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OAK RIDGE NATIONAL LABORATORY
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ABSTRACT

Fused salt fluorination tests were conducted at 600°C to determine the feasibility of recovering plutonium as PuF_6 in the fused salt-fluoride volatility process. Recoveries and material balances were good, although the initial plutonium concentration in 50-50 mole % $NaF-ZrF_4$ or 31-24-45 mole % $LiF-NaF-ZrF_4$ salt was only 2 ppm. The volatilization reaction appeared to be approximately first-order with respect to the plutonium concentration in the salt. Results of absorption of the volatilized PuF_6 on beds of LiF, CaF_2, or NaF indicate that this is a possible method of trapping the material in fluoride volatility processes, possibly separately from UF_6.

CONTENTS

1.0 INTRODUCTION

This report summarizes exploratory work on the volatilization of PuF_6 from fused salts with fluorine. Volatilization of uranium hexafluoride from fused fluorides with elemental fluorine is the basis of a reactor fuel processing method being developed, and the comparable vapor pressures of PuF_6 and UF_6 indicate that plutonium might be recovered with the uranium if the disparity between the chemical stabilities of PuF_6 and UF_6 is unimportant (1-4). The recovery of plutonium as well as uranium is necessary in the processing of reactor fuel of low enrichments.

The work was conducted with 0.1 mc of Pu-239 in each test to determine whether volatilization of PuF_6 from fused salts is feasible and whether PuF_6 absorption on solid beds might be useful in the process. The favorable results obtained in both areas indicate the desirability of extending the work to tests at higher plutonium concentrations and under conditions that would prevail in actual processing. The recovery of plutonium separately from uranium was briefly explored, and this represents another aspect needing further effort.

The authors gratefully acknowledge the assistance of the Analytical Chemistry Group under J. H. Cooper in carrying out the a analytical work. F. L. Moore was particularly helpful in assisting with development of the analytical procedures. The assistance of T. E. Crabtree and C. J. Shipman in the laboratory work and their helpful suggestions were also invaluable.

2.0 FUSED SALT-FLUORIDE VOLATILITY METHOD

In the fused salt-fluoride volatility process being developed for uranium recovery, two steps, namely, dissolution by hydrofluorination and UF_6 volatilization by direct fluorination, are carried out in the presence of a fused salt at a temperature of 500-600°C (4). The third step, absorption of the UF_6 on NaF beds, results in relatively complete decontamination of the UF_6 product from volatile or entrained fission product fluorides. Based on the results reported here, adaptation of the process for recovery of PuF_6 as well as UF_6 appears feasible, although the fluorine utilization in PuF_6 volatilization is low and fluorine recycle would probably be required (Fig. 1). It also appears probable that the volatilized PuF_6 could be recovered by absorption, either together with or separately from UF_6, on a system of fluoride beds. For separate recovery of the PuF_6, a system for separate gaseous transfer and decontamination from fission products would be needed. In many fuels the Pu/U ratio would be low, but the process would also be operable with recycled plutonium fuel where this would not be the case.

In the 18 fluorination experiments shown in Table 1, fused salts containing ~2 ppm of plutonium were used to determine the extent of PuF_6 volatilization and to observe whether the volatilized PuF_6 could be trapped on various solid fluoride beds. The data in Table 2 indicate that NaF, LiF, or CaF_2 is effective in absorption

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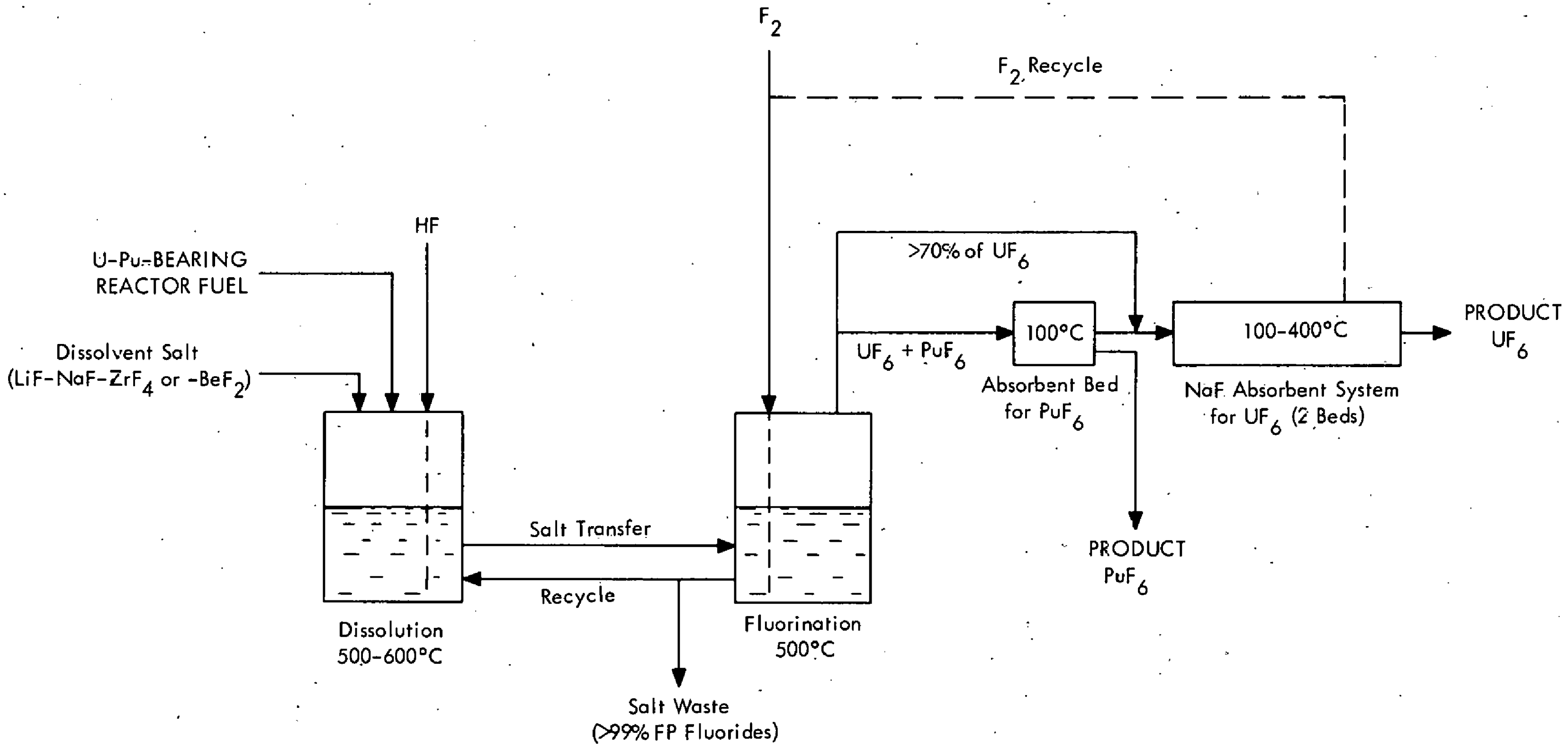

Fig. 1. Processing of U-Pu-bearing reactor fuel by Fused Salt-Fluoride Volatility method. The used NaF is transferred to the molten salt fluorinator. Further absorption steps to purify the plutonium from fission product activities may be necessary as in the case of UF$_6$.

Table 1. Summary of PuF_6 Volatilization Tests with Fused Salt

2 ppm Pu in 50 g 31-24-45 mole % $LiF-NaF-ZrF_4$ at 600°C with exceptions noted in runs 1, 2, 3, 5, 6, and 11

Run	Flow Rate, ml/min			Pu Retention in Fused Salt, %													
No.	F_2	He	Special Remarks	1 hr	2 hr	3 hr	4 hr	5 hr	6 hr	7 hr	8 hr	10 hr	12 hr	14 hr	16 hr	18 hr	20 hr
1	100	--	50-50 mole % $NaF-ZrF_4$	80.4	28.6	21.6	--	14.0	--	--	--	--	--	--	--	--	--
2	100	--	50-50 mole % $NaF-ZrF_4$	--	--	--	--	--	15.3	--	--	--	--	--	--	--	--
3	100	--	50-50 mole % $NaF-ZrF_4$	--	--	--	--	--	27.2	--	--	--	--	--	--	--	--
4	100	--		--	--	--	--	--	37.7	--	--	--	--	--	--	--	--
5	100	--	50-50 mole % $NaF-ZrF_4$	--	--	39.1	--	--	16.0	--	--	--	--	--	--	--	--
6	100	--	50-50 mole % $NaF-ZrF_4$	75.6	70.5	56.4	41.7	28.2	--	--	--	--	--	--	--	--	--
7	100	--		92.3	74.4	57.7	48.1	40.4	--	--	--	--	--	--	--	--	--
8	100	--		--	69.8	--	52.6	--	34.6	--	23.1	15.4	10.5	7.6	4.3	2.7	1.4
9	50	--		85.9	74.4	68.6	73.0	60.2	53.8	--	--	--	--	--	--	--	--
10	150	--		67.9	48.7	42.3	33.3	20.5	16.7	--	--	--	--	--	--	--	--
11	100	--	500°C	75.6	65.4	54.5	44.9	39.7	34.0	--	--	--	--	--	--	--	--
12	100	--	7 mole % UF_6 in F_2 stream	77.0	62.0	48.0	38.5	31.7	30.1 (5.3 hr)	--	--	--	--	--	--	--	--
13	25	75		75.6	70.2	79.5	87.2	65.4	65.4	--	--	--	--	--	--	--	--
14	100	--	2.98% UF_4 in original salt	74.4	40.4	35.2	26.9	27.6	20.5	--	--	--	--	--	--	--	--
15	50	50		53.2	47.5	40.4	41.0	46.2	34.0	--	--	--	--	--	--	--	--
16	100	--		40.8	34.8	28.8	19.1	15.9	--	--	--	--	--	--	--	--	--
17	100	--	13.5-mil-i.d. capillary gas inlet	65.4	52.6	48.1	30.8	29.5	20.5	--	--	--	--	--	--	--	--
18	150	--	30-mil-i.d. capillary gas inlet	73.1	53.2	41.0	29.5	23.7	21.2 (5.5 hr)	--	--	--	--	--	--	--	--

Table 2. Summary of PuF_6 Absorption Results and Material Balances

(See Table 1 for Summary of Conditions in Fused Salt Fluorination Tests)

Run No.	Description of Absorption Beds			Pu Distribution, % of total							Pu Material Balance, %	
	No. 1	No. 2	No. 3	Fused Salt	Salt Samples	Tubing Walls	Bed No. 1	Bed No. 2	Bed No. 3	Bed Walls[d]	Of Total Pu	Of Volatile Pu
1[a]	200 g NaF, 25°C	--	--	19.4	--	1.9	--	--	--	--	--	--
2[a]	8 g NaF, 25°C	8 g NaF, 25°C	--	27.1	--	0.8	46.6	0.2	--	1.3	76.0	67.1
3[a]	8 g LiF, 25°C	8 g NaF, 25°C	--	29.0	--	1.3	58.4	1.4	--	7.4	97.5	96.5
4[b]	8 g LiF, 25°C	8 g NaF, 25°C	--	37.7	--	3.2	62.6	0.1	--	5.1	109.	114.
5[c]	8 g CaF_2, 25°C	8 g NaF, 25°C	--	16.0	13.1	6.1	48.1	0.1	--	4.5	87.9	83.0
6	8 g LiF, 25°C	8 g NaF, 25°C	--	23.2	7.5	1.2	52.8	0.4	--	6.7	96.8	95.2
7	8 g NaF, 25°C	8 g NaF, 25°C	--	40.4	6.5	--	55.1	1.7	--	--	104.	107.
8	8 g CaF_2, 25°C	8 g NaF, 25°C	--	1.4	5.6	2.5	92.9	~0	--	--	102.	103.
9	8 g NaF, 25°C	8 g NaF, 25°C	--	53.8	8.9	4.8	28.8	5.1	--	--	101.	104.
10	8 g CaF_2, 100°C	8 g CaF_2, 25°C	--	16.7	6.2	2.0	73.0	0.1	--	--	98.0	97.5
11	8 g CaF_2, 400°C	8 g CaF_2, 50°C	--	34.0	8.7	4.2	59.6	0.5	--	--	107.	112.
12	8 g CaF_2, 100°C	8 g NaF, 25°C	25 g NaF, 25°C	30.1	7.3	3.7	62.2	0.1	~0	--	103.	105.
13	8 g NaF, 400°C	8 g NaF, 70°C	--	65.4	9.8	8.3	14.7	0.7	--	--	98.9	95.5
14	8 g LiF, 400°C	8 g NaF, 100°C	8 g LiF, 100°C	20.5	5.6	12.2	3.7	51.9	1.3	--	95.2	93.7
15	8 g LiF, 400°C	8 g LiF, 100°C	--	34.0	5.7	37.3	0.7	15.3	--	--	93.0	88.5
16	8 g LiF, 100°C	8 g NaF, 100°C	--	15.9	6.1	15.1	43.8	0.2	--	--	81.1	75.9
17	5 g NaF, 100°C	5 g LiF, 100°C	--	20.5	10.1	18.5	11.9	0.2	--	--	61.2	44.1
18	5 g NaF, 100°C	5 g NaF, 25°C	--	21.2	4.2	27.3	19.2	0.1	--	--	72.0	62.5

[a] Analytical method No. 1 (Sect. 4.2).

[b] Analytical method No. 2 (Sect. 4.2).

[c] Analytical method No. 3 used in runs 5-13 (Sect. 4.2).

[d] Mainly absorbent dust.

of plutonium volatilized as PuF_6. The material balances for many of the tests were in the range 90-100% despite the use of only 0.1 mc of Pu-239 per run. In two tests with uranium present the possibility of separating recovered plutonium and uranium was indicated.

2.1 Retention of Plutonium in Fluorination of Fused Salts

In all tests the plutonium retained in the fused salt decreased during fluorination, indicating volatilization of PuF_6 (Table 1). The rate of disappearance of plutonium from the fused salt had an approximately first-order dependence on the concentration in the salt. Deviations from the first-order dependence could be due to inhomogeneities in the gas or salt mixing.

In three tests (runs 7, 9, and 10) with fluorine flow rates of 50, 100, and 150 ml/min, the volatilization rate constants (assuming a first-order dependence) were approximately proportional, being, respectively, 0.11, 0.18, and 0.31 hr^{-1} (Fig. 2a). The corresponding half-value times in these tests were 6.5, 3.8, and 2.2 hr. It was concluded from this that the amount of plutonium transfer or volatilization depends approximately on the total amount of gas passed through the salt.

A special long-duration test of 20 hr (run 8) demonstrated that the initial plutonium concentration of 2 ppm could be reduced to 1.4% or 0.028 ppm, with no indication that this was a lower limit (Fig. 2b). The half-value time in the initial part of this run, 4.0 hr, duplicated the result in run 7 under about the same conditions. The curvature of the plotted data indicates that the fractional volatilization rate increased to some extent as the experiment proceeded.

Tests at 600°C with 50-50 mole % $NaF-ZrF_4$ salt (Fig. 3) instead of with 31-24-45 mole % $LiF-NaF-ZrF_4$ salt gave little indication that salt composition was a major variable. The half-value time (run 6) was about 3.4 hr. There was no significant change in the volatilization rate at 500°C.

Fluorination with fluorine gas diluted with helium gave anomalous data. With a 50/50 F_2/He gas mixture, there was initially a rapid decrease of plutonium in the salt, but then the rate of disappearance decreased so that the final salt concentration of 34.0% (of initial level) in run 15 compares closely to the 34.6% in run 8. A 25/75 F_2/He gas mixture definitely gave a slower plutonium disappearance rate. Both runs were characterized by abnormally erratic data (Fig. 4).

Evidence was obtained that the rate of plutonium transfer from the fused salt is enhanced by increasing the degree of dispersion of the F_2 in the salt. In the first 16 runs the gas bubble size was the result of using a 1/4-in. tube immersed in the salt. In runs 17 and 18, capillary fluorine inlets were used, giving half-value times of about 2.5 hr, with little difference noted between fluorine flow rates of 100 and 150 ml/min (Fig. 5).

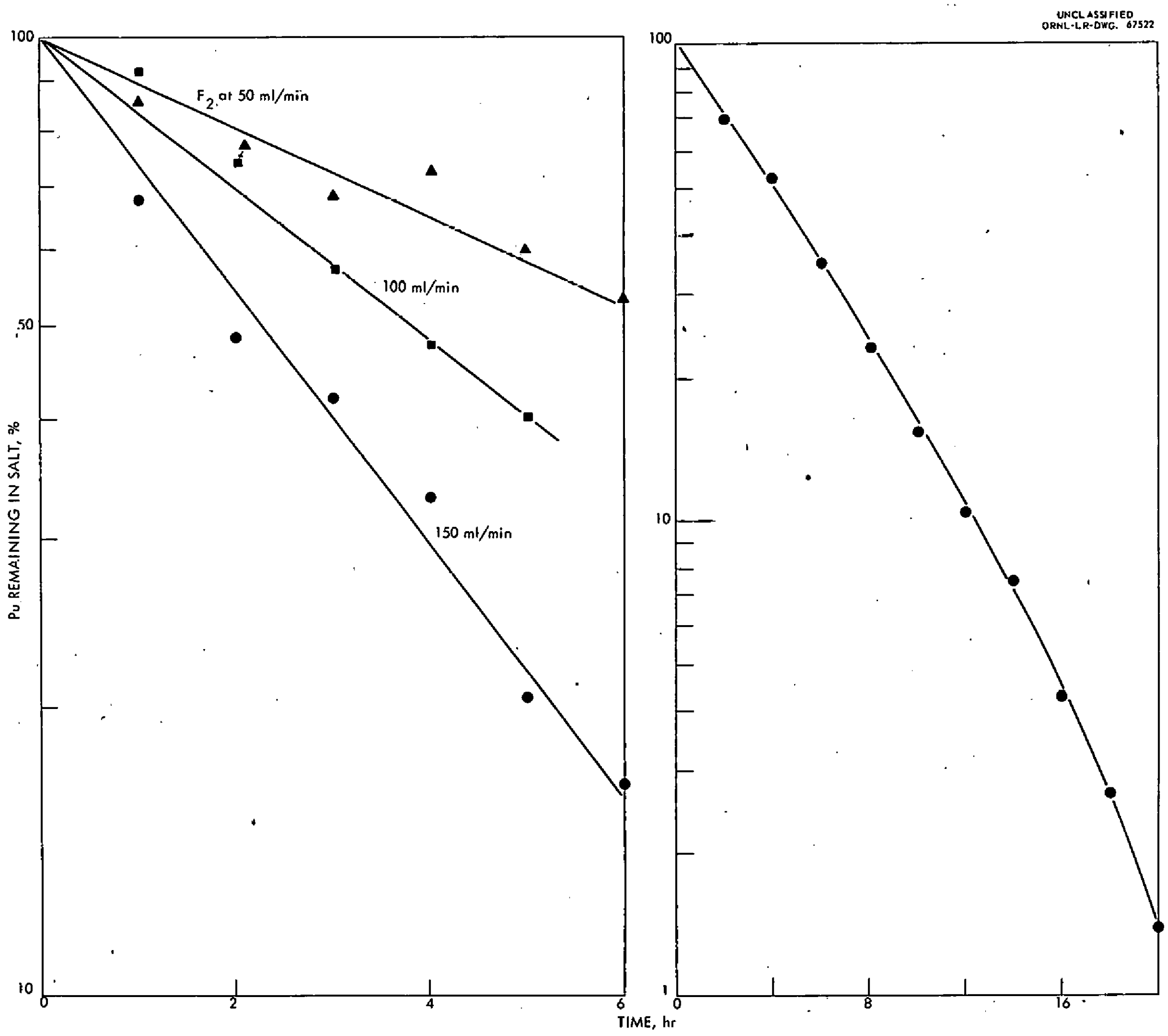

Fig. 2. Volatilization of PuF_6 from fused 31-24-45 mole % LiF-NaF-ZrF_4 at 600°C. (a) 6-hr runs at different fluorine flowrates; (b) 20-hr run at fluorine flowrate of 100 ml/min.

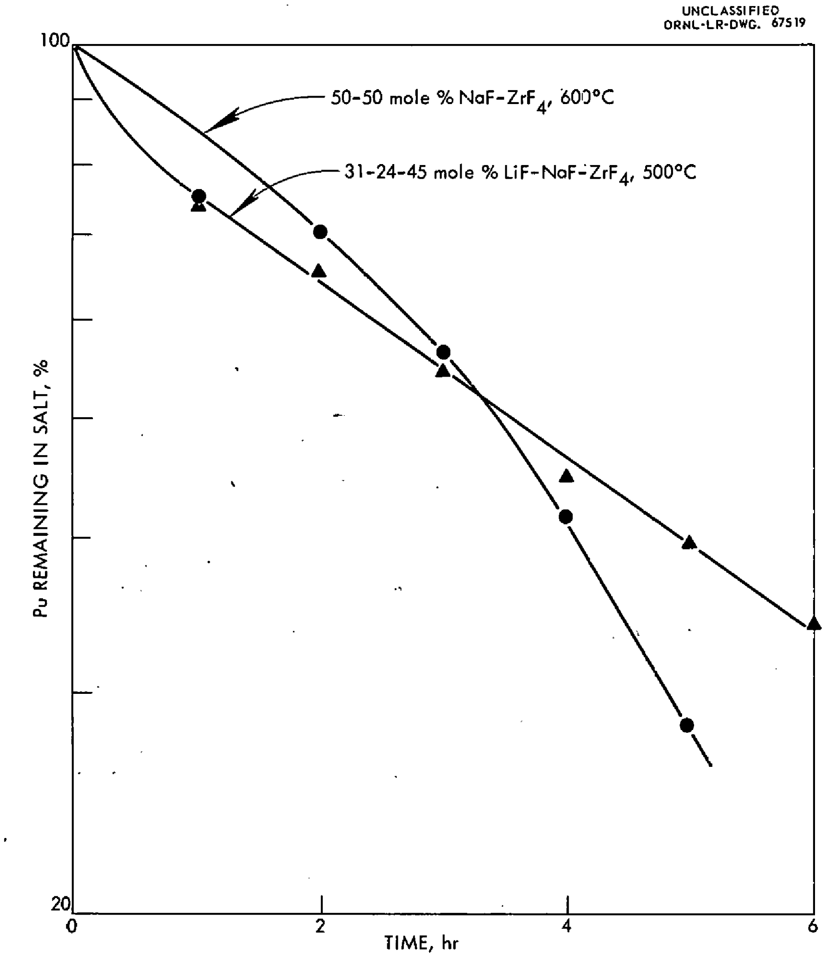

Fig. 3. Comparative PuF_6 volatilization rate from 50-50 mole % $NaF-ZrF_4$ at 600°C and 31-24-45 mole % $LiF-NaF-ZrF_4$ at 500°C. Fluorine flowrate 100 ml/min.

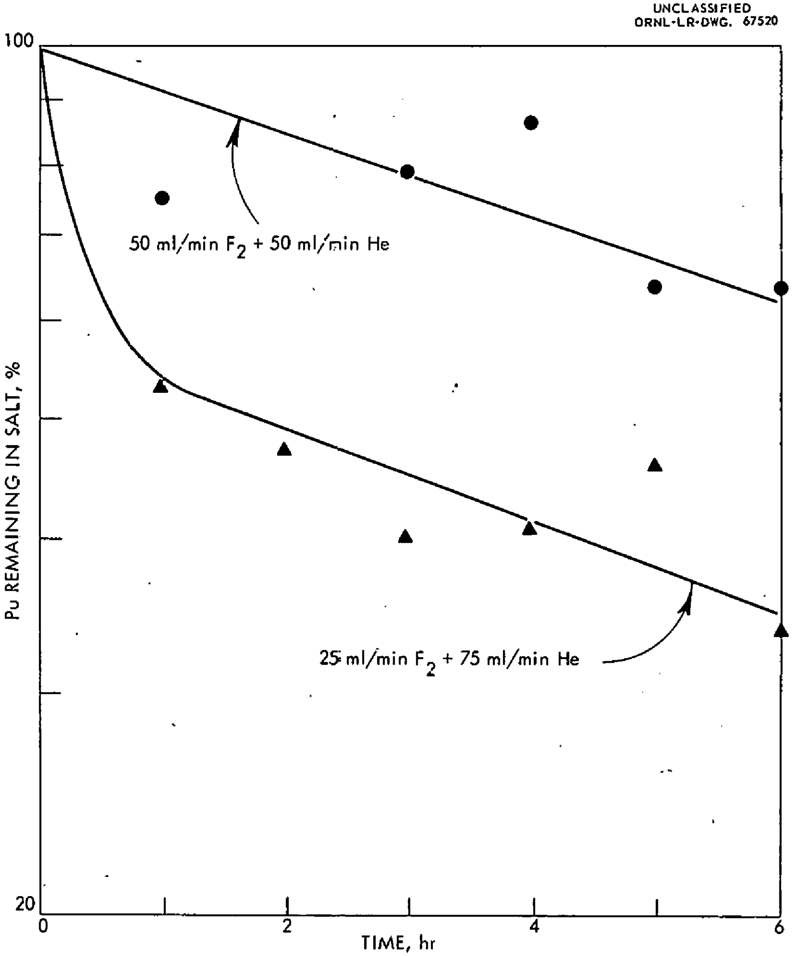

Fig. 4. Effect of dilution of fluorine with helium in volatilization of PuF_6 from 31-24-45 mole % $LiF-NaF-ZrF_4$ salt at 600°C.

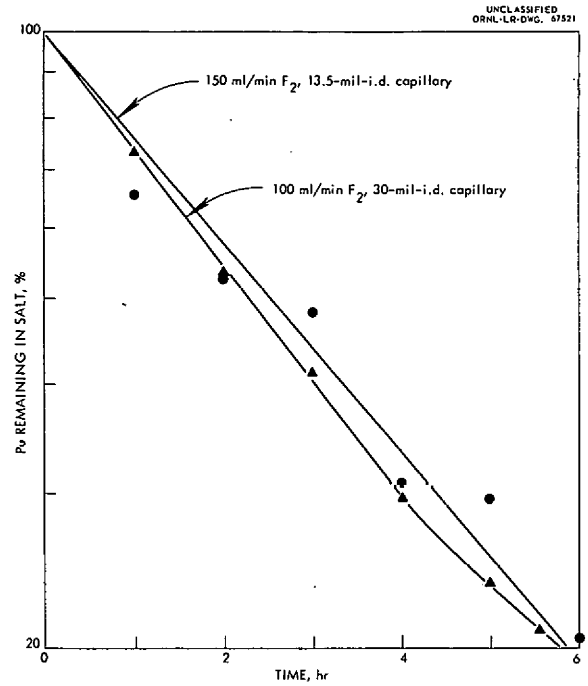

Fig. 5. Volatilization of PuF_6 from 31-24-45 mole % LiF-NaF-ZrF_4 at 600°C with capillary fluorine inlet tubes.

2.2 Behavior of PuF_6 Volatilized in Fused Salt Tests

In all except the first run the data indicated that the bulk of the volatilized PuF_6 was trapped in dry fluoride beds consisting of NaF, LiF, or CaF_2 (Table 2). These materials appeared equally effective in the 25-100°C temperature range. CaF_2 was effective also at 400°C (run 11). There was some indication of a plutonium breakthrough with NaF at 400°C (run 13), and there was definite nonsorption on LiF at 400°C (runs 14 and 15).

2.3 Overall Plutonium Material Balances

A good material balance was obtained in most of the runs, not only for the total plutonium used in the test, but also for the part that was volatilized from the salt (Table 2). The latter was calculated on the basis of the final fused salt concentration, corrected for the amount of plutonium removed in fused salt samples. The material balances obtained appear to be reasonable in view of the small amount of initial plutonium used (100 μg) and of the large number of samples that had to be analyzed.

The data show that some plutonium was retained on all wall surfaces within the system. This was expected since such a small amount of plutonium was used. The experience of other workers with PuF_6 indicates that such loss is insignificant, on a percentage basis, when handling 50-100 g quantities.

2.4 Absorption as a Separation Method for PuF_6-UF_6 Mixtures

In one test the fluorine used in the volatilization step contained 7 mole % UF_6 (run 12). The absorption results (Table 3) show that it is possible to effectively separate PuF_6 and UF_6. In this test with a 7% UF_6-F_2 mix the final fused salt contained less than 100 ppm of uranium.

In a second test the initial fused salt contained 2.26% uranium (as UF_4) in addition to the usual plutonium spike (run 14). With a LiF bed at 400°C, however, the PuF_6 broke through to the following NaF bed. Similar plutonium behavior was evident in run 15 where no uranium was present.

Table 3. Relative Absorption Effects[a] for PuF_6 and UF_6

	Amount Absorbed							
	Plutonium, μg				Uranium, g			
Run No.	Walls	Bed[b] 1	Bed[b] 2	Bed[b] 3	Walls	Bed 1	Bed 2	Bed 3
12	3.7	62	0.1	<0.1	0	0.05	6.02	17.9
14	12	3.7	52	1.3	$<10^{-3}$	$<10^{-3}$	1.1	$<10^{-3}$

[a] Separation factors in run 12 for plutonium on bed 1 = 450 and for uranium on beds 2 and 3 = 330.

[b] See Table 2 for description of the absorption beds used in these experiments.

3.0 DISCUSSION

3.1 Fused Salt Volatilization

The conditions under which the above results were obtained do not duplicate the conditions that might be expected in actual processing of reactor fuels. For example, irradiated low-enrichment UO_2 might be expected to have a plutonium content of 5000 g/tonne after use as power reactor fuel. When this fuel is dissolved in fused salt, a reasonable uranium concentration would be about 5% with a plutonium concentration of 250 ppm, which is far above the level of 2 ppm used in this work. However, the adequate volatilization and recovery obtained at the 2 ppm level indicate that little trouble would be encountered at the higher level.

The PuF_6 volatilization process appears to be primarily a sweep-out or sparging action, as is also the case with UF_6 volatilizations at low concentrations (<1%).

In a typical run at a fluoride flowrate of 100 ml/min, the plutonium transfer from salt to gas in the first minute of operation (assuming a first-order rate effect) was ~0.3 μg. At a fused salt concentration of 250 ppm in actual fuel processing, the initial transfer value would be increased to 37.5 μg. This is still well below the value of about 500 μg in the first 100 ml of fluorine gas obtained by using data for the equilibrium $PuF_4 + F_2 \rightleftharpoons PuF_6$ at 150°C (2,3).

The data presented indicate that PuF_6 volatilization from fused salt is slow, but this does not mean that it is impractical as a processing technique. The superficial linear velocity of the fluorine gas in the reactor (1 in. dia) varied from 4 to 12 in./min, to give half-times of 2.5-4 hr. Probably shorter half-times would be achieved by increasing the gas flowrates to a superficial linear velocity of as much as 500 in./min. Flows of this magnitude have been used in the HF sparging of salt in the Oak Ridge Volatility Pilot Plant (5).

3.2 Absorption of PuF_6

The existence of chemical complexes of PuF_6 with LiF, NaF, or CaF_2 is indicated by the results. Although less than 100-μg quantities were used, they were trapped by these materials. It is dubious that this was due only to surface adsorption, to a hydrolytic mechanism, or to simple filtration of entrained material. The chemical complex concept, however, is consistent with the behavior of UF_6 with NaF, forming the complex $UF_6 \cdot 3NaF$ (6). An adsorption mechanism is unlikely due to the fact that the specific surface areas of these materials are 1 m^2/g or less. The hydrolytic mechanism is discounted because of the large excess of fluorine.

The similar behavior of PuF_6 to that of UF_6 in forming chemical complexes or compounds is supported by similar reactions of NaF with other hexafluorides, e.g. MoF_6, TcF_6, and NpF_6 (7). The dissociation pressures of the UF_6-NaF and MoF_6-NaF complexes over a wide temperature range have been studied, and similar work is needed on the other compounds. The UF_6-NaF, MoF_6-NaF, and probably the NpF_6-NaF complexes appear completely reversible. The behavior of PuF_6 with NaF and CaF_2 at 400°C indicates that these complexes might be irreversible under practical conditions. The breakthrough of PuF_6 in a LiF bed at 400°C, in contrast to the behavior at lower temperatures, shows that this complex might be more easily reversible than the others. UF_6 does not complex with LiF or CaF_2, whereas PuF_6 apparently does.

In the one test (run 12) with both PuF_6 and UF_6 entering the absorption bed train, the absorption of PuF_6 in the presence of a large excess of UF_6 further supports the view that a PuF_6-CaF_2 complex was formed. If surface adsorption had occurred, it is reasonable to assume that the UF_6 gas would have "washed off" the PuF_6 since the condensation and vapor properties of the two materials are similar: sublimation temperature of UF_6 56.5°C, boiling point of PuF_6 62.3°C.

4.0 EXPERIMENTAL TEST EQUIPMENT AND PROCEDURE

4.1 Fluorination Work

The equipment for the plutonium work was mainly nickel vessels connected by 1/4-in. copper tubing with compression-type tube fittings (Fig. 6). The nickel fluorinator was constructed from 1-in.-dia tubing and was 6 in. long. A 1/2-in. entry port was provided for introduction of salt and plutonium spike solution. The outlet was 1/4-in. nickel tubing about 6 in. long. In runs 1-16 the fluorine inlet was a 1/4-in. dip tube, welded into the side of the vessel, which extended down to about 1/4 in. from the reactor bottom. In runs 17 and 18 special capillary inserts were attached to the end of the dip tube before insertion and welding. The 8-g absorption traps were made from 1/2-in.-dia nickel tubing and were about 5 in. long. Nickel-wool plugs were used to retain the absorbent (12-20 mesh) in the trap. The 5-g absorption traps were slightly shorter and made with a level cut in

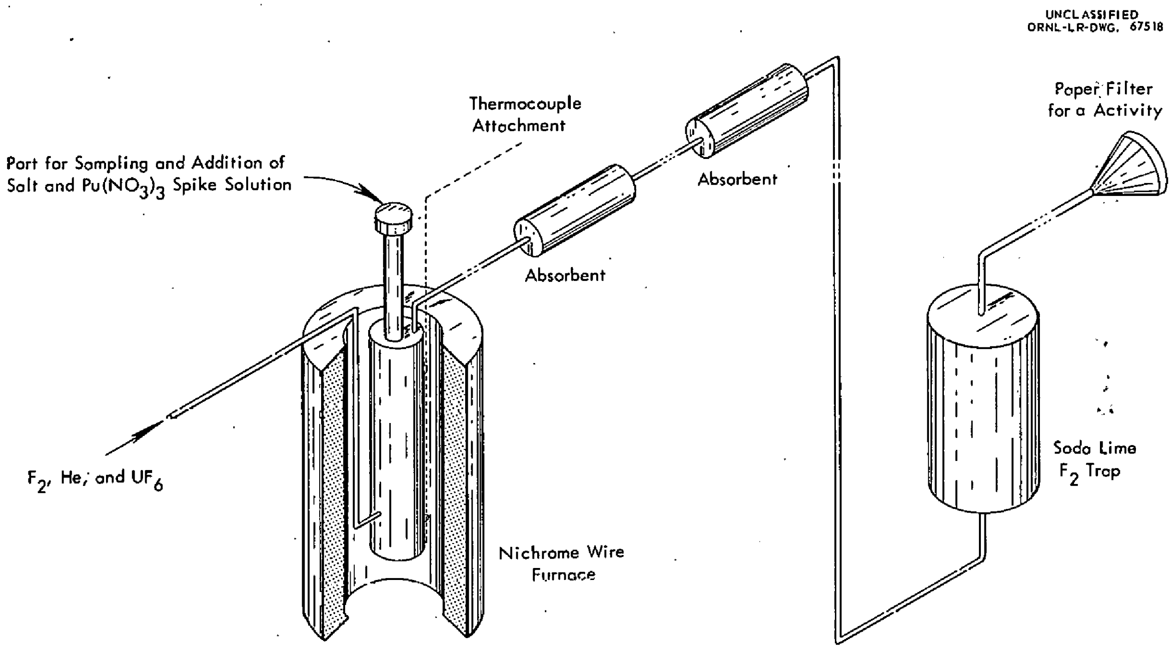

Fig. 6. Schematic of experimental equipment.

a V-form to eliminate the necessity of using nickel wool retainer plugs.

The procedure consisted in inserting the LiF-NaF-ZrF_4 or NaF-ZrF_4 salt, broken up into pea-size lumps, into the fluorinator, after which 100 µl of $PuO_2(NO_3)_2$ solution (~1 g Pu/liter) was placed directly on the salt, with care to avoid contact with the metal walls of the reactor. The reactor was then inserted in the furnace and connected to the gas tubing system. Heating was carried out slowly and carefully with a helium sparge to decompose the aqueous plutonium spike solution. After the reactor had reached the operating temperature of 500-600°C, the helium flow was stopped and fluorine flow was started through the by-pass circuit to condition the tubing, vessel walls, and absorption material. This was continued for about 1 hr. All the apparatus was at operating temperature during the conditioning period.

Salt samples were taken at intervals during fluorination. Each sampling was preceded by a short helium sparge, and this time was not counted in the total fluorination time. The salt samples (~0.5 g each) were taken with a 1/8-in.-dia nickel rod by the quick-freeze technique, i.e. by quickly inserting the cold rod and withdrawing it before the frozen salt could remelt. Experience with uranium and radioactivity determinations has shown that this is a reliable method of sampling since the frozen salt does not have a porous structure and the time involved (2-4 sec) is short.

A large safety trap containing ~1 kg of soda lime was placed at the end of the gas system to absorb the fluorine and to ensure that no plutonium would leave the system and contaminate the external working area. This worked efficiently, only one replacement being made over the entire series of tests. No plutonium α activity was ever detected on a paper gas filter placed at the exit of this trap.

The fluorine gas used in these tests was supplied by the Oak Ridge Gaseous Diffusion Plant. It was passed through NaF to remove 3-5% HF before use. The purity after this treatment was in the range 93-97%.

Gas flowrates were controlled and measured with 50-mil-dia capillary flowmeters, using 0.25 psi input differential pressure instruments to measure the Δp.

4.2 Analytical Methods

Suitable fluoride salt dissolution procedures had to be developed during the test runs because of difficulties in initial analytical tests in achieving reproducible results. Aluminum nitrate solution (1 M) was used initially to dissolve fluorination salt samples as well as absorbent bed fluorides. Erroneous and erratic results were obtained in using LaF_3 precipitation followed by TTA extraction to measure the plutonium α activity. Consultations with F. L. Moore and J. H. Cooper at ORNL indicated that the low plutonium analytical recoveries were due to Al^{3+} and F^- interference in LaF_3 precipitations and to ZrF_4 interference in counting due to its extraction by TTA.

A satisfactory analytical method found was to use dilute aqua regia as the solvent (method 3, below). However, even in this case the presence of dissolved ZrF_4, NaF, and LiF in the dilute aqua regia affected the plutonium α determinations. The percentage recovery appeared reproducible, however, and method 3 was therefore used with all types of material to obtain comparative but not absolute values.

Method 1: Filter Paper Technique. In the first fluorination tests the fluoride salts were dissolved in 1 M $Al(NO_3)_3$ solution (1 g of salt in ~20 ml of solution). Aliquots of these solutions were used successfully with a filter paper technique since results could be quickly obtained and the fluorinations could be monitored as they proceeded. Comparison of the filter paper results with LaF_3-TTA results indicated greater reliability of the former over the latter, and hence the filter paper results were used exclusively in the first three runs.

The filter paper technique consisted simply in slowly dropping 0.050 ml of the 1 M $Al(NO_3)_3$ solution onto a piece of 5-cm filter paper and allowing it to air dry at 80-90°. The plutonium α activity was then counted with a scintillation counter at 41% geometry.

Method 2: HNO_3 Dissolution. Dissolution of some of the absorbent bed materials (run 4) in 4 M HNO_3 gave accurate data by a standard LaF_3-TTA analytical method. However, since the method was not suited for use with fluorination salt samples, no further work was done with it.

Method 3: Dilute Aqua Regia Dissolution. Dilute aqua regia (4 M HNO_3-4 M HCl) at 95°C, in polythene containers, was suggested by C. J. Shipman as a general dissolvent for all the fluorides. In 10 analytical runs in which an aliquot spike of the standard $Pu(NO_3)_3$ solution in a synthetic salt solution (1 g of 31-24-45 mole % LiF-NaF-ZrF_4 in 25 ml of dilute aqua regia) was used, plutonium recoveries were 74.7, 77.9, 83.2, 81.5, 85.5, 74.3, 92.2, 95.6, 83.4, and 87.6%. The average was 83.6%, with a standard deviation of 6.65. The counting level in these tests was in the range 700-900 cpm/ml. The average recovery again demonstrated that some interference (probably from ZrF_4) was recurring in the analytical LaF_3 precipitation-TTA extraction procedure; however, the recovery was much higher than when aluminum nitrate solution was used, and the variation in error was low.

In fused salt runs 6-10, inclusive, a statistical test of the zero-time values for plutonium in the salt after spiking, melting, and helium sparging with duplicate sampling showed the same average recovery value of 83.6% with a standard deviation of 6.35. The values obtained were 77.5, 90.4, 79.1, 95.2, 89.3, 89.3, 78.6, 81.3, 77.0, and 78.6%.

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