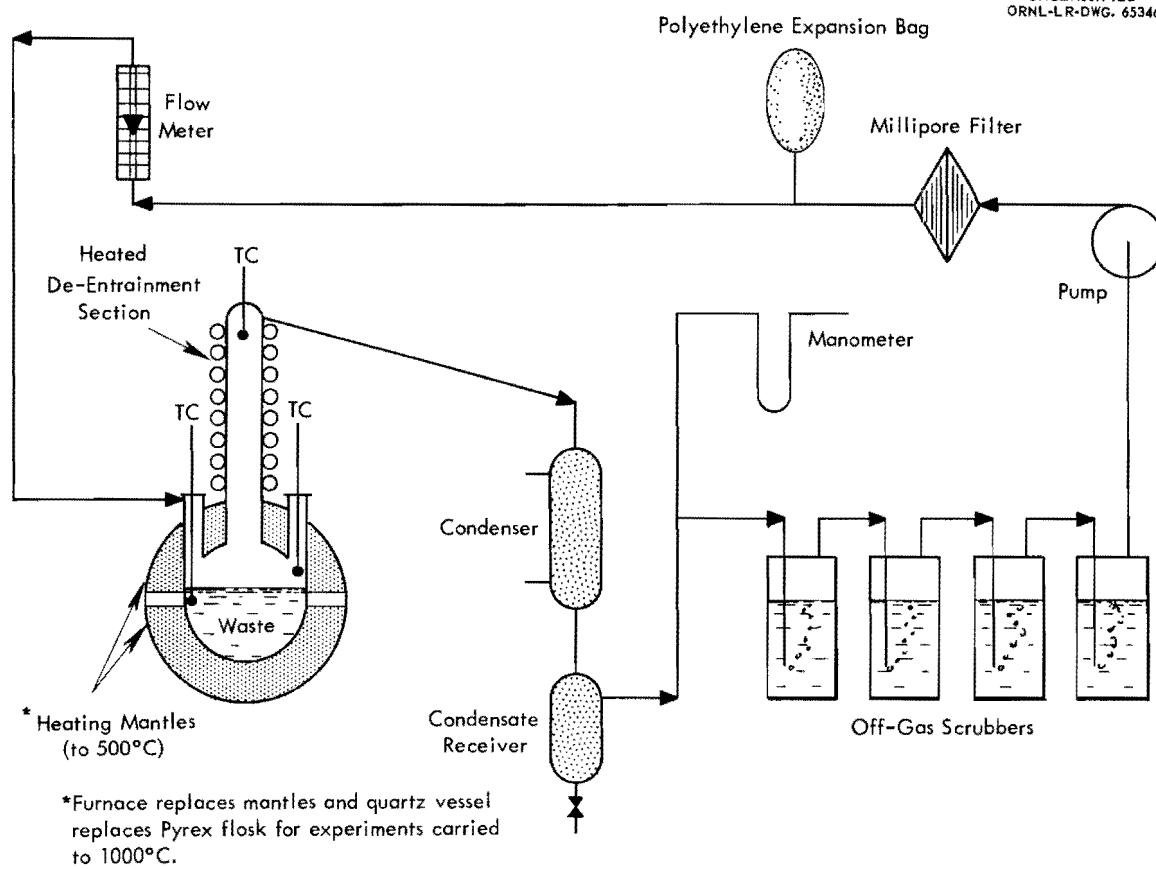


Fig. 2. Schematic of apparatus for studying ruthenium volatility during batch evaporation-calcination was studied in glass and quartz equipment.



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

Component	Darex (24 gal/kg U-235)	Purex (40 gal/ton U processed)	TBP-25 (106 gal/kg U-235)
Al <sup>3+</sup>		0.1 <u>M</u>	1.72 <u>M</u>
Fe <sup>3+</sup>	1.25 <u>M</u>	0.5 <u>M</u>	0.16 mg/ml
Cr <sup>3+</sup>	0.38 <u>M</u>	0.01 <u>M</u>	
Hg <sup>2+</sup>			4.02 mg/ml
Ni <sup>2+</sup>	0.18 <u>M</u>	0.01 <u>M</u>	
Mn <sup>2+</sup>	0.04 <u>M</u>		
Na <sup>+</sup>		0.6 <u>M</u>	2.4 mg/ml
NH <sub>4</sub> <sup>+</sup>			0.05 <u>M</u>
H <sup>+</sup>	0.75 <u>M</u>	5.6 <u>M</u>	1.26 <u>M</u>
Ru	0.2 mg/ml	0.2 mg/ml	0.2 mg/ml
Cl <sup>-</sup> *	260 ppm	165 ppm	160 ppm
NO <sub>3</sub> <sup>-</sup>	6.0 <u>M</u>	6.1 <u>M</u>	6.6 <u>M</u>
SO <sub>4</sub> <sup>=</sup>		1.0 <u>M</u>	
Density, g/ml	1.33	1.30	1.32

\* Except for the Darex waste, which normally contains ~100 ppm Cl<sup>-</sup>, all Cl<sup>-</sup> present was added with the Ru as RuCl<sub>3</sub>.

#### DATA

##### 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 (Fig. 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<sub>3</sub>PO<sub>4</sub>, 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

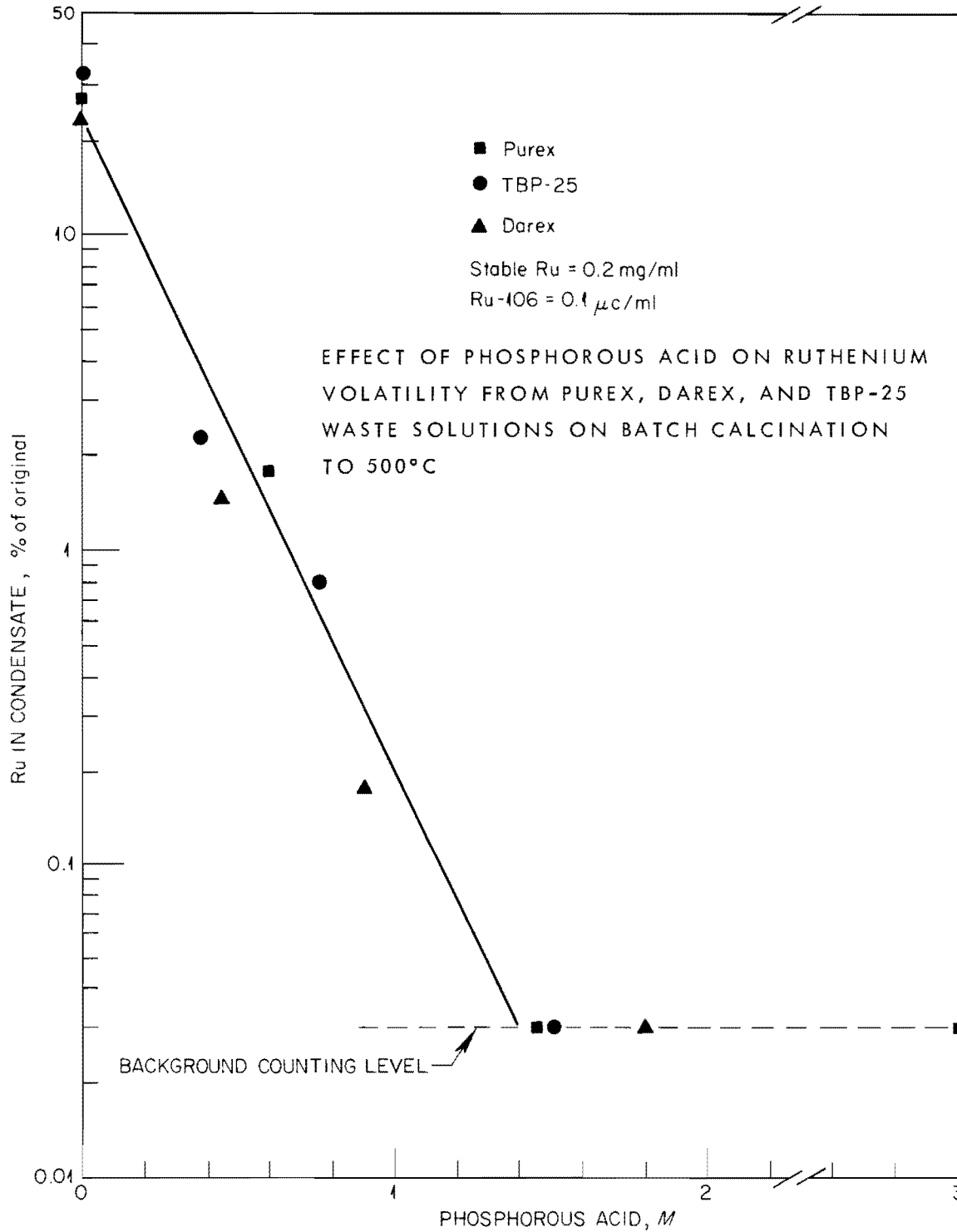


Fig. 3. Phosphorous acid, 1.5 M, lowered the volatility of Ru-106 to background levels in batch calcination of Purex, TBP-25, and Darex waste solutions to 500°C.

no additive. The percentage of original ruthenium in the condensate was not increased by increasing the ruthenium in the feed from 0.2 mg/ml to 20 mg/ml in the presence of 1.5 M  $\text{HPO}_3^-$ .

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. With a stainless steel container the percentages in the condensate were factors of 1.5 to 6.5 less (Fig. 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 g/ml, incorporated 35 to 45 wt % 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 (Fig. 5) contained 29, 21.8, 21.5, 10.7, 8.7, and 6.4 wt %, respectively, of  $\text{P}_2\text{O}_5$ ,  $\text{Na}_2\text{O}$ ,  $\text{SO}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{MgO}$ , and  $\text{B}_2\text{O}_3$ . 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 (Fig. 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 g/ml, contained from 26 to 35 wt % oxides from waste, and represented volume

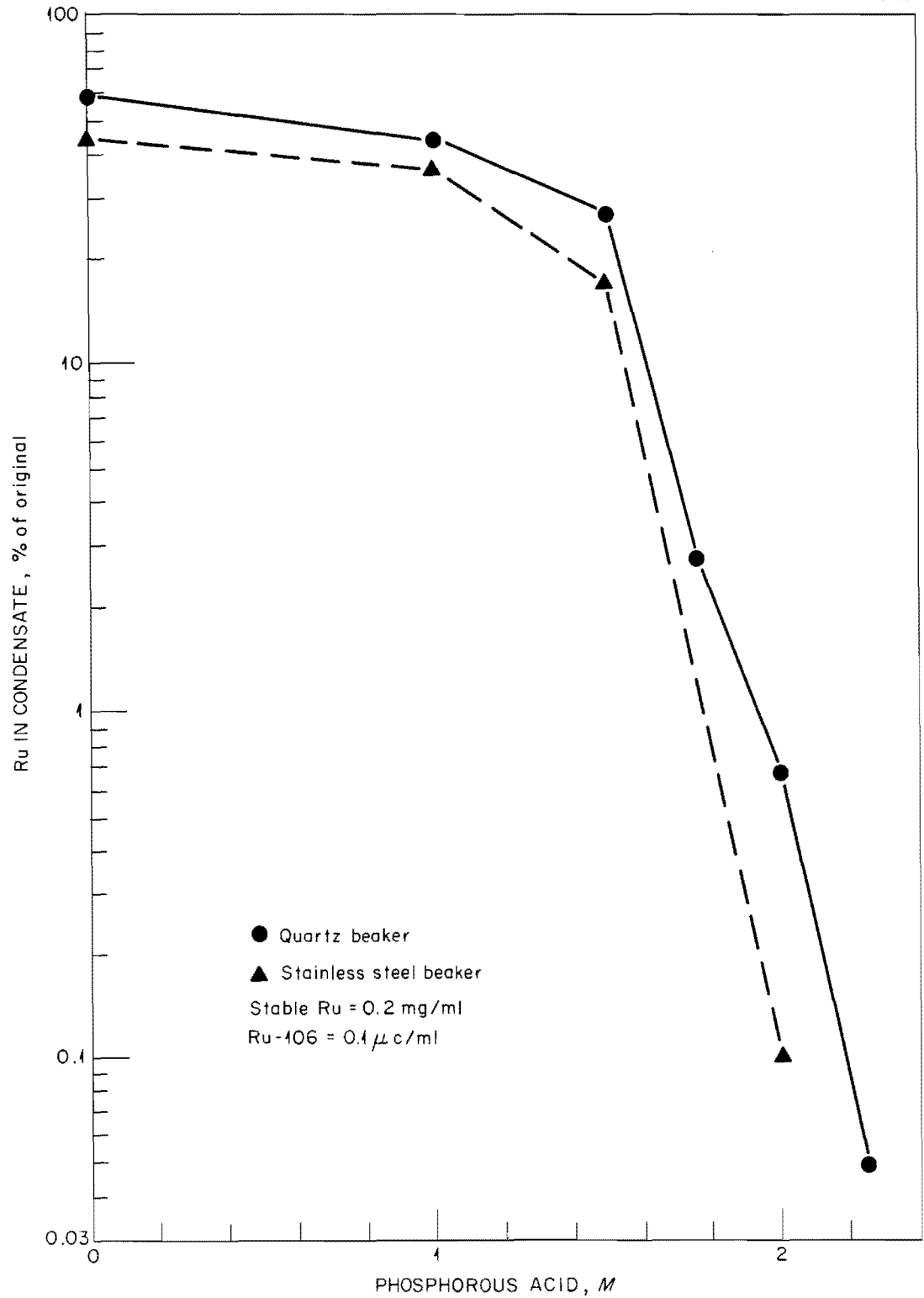


Fig. 4. Phosphorous acid, 2-2.3 M, reduced the volatility of ruthenium to 0.1% or less in batch calcination of TBP-25 waste to 1000°C.

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 wt %, respectively, of  $P_2O_5$ ,  $Al_2O_3$ ,  $Na_2O$ , and  $PbO$  (Fig. 6). Leaching (dissolution) rates for this glass in distilled water, determined by the leaching of Cs-137 tracer, decreased from  $2.1 \times 10^{-6}$  to  $2.5 \times 10^{-7}$  g/cm<sup>2</sup>·day 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 water.

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 g/ml, contained 13.4 to 32.3 wt % 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 wt %, respectively, of  $P_2O_5$ ,  $Al_2O_3$ ,  $Na_2O$ ,  $Fe_2O_3$ ,  $B_2O_3$ ,  $Cr_2O_3$ , and NiO.

#### 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 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 (Fig. 7). This effect is generally consistent with observations made elsewhere\*<sup>(13)</sup>. 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) ( $8 \times 10^{-4}$  vs  $4.9 \times 10^{-3}$  M). The layers were miscible on stirring. A black deposit, presumably  $RuO_2$ , was noted in the cooler parts of the condenser.

\* 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.

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Fig. 5. Purex waste oxides were 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  $NaOH$ , evaporated to dryness, and heated to  $850^\circ C$ . Density 2.7 g/ml; product contained 39 wt % waste oxides; volume reduction factor, 5.5.

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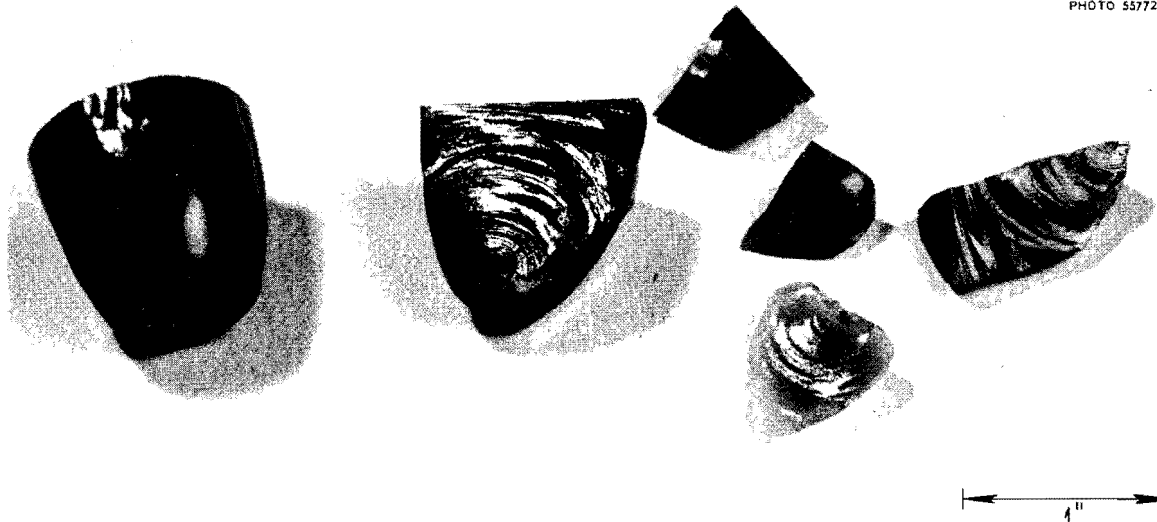


Fig. 6. TBP-25 waste oxides were incorporated in phosphate-lead glass. Waste made 2 M  $NaH_2PO_2$  and 0.25 M in  $PbO$ , evaporated to dryness and heated to  $1000^\circ C$ . Density 2.8 g/ml; 26 wt % waste oxides; volume reduction factor, 8.

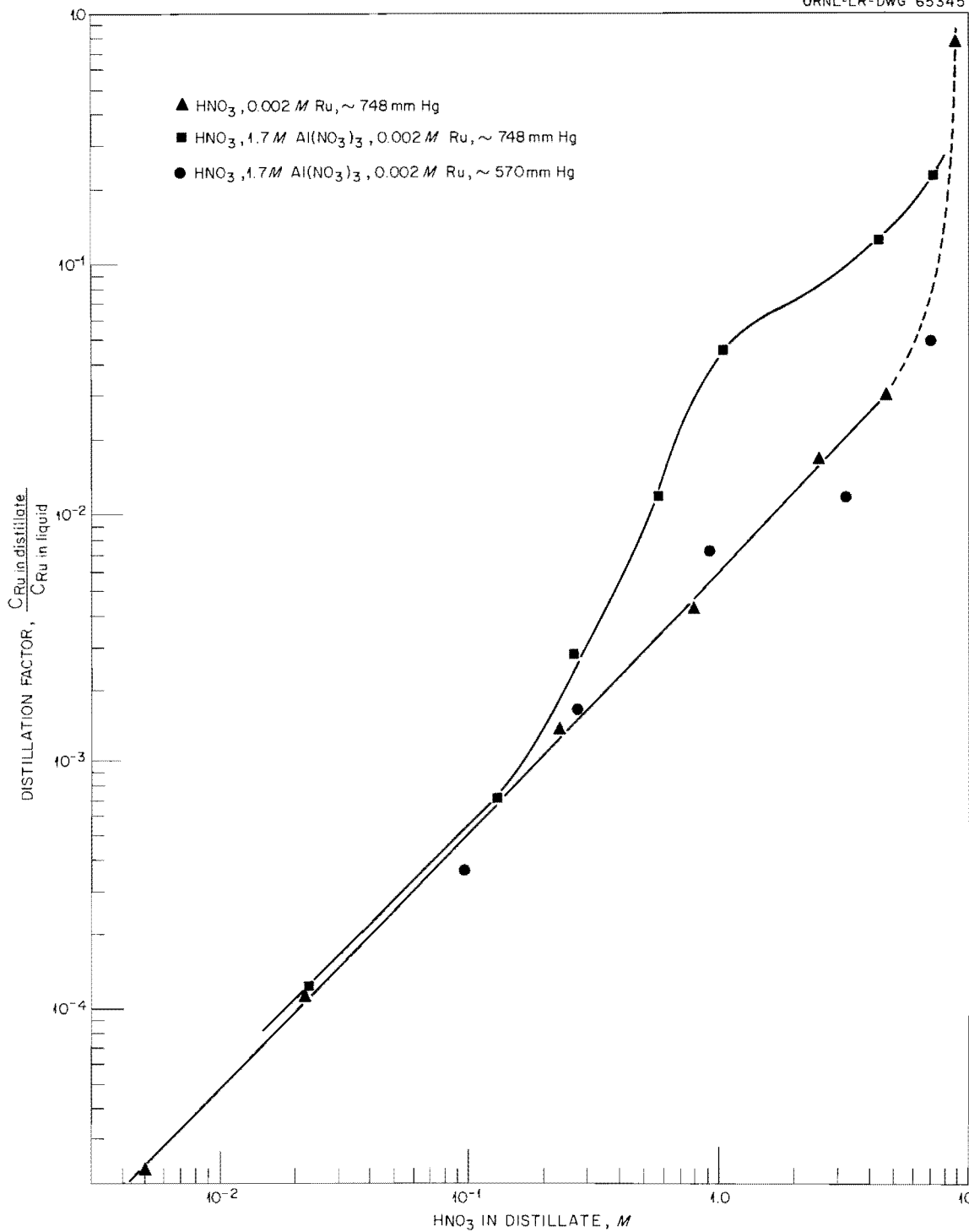


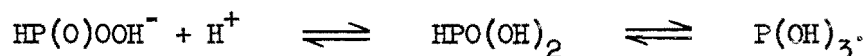
Fig. 7. Volatilization of ruthenium from acid nitrate solutions.

Boiling  $\text{HNO}_3$  solutions, 1.7 M in  $\text{Al}(\text{NO}_3)_3$  behaved similarly to pure  $\text{HNO}_3$  solutions as long as the free  $\text{HNO}_3$  concentration in the distillate was less than 0.1 M. At higher distillate acid concentrations the ruthenium volatility increased rapidly (Fig. 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 Hg, the ruthenium volatilities approximated those from  $\text{HNO}_3$  alone (Fig. 7).

The effectiveness of relatively small concentrations of phosphite in suppressing ruthenium volatility is shown by the fact that 0.1 M  $\text{H}_3\text{PO}_3$  added to nitric acid and synthetic waste solutions prior to Gillespie still distillation lowered ruthenium volatility by factors varying from 447 for 12 M  $\text{HNO}_3$  to 38.8 for Darex waste solution and increased nitric acid separation factors correspondingly (Table 2).

### C. Reaction Mechanisms

Chemical reduction of  $\text{H}_3\text{PO}_3$  is reported to take place through the symmetrical, undissociated form of the acid,  $\text{P}(\text{OH})_3$ <sup>(16)</sup>. Increasing the concentration of hydrogen ion in the solution favors formation of  $\text{P}(\text{OH})_3$ :



In preliminary work on the stoichiometry of the phosphite-nitrate reaction, visible reaction between 1.0 M  $\text{H}_3\text{PO}_3$  and 6 M  $\text{HNO}_3$  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  $\text{H}_3\text{PO}_3$ , 2 M  $\text{Al}(\text{NO}_3)_3$ ), the reaction was still incomplete at 250°C. Small concentrations of  $\text{Ag}^+$ ,  $\text{Hg}^{++}$ , Pd, and oxides of nitrogen aid in initiating the reaction<sup>(17,20)</sup>. The phosphite is oxidized to phosphate, while the nitrate is reduced to a mixture of NO and  $\text{NO}_2$  with smaller amounts of  $\text{N}_2\text{O}$  and  $\text{N}_2$ . The proportions of the products vary with the conditions of the experiment, but the principal product is normally NO<sup>(20)</sup>.

### CONCLUSIONS

Relatively small concentrations of phosphite and/or hypophosphite are very effective in preventing volatilization of ruthenium from Purex, TBP-25,



Table 2. Effect of 0.1 M H<sub>3</sub>PO<sub>3</sub> on Ruthenium Volatility During Distillation of Nitrate Solutions

Solution	Activity of Ru in Distillate, c/min·ml		Reduction Factor	Separation Factor,* HNO <sub>3</sub> from Ru	
	No H <sub>3</sub> PO <sub>3</sub>	0.1 M H <sub>3</sub> PO <sub>3</sub>		No H <sub>3</sub> PO <sub>3</sub>	0.1 M H <sub>3</sub> PO <sub>3</sub>
12 M HNO <sub>3</sub>	2.78 x 10 <sup>5</sup>	662	420	0.956	378.
6 M HNO <sub>3</sub> **	3.76 x 10 <sup>5</sup>	145	2593	0.126	364.
1.7 M Al(NO <sub>3</sub> ) <sub>3</sub> -2 M HNO <sub>3</sub>	1.77 x 10 <sup>4</sup>	85	208	11.1	2055.
<u>Synthetic Waste Solutions***</u>					
TBP-25	9.08 x 10 <sup>3</sup>	40	227	13.8	4920.
Purex	3.17 x 10 <sup>4</sup>	650	48.8	2.22	117.
Darex	9.64 x 10 <sup>4</sup>	2.51 x 10 <sup>3</sup>	38.4	1.50	57.5

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

\*\* Ru added as the nitrosyl hydroxide.

\*\*\* Compositions listed in Table 1.

and Darex waste solutions during evaporation. Ruthenium volatility was decreased to less than 0.1% during high-temperature ( $\sim 1000^{\circ}\text{C}$ ) 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 ( $850\text{-}1050^{\circ}\text{C}$ ) 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.

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