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Combustion Aerosols Formed During Burning of Radioactively Contaminated Materials — Experimental Results

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EXECUTIVE SUMMARY

Pacific Northwest Laboratory conducted two series of experiments to measure aerosols generated by burning contaminated combustibles. Data from these experiments will replace or expand models currently in FIRIN, a fire compartment computer code that predicts source term releases from fires in nuclear fuel cycle facilities.

Four materials were burned in the combustible solid experiments: polychloroprene (PC), polystyrene (PS), polymethylmethacrylate (PMMA), and cellulose. In two runs a combination of fuels was burned. Contaminant form, configuration, and concentration; heat flux; percent oxygen air flow; and type of ignition were varied in the experiments. The highest releases, up to 4.5 wt% uranium, came from PMMA with powder contaminant. Cellulose burns had the lowest fraction releases, approximately 0.01 wt%. Most of the uranium released from PMMA and PS burns came off during the melting and bubbling preignition stage before the material burned. Uranium release rates from polychloroprene correlate well with smoke release rates. Cellulose seemed to give off uranium at a steady rate throughout the burn. Uranium release rates correlated well with mass loss rate for that material. One particle size distribution was measured for each of the fuels. For cellulosic material, more than 97% of the particles carrying uranium were less than 10 μm. However, only 16% of the particles less than 10 μm carried uranium for PC. Sizes of radioactive particles from burning contaminated PS and PMMA were between the two extremes.

A mixture of 30% tributyl phosphate in kerosene was burned in the contaminated combustible liquid experiments. The fuel was combined with nitric acid in the following four configurations:

1. acid/fuel with uranium
2. acid with fission products/fuel with uranium
3. acid with fission products and uranium/fuel
4. acid with fission products and uranium/fuel with uranium.

Weight percent of uranium airborne ranged from 0.2 to 7.1, with the highest releases coming from a burn of pure organic fuel over acid with fission products and uranium. In all configurations, the mass rate of uranium airborne seemed to be proportional to the mass rate of smoke airborne. Fission product analysis failed to provide accurate data, so it is not included in this report. One particle size distribution was measured. An aerodynamic mass median diameter of 0.6 μm was calculated for airborne particles containing uranium from burning contaminated combustible liquids.

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INTRODUCTION

Aerosol generation characteristics of accidental particle releases in nuclear fuel cycle facilities are being investigated at Pacific Northwest Laboratory (PNL). The work is sponsored by the U.S. Nuclear Regulatory Commission, Office of Nuclear Materials Safety and Safeguards. Safety analysis reports and environmental impact statements must evaluate the consequence of postulated accidents in or involving the facility in question. During an accidental release from a nuclear fuel cycle facility, the dominant pathway to man is usually through airborne particles, so it is necessary to determine an aerosol source term (i.e., the quantity of material initially airborne from an accident).

This report presents the results of two series of experiments investigating the release of radioactive aerosols in fuel cycle facility fires. The first series consists of 40 experiments involving combustible solids contaminated with uranium dioxide powder, uranyl nitrate hexahydrate (UNH) solutions, and UNH salt. The second series consists of 16 experiments involving combustible liquids [kerosene/tributyl phosphate (TBP) mixtures] with solutions of nitric acid. The acid contains uranium, cesium, strontium, and ruthenium, while the kerosene/TBP was used both in a pure state and contaminated with uranium.

The purpose of the experimental program is to provide time-dependent radioactive aerosol source term data for safety analyses and for the FIRIN computer code. FIRIN is a fire compartment source term code for fuel cycle facility fires. The current FIRIN radioactive source term models are based on the results of a series of experiments measuring the fractional release from specific types of fire accidents (Mishima and Schwendiman 1973a,b,c). The results obtained in these experiments were not time dependent and are applicable mainly to the accident configuration for which they were performed. The subject series of experiments was done to develop models that can be applied to many fire situations and to provide information on the rate and particle size of radioactive particles released in a fire.

Experimental procedures and results are discussed in this report for the burning of contaminated combustible solids and liquids.

EXPERIMENTAL PROCEDURES

Two series of experiments were performed to determine the rate and amount of airborne material and the aerodynamic particle size of the aerosols generated by burning contaminated combustibles. Four solid combustibles were burned in a set of 40 experiments in which contaminant form, configuration, and concentration; airflow; heat flux; and type of ignition system were varied. In a second series consisting of 16 experiments involving combustible liquids, a mixture of 30% TBP in kerosene was burned over a nitric acid solution. Four configurations were examined: acid/fuel with uranium, acid with fission products/fuel with uranium, acid with fission products and uranium/fuel, and acid with fission products and uranium/fuel with uranium. The apparatus, sample preparation, procedures, and analysis are described in the following sections.

APPARATUS

The experimental apparatus used in the burn experiments is called the Combustion Aerosol Release Equipment (CARE). Figure 1 is a diagram of the equipment. An adjustable-height aluminum plate supports a milled aluminum base. The base was machined to provide support for a 0.8-m-long, 18-cm inside-dia quartz tube. Burning of solid combustibles takes place in a shallow pan positioned approximately midway in the tube. Air (or oxygen) enters the base and travels through a 13-cm layer of glass beads, which disperse and equalize the airflow in the tube. The combustion gases exit through the top of tube and are drawn into a hood where they are diluted with air. The combustion gases and air enter an insulated pipe and the filtration manifold. Four 8- x 10-in. filter holders in the manifold sequentially sample the aerosol in a predetermined order. After leaving the manifold, the gas velocity is measured with a hot wire anemometer. A High-Efficiency Particular Air (HEPA) filter bank is located downstream to trap any remaining aerosol. Except for the filtration manifold, the CARE was designed to be similar to small-scale burn apparatus at Factory Mutual Research Corporation, Norwood, Massachusetts (Steciak, Tewarson, and Newman 1983).

The CARE is located within the Radioactive Aerosol Release Tank (RART), which acts as a secondary containment system. The RART is a stainless steel cylindrical enclosure approximately 3 m high and 2.9 m in diameter with a 20-m^3 volume, about the size of a small room.

Radiant heat flux to simulate nearby fires or other heat sources is provided by four radiant panels mounted symmetrically around the quartz tube above the sample. A radiometer is used to calibrate the output of these panels.

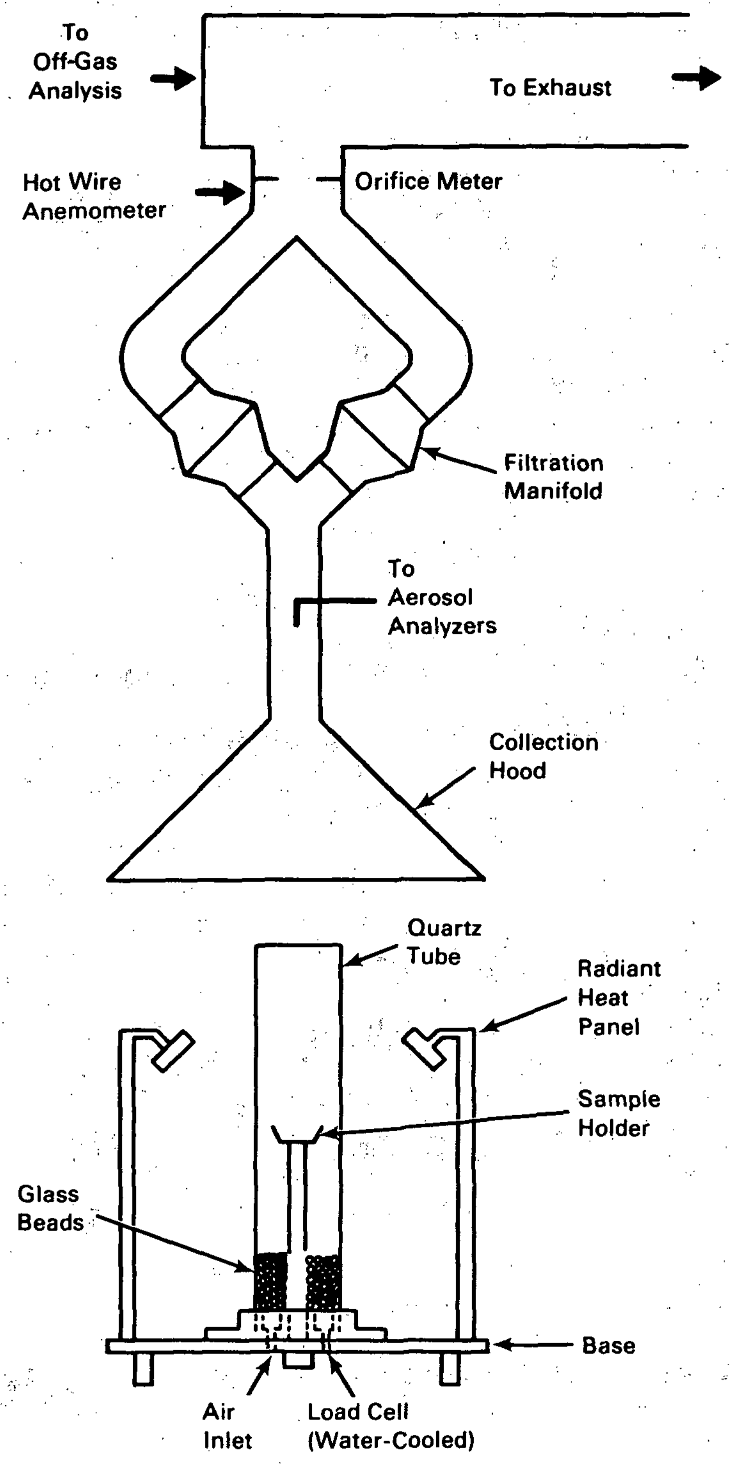

FIGURE 1. CARE Combustion Apparatus

For combustible liquids, the apparatus was modified slightly. A metal or glass beaker was used in place of the shallow pan. A heat tape was used to provide an external heat flux to the liquid. A single radiant panel provided external heat flux to the top surface of the liquid in several experiments.

The mass loss rate of the burning material is measured by a load cell coupled to the sample holder. Aerosol number concentration and particle size distribution is measured using an optical particle counter. A cascade impactor is also used for particle sizing. Data from the particle analyzer are fed to a computer. Combustion gases are analyzed with real-time analyzers for oxygen, hydrocarbons, carbon dioxide, and carbon monoxide. Temperature profiles are measured by thermocouple rakes located above the quartz tube and in the off-gas ducts. Data from the off-gas analyzers and thermocouples are fed to a data logger.

Air flow in the apparatus varies with location. Air velocity in the large duct below the filtration manifold is about 15.24 m per minute (fpm). This duct has a diameter of 25.4 cm, so the total airflow is about 0.013 cubic meters per second (m^3/s). This number is fairly constant throughout the apparatus, although there is some air leakage around the valves in the filtration manifold. The highest air velocities are in the 15-cm ducts directly below each filter holder. The air velocity here is about 0.066 m^3/s. The lowest air velocity, 0.002 m^3/s, occurs across the face of the 0.9-m outside-dia (OD) dilution hood.

COMBUSTIBLE SOLID EXPERIMENTS

Parameters for the combustible solid experiments are shown in Table 1. A total of 40 experiments were performed.

Experimental Parameters

Four combustible materials were burned in these experiments; each is listed below along with its use in nuclear fuel cycle facilities.

- polymethylmethacrylate (PMMA)--glove box walls, hood windows
- polychloroprene (PC)--rubber gloves, window gasketing
- polystyrene (PS)--ion exchange resins
- cellulose--paper towels, rags, glove box liners.

The contaminant material used in all the experiments was depleted uranium in various chemical forms. Depleted uranium dioxide has been shown to be a suitable surrogate for plutonium and mixed oxides in previous studies (Sutter 1983). The depleted uranium was used as a powdered dioxide, in solution with nitric acid, and as a salt of uranyl nitrate hexahydrate.

TABLE 1. Parameters for Contaminant Combustible Solid Experiments

Run#	Material	Contaminant	Heat Flux, %	Ignition[(a)]	Oxygen, %	Comments
3	cellulose	liquid	100[(b)]	Y	21	
4	cellulose	powder	100	N	21	
5	cellulose	liquid	50	N	21	
6	PMMA	powder	100	Y	21	
7	PC	powder	100	Y	21	
8	PC	liquid	100	Y	21	
9	PC	powder	100	N	21	
10	PMMA	liquid	100	Y	21	Data incomplete
11	PC	powder	70	Y	21	
12	cellulose	powder	100	Y	21	
13	PMMA	powder	100	N	21	
14	PMMA	salt	100	Y	21	
15	cellulose	powder	70	Y	21	
17	PS	liquid	100	Y	21	
18	cellulose	liquid	100	Y	21	
19	PMMA	liquid	100	Y	21	
20	cellulose	salt	100	N	21	
21	PMMA	liquid	100	Y	21	
22	cellulose	salt	70	Y	21	
23	cellulose	powder	100	Y	21	
24	PC	salt	100	Y	21	
25	PS	liquid	100	Y	21	
26	cellulose	powder	10	Y	12	
27	cellulose	powder	100	Y	6	
28	cellulose	powder	100	Y	6	High air flow
29	PMMA	powder	100	Y	21	Thick layer
30	PMMA	powder	100	Y	21	Thick layer
31	PMMA	powder	100	Y	21	Pile
32	PMMA	powder	100	Y	21	Pile
33	PMMA	powder	100	Y	21	PMMA over powder
34	PMMA	powder	0	N	21	Background
35	cellulose	powder	0	Y	21	Data incomplete
36	PMMA	powder	100	Y	21	Data incomplete
37	cellulose	powder	0	Y	21	Data incomplete
38	PS	liquid	100	Y	21	Data incomplete
39	cellulose	powder	0	N	21	Background
40	mixed	powder	100	Y	21	Data incomplete
41	mixed	liquid	100	Y	21	Data incomplete
51	PMMA	powder	0	Y	21	
55	PS	liquid	100	Y	21	

(a) Y indicates an ignitor was used, N indicates that no ignition system was used.

(b) 100% heat flux is 24 kW/m^2.

External radiant heat fluxes of 13 and 24 kW/m^2 were used to simulate various-sized fires. Oxygen concentrations were kept at 21% (ambient) except for three experiments (26, 27, and 28). Twelve percent oxygen was used in run 26. Six percent oxygen was used in runs 27 and 28. In one run air flow through the tube was raised from 8 Lpm to 11 Lpm.

Two different ignition systems were used. Both systems used a methane gas flame to provide ignition, but one nozzle gave a flame approximately 2.5 cm above the surface of the sample, while the other nozzle could be adjusted to give a flame that impinges on the sample surface. Using the first nozzle, ignition occurred when a flammable mixture of vapors formed above the sample and was ignited, causing flashover to the surface. With the second nozzle, the flame impinging on the surface caused higher surface temperatures and higher vapor generation rates than the first nozzle and also ignited the vapors as soon as they left the surface of the sample. Much shorter ignition times were observed with the second nozzle.

Sample Preparation

The configuration of combustible and contaminant can have a significant effect on uranium release. Configuration was varied for each contaminant material and is described in the following paragraphs. Specific details for individual runs are also given.

Three contaminant materials were used in these experiments. The first was a depleted uranium dioxide (DUO) powder. Previous work by Sutter, Johnston, and Mishima (1981) using sedimentation methods characterized the powder as having a mass median diameter (MMD) of 1.0 µm and a σ_g of 2. Table 2 gives the particle size distribution of the powder. Scanning electron microscope (SEM) measurements of the agglomerates on a PMMA slab were also made and are shown in Table 2. SEM measurements show an MMD of 5.0 µm and a σ_g of 2.

The second contaminant material was a solution of uranium as UNH in nitric acid. Uranium concentration is 208.7 g/L (Sutter, Johnston, and Mishima 1981). Other properties characterizing UNH include a density of 1.54 g/cm^3, viscosity of 1.70 cp, and a surface tension of 62.88 dyne/cm (Sutter 1983).

The third contaminant material was an air-dried salt cake of the UNH solution. The cakes were prepared by placing a known amount of UNH solution on the combustible material and placing the sample in a warm oven with forced air circulation. The samples were left in the oven until no liquid was visible and then allowed to sit at room temperature for at least a day. The uranium is presumably in the form of UNH crystals, but the exact amount of hydrated water is not known.

TABLE 2. DUO Particle Size Distribution

Size, µm	Measured Cumulative Mass[a], %	Measured Cumulative Mass[b], %
20	97	96
10	95	92
8	94	82
6	93	60
4	92	32
2	85	13
1	50	2
0.8	40	0.2

(a) Sutter, Johnston, and Mishima (1981) - liquid sedimentation.

(b) SEM analysis of agglomerators on PMMA Slab.

Four combustible materials were used in these experiments: cellulose, PS, PC, and PMMA. Cellulose was used in the form of paper towels. Preliminary testing showed that about 1 g of powder could be picked up on each towel using reasonably good laboratory practice. This amount was used as an upper limit for the amount of powder placed on a towel. Other tests showed that up to 60 cc of UNH solution saturated a towel. A towel that was wrung out before disposal might have about 20 cc of solution still in it. These numbers were used in setting the amount of contaminant solution to add to each towel. Each towel was 0.3 x 0.4 m and folded in quarters. Towels contaminated with powder were usually contaminated on one side only and then folded to expose only uncontaminated sides. Towels soaked in liquid were crumpled to a ball shape before burning. Towels with air-dried solutions were left flat and folded in half again (a total folding into eighths).

Polystyrene was burned in the form of small angular beads. Enough UNH solution was added to each pile of beads to coat them without leaving free liquid in the void space.

Polychloroprene was cut in the form of approximately 8- x 8-cm pieces out of an actual glove box glove. The PC was placed flat in the bottom of the burning pan and contaminated to a level of 7.5 g/m^2 uranium, the amount corresponding to a coating of powder visible to the unaided eye (Mishima, Schwendiman, and Ayer 1979, p. 44).

Polymethylmethacrylate was used in the form of 5-715-cm-dia circles of 1.27-cm-thick Plexiglas®. This particular form of PMMA not only supports combustion, but is also combustible. The PMMA normally used in glove box windows will not support combustion by itself but can supply fuel. The contaminant material was poured or spooned onto the top surface of the PMMA slab and then either spread out with a spatula to obtain an approximately even layer or left in a pile. In an actual facility the contaminant material would probably settle out of the air onto the surface. The amount of powder contaminant used ranged from 0.02 (a level of 7.5 g/m^2) to 1.7 g. Liquids were poured on the surface to form pools of various thicknesses.

The following is a description of the sample preparation for each run in which data were successfully collected. Amounts of combustible material and contaminant material, and a brief description of the preparation process are given.

- Run #3 - Three paper towels were soaked in 100 mL of UNH solution. The towels were crumpled.
- Run #4 - Three paper towels were contaminated with ~1 g of DUO each. The DUO was placed in a stainless steel pan and wiped on the surface of the towels. The towels were then crumpled to expose only uncontaminated surfaces.
- Run #5 - Three paper towels were soaked in 75 mL of UNH solution and crumpled.
- Run #6 - A 7.6-cm-dia, 1.3-cm-thick PMMA slab was coated with 0.05 g of DUO. The DUO was weighed out, placed on the surface of the slab, and evenly spread with a spatula on the surface.
- Run #7 - A piece of PC, approximately 8 by 8 cm, was coated with 0.08 g of DUO. The DUO was weighed out, placed on the surface, and spread with a spatula.
- Run #8 - 5 mL of UNH solution were placed on a piece of PC. Because the PC was larger than the bottom of the burning pan, the UNH collected at the bottom, giving non-uniform coverage of the PC surface.
- Run #9 - Sample preparation same as Run #7.

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- Run #10 - A 5.7-cm-dia, 1.3-cm-thick PMMA slab was coated with 5 mL of UNH solution. The solution covered the whole surface in a continuous layer.

- Run #11 - Sample preparation same as Run #7.

- Run #12 - Sample preparation same as Run #1, except that 0.3 g DUO per towel was used.

- Run #13 - Sample preparation same as Run #6, except that a 5.7-cm-dia PMMA slab with 0.02 g DUO was used.

- Run #14 - A PMMA slab similar to that of Run #10 was coated with UNH solution, placed in a low-temperature forced convection oven for several days, and air dried in a ventilated hood for several more days. Approximately 1.2 g of UNH salt were deposited in a fairly even layer over the surface of the slab.

- Run #15 - Sample preparation same as Run #1.

- Run #17 - 35 g of PS beads were soaked in 6 mL of UNH solution. No free liquid was observed on the surface of the beads.

- Run #18 - One fourth of a paper towel was soaked in 10 mL of UNH solution. The remaining 3/4 of the towel was left dry and uncontaminated and was wrapped around the wet 1/4 towel. Another uncontaminated towel was crumpled and placed next to this towel in the burning pan.

- Run #19 - Sample preparation similar to Run #10 except that 2 mL of UNH solution were used.

- Run #20 - A paper towel was soaked in 8 mL of UNH solution, placed in a low-temperature forced convection oven for several days, and air dried in a ventilated hood for several days. The towel has ~3.5 g UNH salt in it. The now stiff towel was broken in half and the two halves piled on top of each other in the burning pan.

- Run #21 - Sample preparation same as Run #10 except that 0.1 mL UNH was used on a 5.7-cm-dia PMMA slab.

- Run #22 - Sample preparation similar to Run #20.

- Run #23 - Sample preparation similar to Run #1 except that two towels and 2 g DUO were used.

- Run #24 - A piece of PC, approximately 8 x 8 cm, was coated with 3 mL UNH, placed in a low-temperature forced convection oven for several days, and air dried in a ventilated hood for several days. The PC had ~1.5 g of UNH salt on it.

- Run #25 - Sample preparation similar to Run #17 except that 10 g of PS beads and 4 mL of UNH solution were used.

- Run #26 - Sample preparation similar to Run #23.

- Run #27 - Sample preparation similar to Run #23.

- Run #28 - Sample preparation similar to Run #23.

- Run #29 - Sample preparation similar to Run #13 except that 0.44 g DUO was used.

- Run #30 - Sample preparation similar to Run #13 except that 0.36 g DUO was used.

- Run #31 - Sample preparation similar to Run #13 except that 1.13 g DUO were used in a pile.

- Run #32 - Sample preparation similar to Run #13 except that 1.35 g DUO were used in a pile.

- Run #33 - Sample preparation similar to Run #13 except that 1.66 g DUO were placed under slab.

- Run #34 - Sample preparation similar to Run #13 except that 0.05 g DUO was used.

- Run #35 - Sample preparation similar to Run #1.

- Run #36 - Sample preparation similar to Run #34.

- Run #37 - Sample preparation similar to Run #35 except a different brand of paper towel used.

- Run #38 - Sample preparation similar to Run #17 except that 6 g of PS beads and 3 mL of UNH solution were used.

- Run #39 - Sample preparation similar to Run #1.

- Run #40 - 1 g of DUO was placed on a paper towel, and 1 g DUO was placed on 15 g of PC.
- Run #41 - 25 mL of UNH liquid was absorbed by a paper towel, and 3 mL UNH liquid was placed on a PMMA slab.
- Run #51 - Sample preparation similar to Run #13.
- Run #55 - Sample preparation similar to Run #17 except that 25 g of Amberlite IRA-900 ion exchange resin was soaked in 15 mL of UNH solution.

COMBUSTIBLE LIQUID EXPERIMENTS

The complete combustible liquid experiments are shown in Table 3. A total of 15 experiments were performed, and complete sets of data were obtained for 11 experiments.

The combustible liquid used in these experiments was a mixture of 30% TBP in normal paraffin hydrocarbon (NPH) (a kerosene-like diluent). This liquid was chosen because of its use as a solvent in the plutonium uranium reduction oxidation extraction (PUREX) process. Other combustible liquids that might be found in fuel cycle facilities include fuel oils, lubricating oils and greases, and other solvents. Properties of the NPH solvent (Mishima and Schwendiman 1973b) are:

Specific Gravity	60/60°F or 15.6/15.6°C	0.76
Viscosity at 25°C		1.8 cp
Flash point		70°C

The TBP/NPH mixture was burned as a layer over a ~3 M nitric acid solution. Nitric acid is present in large quantities in fuel reprocessing facilities, and it was felt that in many accidents both nitric acid and the TBP/NPH mixture could be involved.

The TBP/NPH mixture was used in both a "pure" form (no simulated radioactive contamination) and containing 101.2 g/L depleted uranium. The nitric acid was used in pure form, containing simulated fission products (as listed in Table 3), and containing both simulated fission products and depleted uranium.

The TBP/NPH mixture was placed over a layer of nitric acid in a glass or metal beaker. A heat tape was wrapped around the outside of the beaker to

TABLE 3. Contaminated Combustible Liquid Experiments

Run #	Acid Phase	Organic Phase	Comments
42	pure	pure	Data incomplete
43	pure	pure	Data incomplete
44	pure	spiked (a)	Data incomplete
45	pure	spiked (a)	
46	pure	spiked (a)	
47	spiked (b)	spiked (a)	
48	spiked (b)	spiked (a)	
49	spiked (b)	pure	
50	spiked (c)	pure	Data incomplete
52	spiked (c)	pure	
53	spiked (c)	pure	
54	pure	pure	
56	spiked (c)	spiked (a)	150 mL acid/50 mL organic (d)
57	spiked (c)	spiked (a)	100 mL acid/50 mL organic
58	spiked (c)	pure	150 mL acid/50 mL organic

(a) Organic phase contains 101.2 g/L uranium.
(b) Contains 0.16 g/L ruthenium, 0.50 g/L cesium, 1.76 g/L strontium.
(c) Acid phase contains 0.43 g/L ruthenium, 1.34 g/L cesium, 4.69 g/L strontium, 188 g/L uranium.
(d) 100 mL acid/100 mL organic in other experiments.

provide initial vaporization of the TBP/NPH. Ignition was provided by a methane flame located approximately 8 cm above the beaker. Oxygen concentration was not controlled but was left at ambient (~21%).

All of the combustible liquid experiments were made using combinations of the five liquids described below. In each experiment, one organic liquid was paired with one inorganic acid.

The liquids were mixed to specification and have the following compositions:

- 30% TBP in NPH - This is the primary organic solvent used in the PUREX process.
- 30% TBP in NPH with 101.2 g/L uranium - This solution represents the used solvent that contains separated heavy metals.
- 2.5 M nitric acid - The primary dissolution acid in the PUREX process.

- 2.54 M nitric acid with 188 g/L uranium, 0.43 g/L ruthenium, 1.34 g/L cesium, and 4.69 g/L strontium - This solution simulates the PUREX acid solution after dissolution and before contact with the organic phase.

- 2.88 M nitric acid with 0.16 g/L ruthenium, 0.50 g/L cesium, and 1.76 g/L strontium - This solution simulates the PUREX acid solution after contact with the organic phase.

Any of these liquids could be present in an accident in a fuel reprocessing facility. The organic liquids are the only combustible liquids, and only the NPH fraction is combustible. The acid phases were included in these experiments to see if fire and heating would cause migration of the fission products or transuranic elements across the phase boundaries. For example, as a contaminated organic liquid burns, some of the contaminant (uranium in this case) may be driven out of the organic and into the underlying acid phase. The driving force for this would be the change in solubility of uranium in the organic liquid because of changes in temperature and composition.

The liquids chosen for each experiment were placed in metal or glass beakers, and the beakers were wrapped in heat tapes. In some cases, a heat lamp was also used to provide heat flux; however, external heat flux was not a measured or controlled variable in these experiments.

ANALYSIS OF FILTER SAMPLES FOR URANIUM AND FISSION PRODUCTS

A mixed acid leach solution was prepared in the following manner: approximately 400 mL of deionized water were placed in a 1-L flask. Approximately 310 mL of 5N HNO_3 were added to the flask and swirled. Two mL of 0.05N HF were added and swirled. The mixture was allowed to cool to room temperature before adding water to bring the flask to volume.

Each filter was placed in a large glass beaker and covered with 300 mL of the mixed acid leach solution. A stirring rod was used to help break up the filter. Beakers were placed in a water bath at 45 to 55°C for approximately 30 h to aid filter digestion.

Filters were separated from the leachate by vacuum filtration using a large buchner funnel. Leachate was poured into a 500-mL volumetric flask and brought to volume with deionized water.

The amount of uranium in the samples was analyzed using laser fluorometry. This method employs a pulsed nitrogen laser to excite uranium in a solution containing a pyrophosphate reagent. The fluorescent signals are amplified and integrated, and the results are displayed on a meter.

Samples from the contaminated combustible liquid experiments underwent additional analysis to detect ruthenium, cesium, and strontium. Atomic absorption was used for the cesium analysis, while inductively coupled plasma was used to detect ruthenium and strontium.

A separate extraction process was used to separate uranium and fission products from the organic leftover after some of the burns. In this process 10 mL of organic sample were placed in a 100-mL volumetric flask with 20 mL of a solution containing 64.05 g of ammonium carbonate and 27.54 g of sodium bicarbonate/L. The solution was heated to 75°C in a water bath for about 30 h with air bubbling through each sample. In some samples a precipitate occurred. Approximately 30 mL of 2 M HNO_3 were added to these samples and shaken. The aqueous and organic phases of the sample were then separated by decanting, and the volume of the aqueous phase was brought up to 100 mL with 2 M HNO_3.

In all the samples in which fission products were involved in the burn, mass balances of the release and residue indicated a recovery of more than 200% for cesium and strontium and a total lack of ruthenium. The ruthenium absence may be caused by the formation of a ruthenium oxide precipitate in the analytical samples. The excess of cesium and strontium could be attributed to contamination in the sample solution interfering with analysis. Because of the unreliability of the analysis data, fission product release is not presented in this report.

RESULTS

Weight percent airborne ranged from less than 0.01 to 7. Figure 2 shows the range of values for each of the contaminated fuel types burned. As shown by the figure, PMMA gave the highest releases of the solid burn experiments.

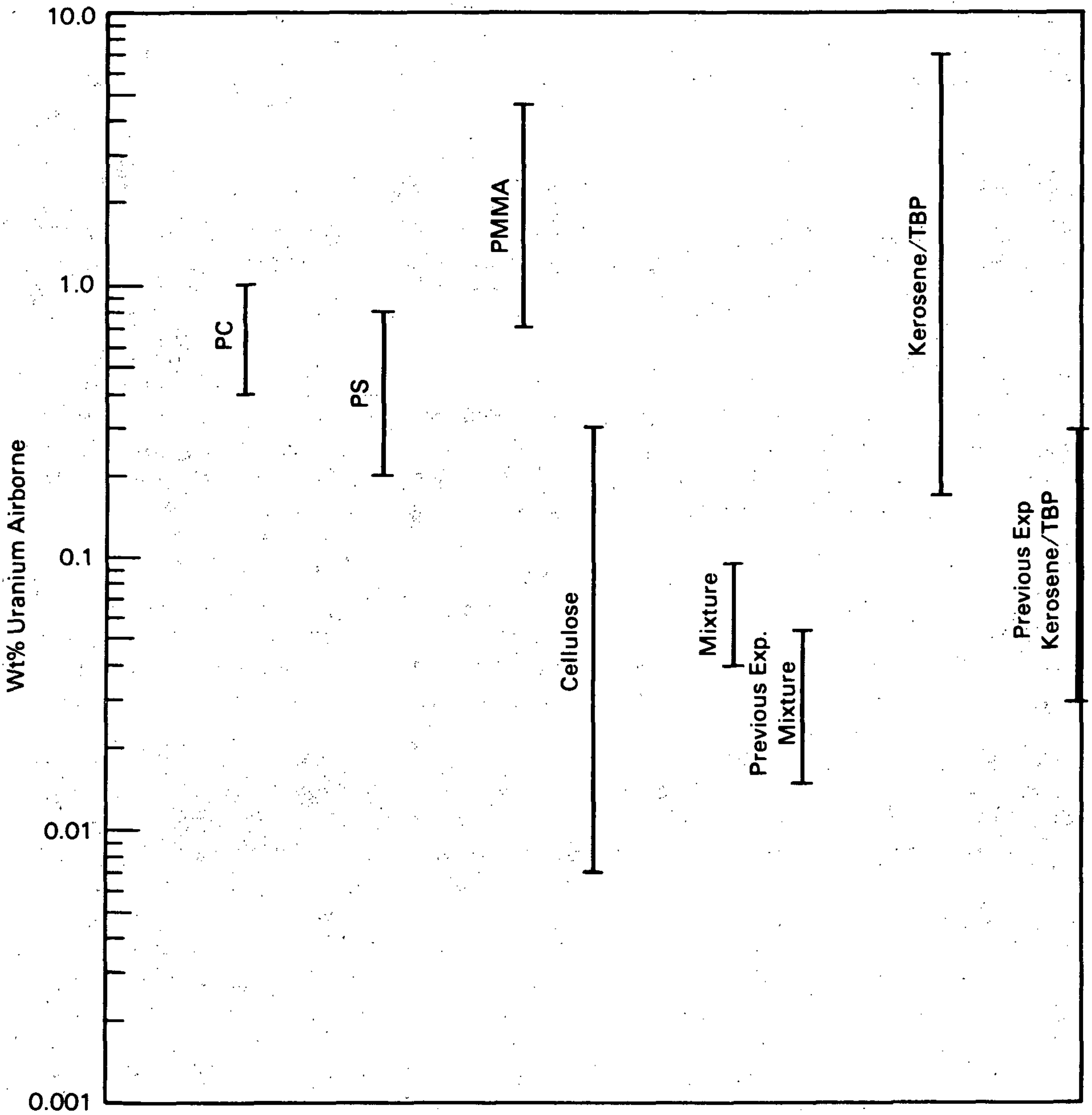

FIGURE 2. Results from Burning Contaminated Combustibles

Cellulose burns gave the lowest releases. Fuel mixtures both from these and previously reported experiments (Mishima and Schwendiman 1973a) gave airborne releases of uranium similar to those of the cellulose burns.

Uranium released from the contaminated combustible liquid burns reached as high as 7%. Much higher releases were achieved in this set of experiments than in similar previously reported experiments (Mishima and Schwendiman 1973b).

Because uranium release is highly dependent on fuel type, results are discussed and presented by fuel type in the following sections. Detailed data on cumulative fractional uranium and smoke releases are given in Appendix A.

COMBUSTIBLE SOLIDS

Results are presented for 46 experiments in which combustible solids were contaminated, then burned. Results are presented by fuel type in the following order: PC, PS, PMMA, cellulose, and mixed fuels.

Polychloroprene (PC)

Five runs (#7, #8, #9, #11, and #24) burned PC contaminated with either DUO powder, UNH solutions, or UNH salt. The smoke fractional releases (weight of smoke produced divided by weight of fuel burned) ranged from 7 to 15%. The uranium fractional release ranged from about 0.4 to 3.5%.

Run #8 was the only PC run with UNH solution. The uranium fractional release of 3.5% was the highest of all contaminant forms, as shown in Figure 3. Most of the uranium was released during the flaming combustion period (between the first and third data points on the graph). The release seems to correlate fairly well with the smoke production rate.

Runs #7, #9, and #11 burned PC with DUO powder. These runs are shown in Figure 4. Runs #7 and #9 were identical except that an ignitor was used in Run #7. These two runs were virtually identical in smoke production and were close to each other in uranium fractional release. The ignited run had a uranium fractional release of 1%, while the release for no ignition was 0.7%. Both runs showed close correlation between uranium and smoke release rates. Appreciable amounts of both smoke and uranium were released before sample ignition. The difference between uranium fractional release for these two runs is small, so the presence of an ignition system may not be important. In these cases, the PC burned for only 30 s even when an ignitor was used. This time is small compared to the 7 min in which the material smoldered and changed shape before flaming combustion.

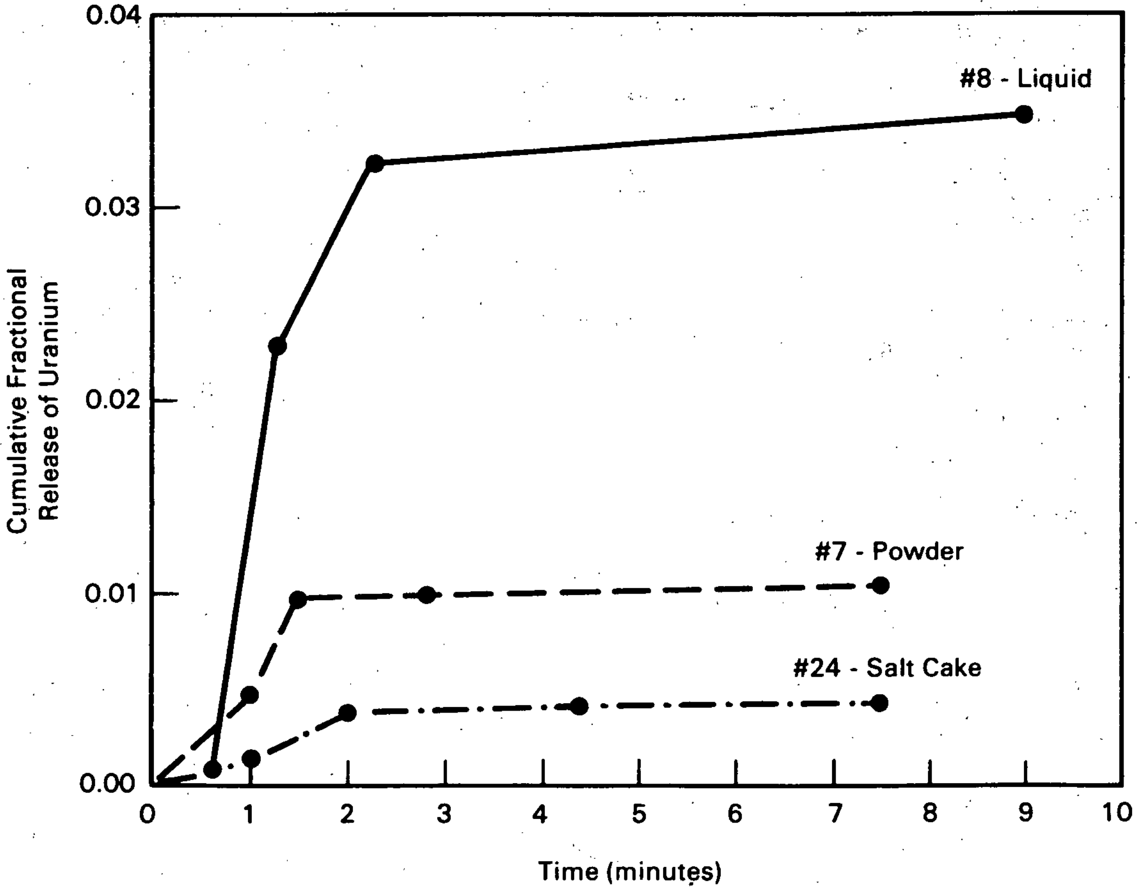

FIGURE 3. Fractional Release of Uranium from Contaminated Polychloroprene

The third powder run, Run #11, was similar to the first two runs with powder, but a lower heat flux was used in the early part of the run. A lower uranium fractional release of 0.4% resulted. The heat lamps were run at 70% power for the first 13 min of the run. The lamps were then increased to 100% power, and ignition occurred 25 s later. The sample flamed for 45 s and then smoldered for about 1 min. The majority of uranium and smoke was released during the time of the lower heat flux; that is, before ignition.

Run #24 burned PC with an air-dried UNH salt cake. The uranium fractional release of 0.4% from this run was lower than the corresponding run with powder contamination. Cohesive forces between the particles in the salt cake are probably greater than cohesive forces in the loose powder. The salt cake may also have greater cohesion to the PC. Most of the uranium release occurred during the flaming period. Uranium release also appears to correlate well with smoke production.

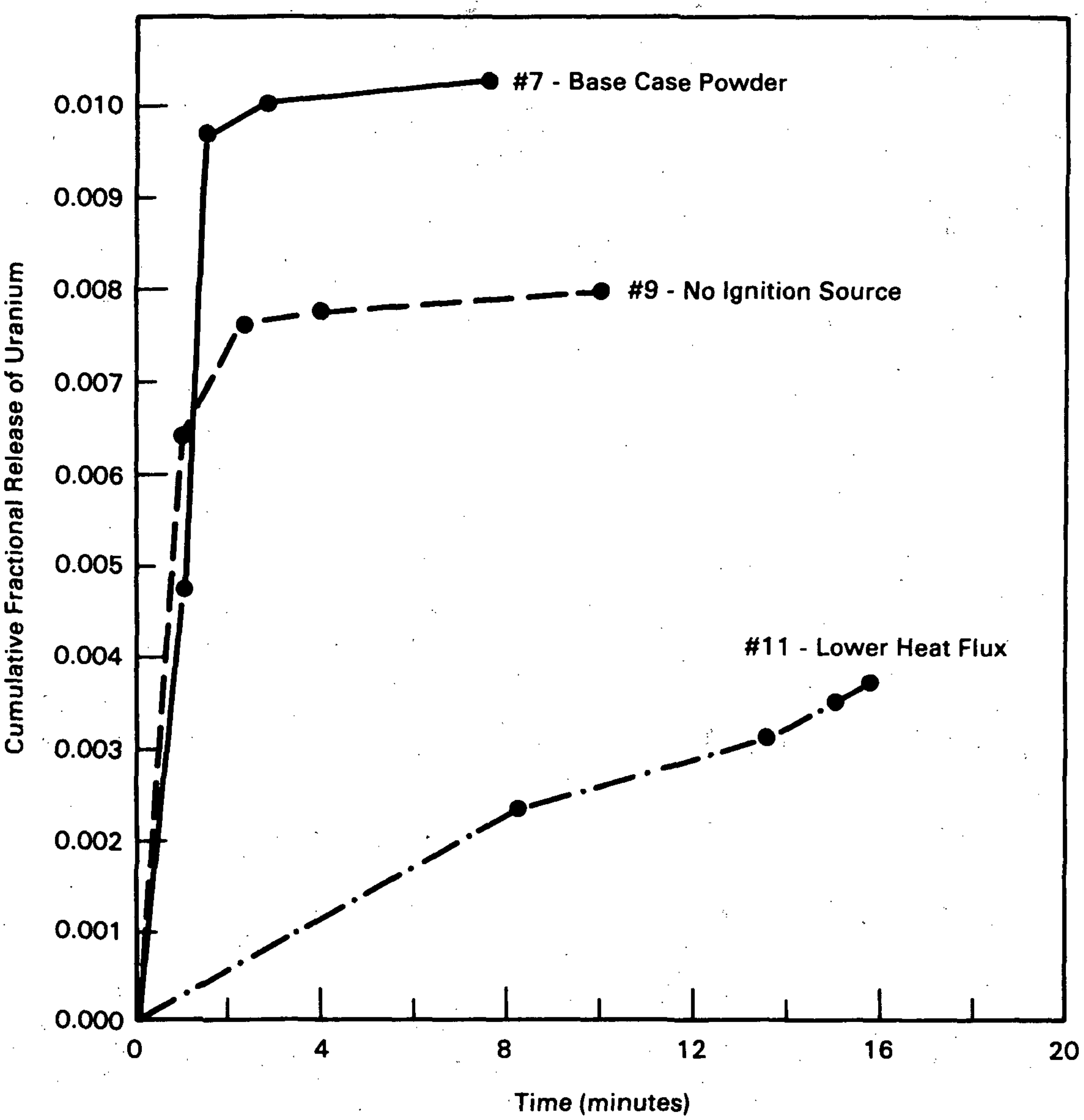

FIGURE 4. Fractional Release of Uranium from Polychloroprene Contaminated with Powder

Polystyrene (PS)

Four runs (#17, #25, #38, and #55) burned PS beads covered with UNH solution. The uranium cumulative fractional release is shown in Figure 5.

Run #25, with a contaminant concentration more than twice that of Run #17, had a uranium fractional release only slightly higher (0.18% versus 0.16%). Run #25 was considerably smokier than Run #17 in terms of smoke fractional

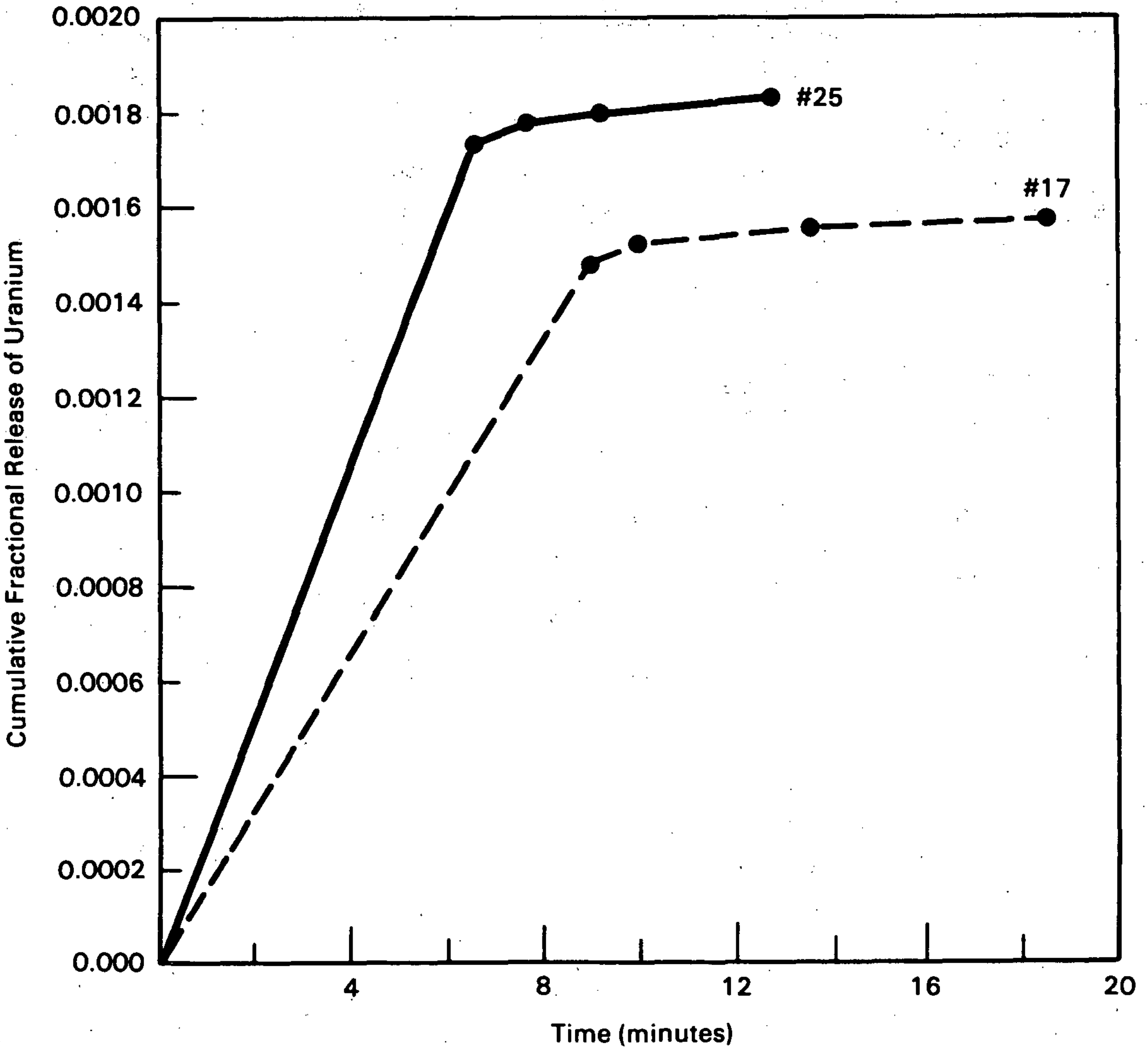

FIGURE 5. Fractional Release of Uranium from Polystyrene Contaminated with Liquid

release, although both runs provided considerable amounts of smoke. A much larger amount of PS was used in Run #25, which could lead to an underventilated, smokier fire. Run #38 had an even higher contaminant concentration and gave a uranium fractional release of 0.8%. Thus, contaminant concentration appears to have some effect on the fractional release of uranium, with higher concentrations giving higher releases.

Essentially all the uranium was released prior to ignition of the PS. Because half (or less) of the mass loss in each run occurred before ignition, it appears that the uranium release rate will not correlate well with mass loss rate. The uranium release rate does not appear to correlate with smoke release rate, either. Most of the mass loss before ignition is caused by evaporation of water from the UNH solution. Apparently, it is this mass flux that is transporting the uranium. Just before ignition, the mass of PS beads and UNH solution melted to a thick, viscous mass of molten PS that had the appearance of mincemeat. The fact that little uranium comes off after ignition would tend to indicate that the majority of uranium has sunk down into the bulk molten mass and been trapped inside. The uranium is therefore not released and remains in the residue left over after the fire burns out.

One observation is that while the uranium and smoke release rates do not correlate well, the total uranium fractional release correlates reasonably well with the total smoke fractional release. This could be useful for estimating uranium release in situations where the rate of release is not important.

Run #55 was made with actual ion-exchange resin rather than just PS beads. It was anticipated that the difference in structure between commercial PS beads and specially designed, cross-linked resins might have an effect on the release. Preliminary lab results indicate that the uranium fractional release is about 0.1%, or about the same as Run #17, which had approximately the same contaminant concentration. Thus, the type of resin is probably not an important factor in the release of uranium in PS fires.

Polymethylmethacrylate (PMMA)

Fourteen runs (#6, #10, #13, #14, #19, #21, #29 through #34, #36, and #51), burned PMMA contaminated with UNH solutions, uranium dioxide powders, and dried UNH solutions. Smoke fractional release ranged from 0.6 to 1.2%, while uranium fractional releases ranged from about 0.25 to 4.5%. The cumulative fractional release of uranium versus time is shown in Figures 6 and 7.

Runs #6, #13, #29 through #34, #36, and #51 burned PMMA with powder contaminant. The first two runs were essentially identical except that Run #6 used an ignition system and Run #13 did not. The uranium fractional releases were 3.2 and 3.0% for Runs #6 and #13, respectively. The difference is slight and is probably caused by experimental variation. Runs #29 through #33 were done as a special study of the PMMA-powder system. Runs #29 and #30 used a layer of DUO similar to Run #6, but much thicker. Runs #31 and #32 used a pile of DUO in the center of the PMMA slab as opposed to a layer. Run #33 used a layer as in Run #6 but for this run the PMMA was placed on top of the layer of DUO. The uranium fractional releases were 3.5% for the runs with thick layers of powder, 4.5% for the runs with piles of powder, and about 1.3% for the single run with DUO under PMMA. Run #34 was a background run involving PMMA and powder but no ignition system. In the absence of a flame, 0.2% of the powder was removed. This amount seems to represent aerodynamic entrainment by the air flow in the system.

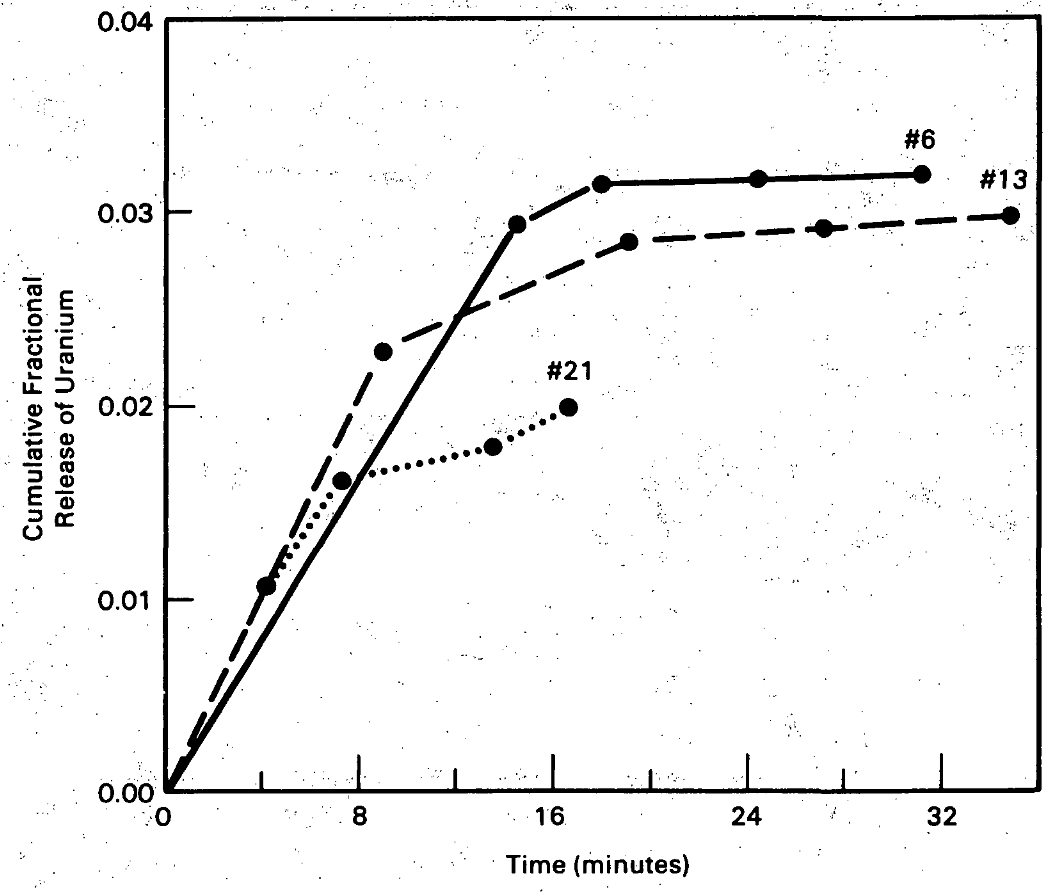

FIGURE 6. Fractional Release of Uranium from Contaminated PMMA

The DUO was released before ignition or early in the burning period. The mechanisms that contribute to the release before ignition are probably entrainment caused by the increase in vapor pressure of the PMMA as it heats up and the explosive ejection of particles by the explosion of gas bubbles in the PMMA slab. These explosions were observed to occur during all PMMA runs.

Run #36 was made specifically to identify how early in the experiment the powder was released. It had been noticed in earlier experiments that most of the powder came off before ignition. In Run #36, two of the filter samples were dedicated to the early part of the run instead of one. The results from this run showed about 31% of the total release coming off before the gas bubbles formed and exploded in the PMMA and another 47% of the total release coming off before ignition. The remaining 21% of the release came off during the flaming period.

Run #51 was made to investigate the effects of different types of ignition. In this run, no external heat flux was applied, and the PMMA slab was

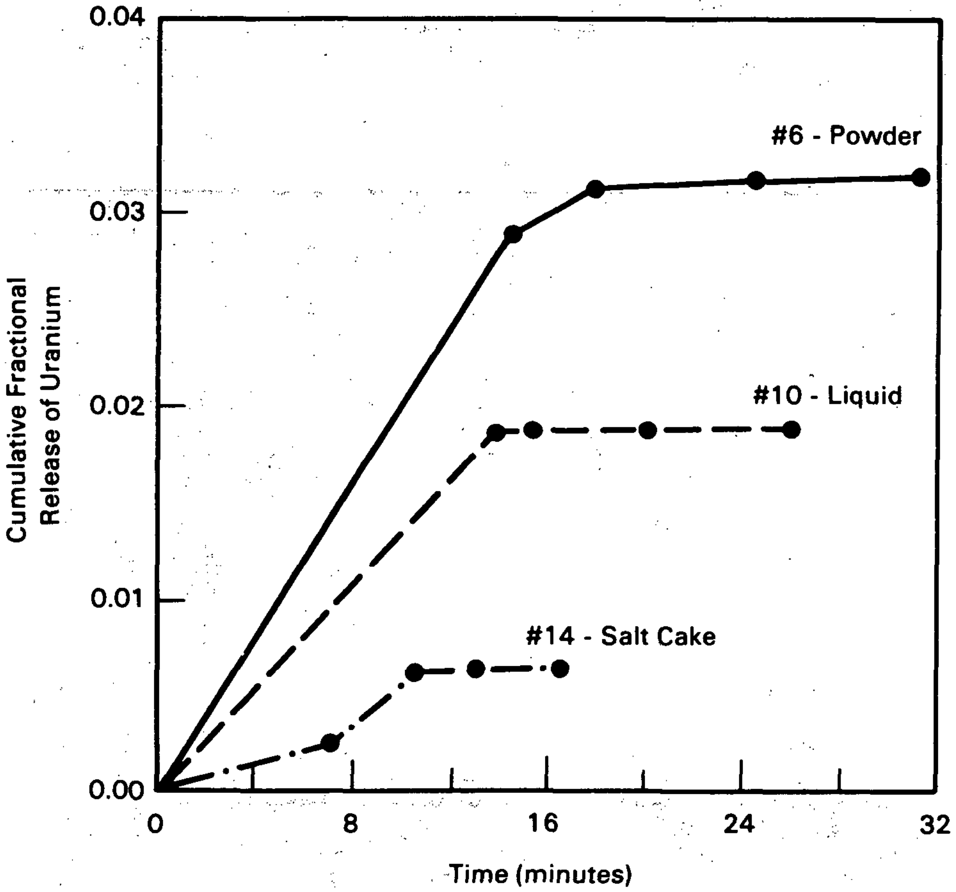

FIGURE 7. Fractional Release of Uranium from PMMA Contaminated with DUO Powder

ignited on two sides simultaneously. The uranium fractional release of about 1.5% in this run is somewhat lower than earlier experiments, but not significantly lower.

Run #14 burned PMMA contaminated with an air-dried UNH solution. Once again, the release of uranium occurred before ignition. The uranium fractional release of 0.65% is much lower than the ~3% obtained from the powder runs. This is probably because of the stronger cohesive forces between the salt cake and PMMA slab.

Runs #10, #19, and #21 all burned PMMA with UNH solution contaminant. The differences in the runs were mainly in the amount of UNH solution placed on top of the PMMA slab. Run #10 used 5 mL, Run #19 used 2 mL, and Run #21 used 0.1 mL. The results were quite similar for Runs #10 and #21, but Run #19 gave a much lower uranium release. The analytical results of Run #19 are suspect, however, so this run will not be considered further. Run #10 had a uranium fractional release of 1.9%, while Run #21 had a release of 2.0%. This difference is quite small and is probably caused by experimental variation. Once again, the release occurred before ignition.

Cellulose

Seventeen runs (1, 2, 3, 4, 5, 12 ,15, 18, 20, 22, 23, 26, 27, 28, 35, 37, and 39) burned cellulose contaminated with powders, solutions, or salt cakes. Smoke fractional release ranged from 0.45 to 19%. Uranium fractional released ranged from 7 x 10^{-5} to 3 x 10^{-3} and are shown in Figures 8 and 9. Data from Runs #1 and #2 are incomplete and are not presented.

Runs #3, #5, and #18 used UNH solutions as the contaminant. Run #3 had the highest contaminant concentration, and an ignitor was used. Run #5 had a slightly lower concentration, no ignitor, and a slightly lower heat flux. Run #18 had a much lower contaminant concentration and an ignitor. Run #3 gave the highest uranium fractional release of the liquid runs at 1.5 x 10^{-4}, twice that of the other two runs. The effect of not having an ignitor was about the same as using a much smaller concentration of contaminant. Uranium fractional releases of 7.5 x 10^{-5} and 7.1 x 10^{-5} were obtained in Runs #5 and #18, respectively.

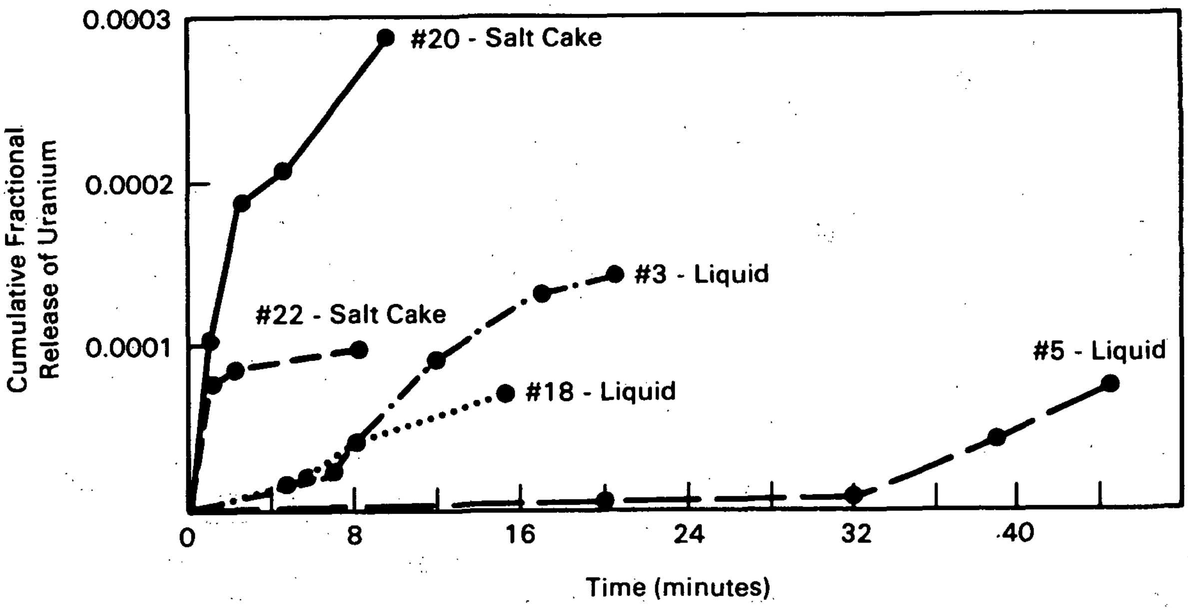

FIGURE 8. Fractional Release of Uranium from Contaminated Cellulose

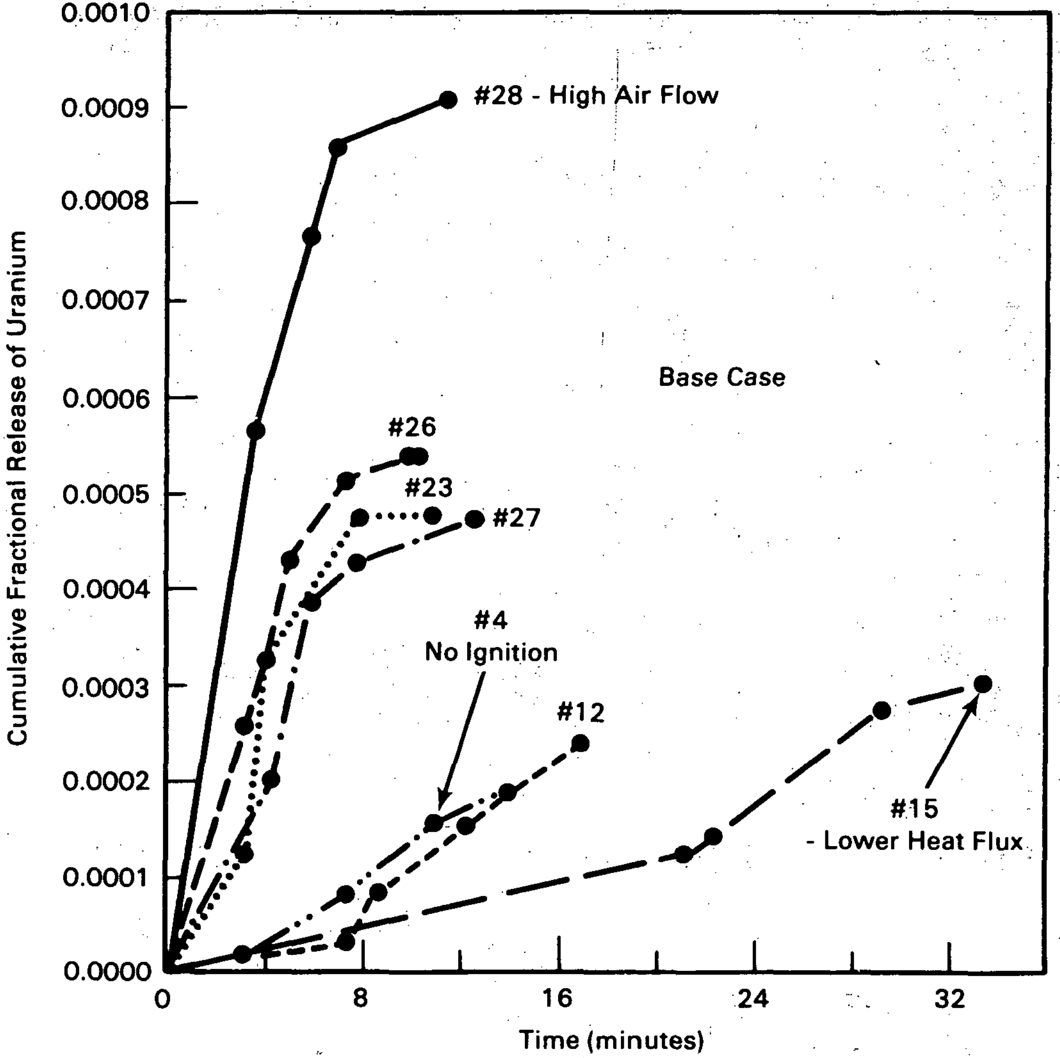

FIGURE 9. Fractional Release of Uranium from Cellulose Contaminated with Powder

Runs #20 and #22 involved air-dried UNH solutions. The uranium fractional release was highest for Run #20, where no ignitor was used. The value of 2.9 x 10^{-4} is slightly higher than the value for the comparable powder run (Run #4). The uranium fractional release for Run #22 is 9.6 x 10^{-5}, which is lower than the comparable powder run (Run #12). Part of the difference may be in fire scaling, as one towel was burned in Runs #20 and #22, while three towels were burned in Runs #4 and #12. The overall range varies by a factor of 3 on these releases, which are the smallest releases measured in these experiments.

Runs #4, #12, #15, #23, #26, #27, #28, #35, and #37 all burned cellulose with DUO powder contamination. Smoke fractional releases ranged from 1.1 to 4.2%. Uranium fractional releases ranged from 1.9×10^{-4} to 1×10^{-3}. Figure 9 shows the release rate of uranium from these runs.

Run #28 gave the third highest uranium fractional release of 9×10^{-4} from burning towels. This appears to be caused by the much higher air flow used in this run. A lower oxygen concentration was used with twice as much air flow. As will be seen later, oxygen concentration did not appear to have a large effect, so air flow appears to be responsible for the release. Runs #23, #26, and #27 had approximately equivalent uranium fractional releases of 4.8×10^{-4} to 5.5×10^{-4}. The only difference in these runs was that the oxygen concentration was varied from 6 to 21%. Runs #4 and #12 were variations of Run #23, with Run #4 having no ignitor and Run #12 having an ignitor but a lower contaminant concentration. Both runs had lower uranium fractional releases as a result of the changes, with the fractional uranium release being about half the release of Run #23. Run #15 was also similar to Run #23, but with a lower heat flux. The uranium fraction release decreased to about 3×10^{-4} from this effect.

Runs #35 and #37 involved the use of an ignitor but no external heat flux. Runs #35 and #37 gave the highest uranium fractional releases of all cellulose runs. The releases were 3×10^{-3} and 1.1×10^{-3}, respectively. The high releases may have been caused by the cellulose immediately beginning flaming combustion instead of charring/smoldering followed by flaming combustion. The releases during flaming combustion for all runs seem to be higher than releases during smoldering or after flaming combustion. The smoldering period may allow the powder to react with the mineral residue and bind to it strongly enough to reduce releases.

Run #39 was a background run involving cellulose and powder but no heat flux or ignition. The uranium fractional release of 1×10^{-4} measured the effect of aerodynamic forces from the air flow in the system.

Mixed Fuels

Runs #40 and #41 were burns of combinations of various fuels. Run #40 involved cellulose and PC, while Run #41 involved cellulose and PMMA. The uranium fractional releases were 9.5×10^{-4} and 3.6×10^{-4}, respectively. The order of magnitude of these fractional releases are similar to releases from cellulosic materials.

COMBUSTIBLE LIQUIDS

Five different liquids, two combustible and three noncombustible, were used in these experiments. These liquids were used in combinations of one combustible and one noncombustible liquid, which resulted in six categories of experiments. Two categories, one where both nitric acid and organic are uncontaminated and the other where nitric acid is spiked with fission products and organic is uncontaminated, did not involve any uranium and are discussed in a later paragraph on fission product release results. The remaining four categories are discussed below. Overall, the experiments in each category were fairly reproducible (with one exception discussion below), and there were obvious differences between the categories.

Pure Acid/Organic with Uranium

Runs #44, #45, and #46 involved uncontaminated nitric acid and 30% TBP in NPH with uranium. Uranium fractional releases ranged from 0.004 to 0.0056 (0.4 to 0.6%). These runs resulted in the lowest uranium fractional releases of the four categories.

The relatively low fractional release of the uranium may be caused by the experimental configuration and apparatus. In these experiments, 100 mL of organic liquid was placed on top of 100 mL of acid in a metal beaker. The beaker was heated to simulate an external heat flux, and the organic vapors were ignited above the beaker. If the beaker was continually heated during the run, the acid eventually boiled over and quenched the burning organic. If heating of the beaker was stopped at the time of ignition, the acid boilover was delayed but not eliminated. Boilover in this case was caused by conductive heat transfer through the walls of the beaker from the flame to the acid. The substitution of a glass beaker for the metal beaker just delayed the boilover. All three runs ended with a boiling acid layer quenching the flame and cooling the organic layer. Significant amounts (from 40 to 60%) of organic layer remained unburned.

While these experiments may show that fires of organic liquid on top of aqueous liquids may put themselves out if sufficient heat is transferred to the aqueous liquid, the numerical values of the uranium fractional releases may not adequately describe those expected from the same configuration without boilover. However, uranium seemed to be released with smoke in these experiments.

Under the experimental conditions, the organic fuel typically ignited in 10 to 20 min and burned for another 10 to 20 min. No great differences were seen in the releases for the shorter runs versus the longer runs. This may be because the run times were only different by a factor of 2, and the actual burn time differences were less than that. Based on these three experiments, a uranium release of 0.6% may represent a lower boundary for the release. If heat transfer from the flame to the acid is reduced, the fire could burn longer, and the release may be higher.

Acid with Fission Products/Organic with Uranium

Runs #47 and #48 involved the nitric acid spiked with fission products and 30% TBP in NPH with uranium. Uranium fractional releases were from 0.025 and 0.027 (2.5 to 2.7%), respectively.

The uranium fractional releases obtained in these experiments are 5 to 6 times greater than those described in the previous section involving uncontaminated nitric acid. The presence of fission products in the acid phase seems to enhance uranium release from the organic phase. The length of these runs is similar to the previously described runs, both in total time and total burn time, so no effect from burn time is noted. Similar quantities of organic material remained after burning, indicating similar amounts of organic consumed. Other than a slight change in the difference in molarity between the two acid solutions (the uncontaminated acid was 3 M, while the acid with fission products was 2.88 M), the difference in the results must be attributed to the addition of fission products in the acid.

One theory is that when fission products are not present in the acid, heat causes some of the uranium to migrate to the acid from the organic. When fission products are present in the acid, this migration is stopped, and more uranium can be released from the organic. However, analysis of the remaining acid layer did not support this theory. Comparable amounts of uranium were found in both the uncontaminated acid and the acid with fission product experiments.

Acid with Fission Products and Uranium/Pure Organic

Runs #52, #53, and #58 involved nitric acid with fission products and uranium with 40% TBP in NPH. Run #58 had a much lower release (0.2%) of uranium than Runs #52 and #53 (6.0 and 7.1%, respectively). Runs #52 and #53 burned to a dry residue, presumably consisting of TBP, residue from the NPH, and salts from the acid. Run #58 did not burn to dry residue, and some of the NPH (~40%) remained after the flame died out. The major differences in the three runs were the amount of liquid used, the beaker material, and the heat input method. Runs #52 and #53 used 100 mL each of acid and organic in a metal beaker with a heat tape around the beaker. Run #58 used 150 mL of acid with 50 mL of organic in a glass beaker with a heat lamp to provide external heat flux.

The high uranium releases appeared to be caused by production of a solid residue. The heat necessary to evaporate the acid probably came from the flame by way of metal beaker, while the glass beaker kept this heat transfer to a minimum. These results give a spectrum of release that might be expected in a fire. As in the earlier liquid runs, heat transfer from the flame to the acid seems to be a critical parameter. Also of significance is the complex uranium/acid/organic material interactions noted in the previous section. Even in Run #58, where very little of the acid layer was boiled off, the fractional release was significant. The uranium may be released during vaporization of the acid and, at the same time, transferred to the organic layer with release during subsequent combustion of the organic.

Acid with Fission Products and Uranium/Organic with Uranium

Runs #56 and #57 involved solutes in both the organic and acid layers. Uranium releases were 1.6% and 0.8% for Runs #56 and #57, respectively. Note that these runs did not burn to a solid residue, so the high releases of Runs #52 and #53 would not be expected, which was indeed the case. The releases obtained in Runs #56 and #57 are lower (by a factor of 3) but higher (by a factor of 5) than Run #58 where no solid residue was produced. Strangely enough, however, the release is less than the release in runs where uranium was present in the organic phase only. No reason can be given for this other than experimental variation.

Fission Product Release

Fission product simulants consisting of cesium, strontium, and ruthenium in "typical" fuel reprocessing concentrations were added to four of the combustible liquid experimental subsets (a total of nine experiments). The analytical method used was not adequate to provide reliable data. Overall mass balances of the release and residue indicate an excess of 200 to 300% in the case of cesium and strontium and a total lack of ruthenium. The ruthenium absence may be caused by the formation of a ruthenium oxide precipitate in the analytical samples. The excess of cesium and strontium could be attributed to a contaminant in the solution that interfered with analysis.

Because of the lack of data, no conclusion as to the airborne release of fission product can be made.

SIZE DISTRIBUTION OF AEROSOLS

The size distribution of airborne particles was measured in at least one run for each of the fuels burned. Table 4 lists the airborne mass median diameter (AMMD) and geometric standard deviation for the runs in which particle size was taken.

TABLE 4. Size of Aerosols from Burning Contaminated Combustibles

Run #	Fuel	AMMD, μm	σ_g	Less than 10 μm
10	PMMA[a]	4.3	2.3	84
11	PC[b]	19.9	4.6	16
12	Cellulose	>0.1		97
14	PMMA	4.9	4.6	63
23	Cellulose	10.5	5.7	40
28	Cellulose	7.8	5.2	47
30	PMMA	3.9	2.5	84
38	PS[c]	1.7	3.8	90
51	PMMA	1.4	4.3	95
56	TBP/NPH	0.6	3.1	99

(a) Polymethylmethacrylate.
(b) Polychloroprene.
(c) Polystyrene.

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APPENDIX A

PERCENT AIRBORNE FROM BURNING CONTAMINATED COMBUSTIBLES

TABLE A.1. Results from Burning Contaminated Polychloroprene (PC)

Run #	Time, min	Cumulative % Release Uranium	 Smoke	Notes
7	0	0.0	0.0	DUO powder
	1.0	0.48	4.4	Heat flux = 23 kW/m^2
	1.5	0.97	11.5	
	2.8	1.01	12.9	
	7.5	1.03	13.9	
9	0	0.0	0.0	DUO powder
	1.2	0.64	8.6	Heat Flux = 23 kW/m^2
	2.3	0.76	13.1	No ignition
	4.0	0.78	14.2	
	10.0	0.80	14.8	
11	0	0.0	0.0	DUO powder
	8.3	0.24	1.5	Heat Flux = 13 kW/m^2
	13.6	0.31	5.0	
	15.1	0.35	6.7	
	15.7	0.37	7.0	
8	0	0.0	0.0	UNH liquid
	0.7	0.11	1.5	
	1.3	2.27	5.8	
	2.3	3.23	7.5	
	9.0	3.47	10.3	
24	0	0.0	0.0	Air-dried UNH
	1.0	0.12	1.2	
	2.0	0.39	6.9	
	4.4	0.42	9.6	
	7.5	0.42	10.0	

TABLE A.2. Results from Burning Contaminated Polystyrene (PS)

Run #	Time, min	Cumulative % Release		Notes
		Uranium	Smoke	
17	0	0.0	0.0	0.17 mL UNH/g PS
	9.0	0.15	0.5	
	10.0	0.15	2.1	
	13.5	0.16	4.7	
	18.5	0.16	5.9	
25	0	0.0	0.0	0.4 mL UNH/g PS
	6.7	0.17	0.6	
	7.8	0.18	2.9	
	9.3	0.18	7.0	
	12.8	0.18	9.6	
38	0	0	0	0.5 mL UNH/g PS
	5.3	0.74	0.8	
	6.5	0.76	2.3	
	12.8	0.77	2.7	
		0.78	3.0	
55	0		0.0	0.6 mL UNH/g PS
	4.0		1.1	Amberlite ion exchange resin
	7.5		3.8	
	20.0		4.0	
			4.1	

TABLE A.3. Results from Burning Contaminated Polymethylmethacrylate (PMMA)

Run #	Time, min	Cumulative % Release Uranium	 Smoke	Notes
6	0	0.0	0.0	DUO powder
	14.5	2.9	0.1	
	18.0	3.2	0.4	
	24.5	3.2	0.8	
	31.5	3.2	0.9	
13	0	0.0	0.0	No ignition
	9.0	2.3	0.1	DUO powder
	19.0	2.8	0.3	
	27.3	2.9	0.5	
	34.6	3.0	0.6	
29	0	0.0	0.0	DUO powder
	4.4	1.0	0.1	Thick layer
	9.1	3.3	0.5	
	14.4	3.4	1.0	
	18.4	3.5	1.1	
30	0	0.0	0.0	DUO powder
	5.0	2.0	0.1	Thick layer
	7.9	3.3	0.6	
	13.7	3.5	1.1	
	17.0	3.6	1.1	
31	0	0.0	0.0	DUO powder
	3.9	0.4	0.1	Pile
	7.8	4.2	0.6	
	14.0	4.4	1.1	
	19.5	4.4	1.2	
32	0	0	0.0	DUO powder
	4.5	0.6	0.1	Pile
	9.3	4.1	0.5	
	14.3	4.3	1.1	
	19.3	4.5	1.1	
33	0	0.0	0.0	DUO powder
	10.3	0.3	0.1	Under PMMA
	13.3	1.1	0.7	
	15.5	1.3	1.1	
	21.0	1.3	1.1	

TABLE A.3. (contd)

		Cumulative % Release		
Run #	Time, min	Uranium	Smoke	Notes
34	0	0.0	0.0	DUO Powder
	5.0	0.07	0.003	No burn, only entrainment
	10.0	0.13	0.003	
	15.0	0.21	0.063	
	20.0	0.25	0.063	
36	0	0.0	0.0	DUO powder
	5.0	1.2	0.07	Early sampling to detect spike
	7.3	2.9	0.17	
	12.8	3.2	0.75	
	18.6	3.7	1.09	
51	0	0	0	DUO powder
	4.5	0.42	0.02	
	14.5	1.10	0.35	
	20.5	1.30	0.56	
	30.3	1.52	0.56	
10	0	0.0	0.0	Liquid UNH
	13.8	1.9	0.2	0.12 mL UNH/g PMMA
	15.4	1.9	0.4	Flame above sample
	20.3	1.9	1.0	
	26.0	1.9	1.1	
19	0	0.0	0.0	Liquid UNH
	6.5	0.1	0.1	0.05 mL UNH/g PMMA
	9.6	0.2	0.5	Impinging flame
	12.8	0.2	0.9	No. 3 uranium sample suspect
	15.5	0.2	1.2	
21	0	0.0	0.0	Liquid UNH
	4.1	1.1	0.1	0.12 mL UNH/g PMMA
	7.5	1.6	0.4	Impinging flame
	13.3	1.8	0.8	
	16.7	2.0	1.1	
14	0	0.0	0.0	Air-dried UNH
	7.3	0.3	0.04	
	10.5	0.6	0.3	
	13.3	0.6	0.6	
	16.6	0.6	0.9	

TABLE A.4. Results from Burning Contaminated Cellulose

Run #	Time, min	Cumulative % Release Uranium	Smoke	Cumulative Mass Loss, g	Notes
4	0	0	0	0	DUO powder
	3.0	0.0014	0.1	2.5	
	7.5	0.0084	0.3	20.0	
	11.0	0.0161	1.1	30.0	0.1 g DUO/g cellulose
	14.0	0.0188	1.6	31.0	Smoldering
12	0	0.0	0.0	0.0	DUO powder
	7.4	0.0033	0.1	14.7	
	8.5	0.0086	0.3	21.9	
	12.2	0.0154	0.4	31.1	0.03 g DUO/g cellulose
	17.0	0.0239	1.7	31.1	
15	0	0	0.0	0.0	DUO powder
	21.3	0.0127	1.1	5.4	Heat flux - 13 kW/m(a)
	22.3	0.0144	1.2	9.2	
	29.3	0.0278	1.4	26.8	0.09 g DUO/g cellulose
	33.5	0.0303	1.5	27.8	
23	0	0	0.0	0.0	DUO powder
	3.2	0.0123	0.8	5.7	
	4.0	0.0328	0.9	11.3	
	7.7	0.0475	1.0	24.4	0.09 g DUO/g cellulose
	11.0	0.0478	1.1	23.9	
26	0	0	0	0.0	DUO powder
	3.3	0.0254	1.9	5.3	
	5.0	0.0430	2.1	15.9	
	7.3	0.0515	2.2	21.6	0.08 g DUO/g cellulose
	10.4	0.0546	2.2	22.5	
27	0	0	0	0	DUO powder
	4.1	0.0204	3.2	10.9	0.09 g DUO/g cellulose
	5.9	0.0381	3.7	20.4	
	7.7	0.0431	3.9	23.0	
	12.7	0.0478	4.2	21.8	
28	0	0	0.0	0	DUO powder
	3.7	0.0566	1.2	4.5	Airflow = 17.5 lpm(b)
	6.1	0.0761	1.3	20.7	0.095 g DUO/g cellulose
	6.8	0.0860	1.4	22.4	
	11.4	0.0903	1.5	23.2	
35	0	0.0	0.0		DUO powder
	1.0	0.273	0.2		No heat flux
	3.0	0.297	0.4		~0.1 g DUO/g cellulose
	7.0	0.299	0.4		
		0.301	0.4		

TABLE A.4. (contd)

Run #	Time, min	Cumulative % Release		Cumulative Mass Loss, g	Notes
		Uranium	Smoke		
37	0	0.0	0.0		DUO powder
	1.8	0.018	0.03		No heat flux
	3.8	0.077	0.1		~0.1 g DUO/g cellulose
	6.3	0.144	0.3		
	10.3	0.153	0.3		
39	0	0.0	0.0		DUO powder
	5.0	0.006	0.01		No heat flux
	10.0	0.0085	0.03		No ignition
	15.8	0.0098	0.03		Entrainment only
		0.0108			Airflow = 11 lpm
					~0.2 g DUO/g cellulose
20	0	0	0.0	0.0	Air-dried UNH
	1.1	0.011	3.7	5.3	
	2.6	0.019	4.8	11.7	
	4.5	0.021	4.9	13.2	
	9.8	0.029	5.1	14.5	
22	0	0.0	0	0	Air-dried UNH
	1.0	0.008	0.3	8.2	Heat flux = 13 kW/m^2
	2.3	0.009	0.4	12.0	
	8.3	0.010	0.5	14.6	
3	0	0	0	0	Liquid UNH
	7.0	0.003		25.8	
	12.0	0.009		62.5	
	17.0	0.013		93.8	
	20.5	0.014		101.7	
5	0	0.0	0	0	Liquid UNH
	20.0	0.0003	2.3	29.9	Heat flux = 7 kW/m^2
	32.0	0.0005	12.0	64.1	
	39.0	0.0043	18.1	84.9	
	44.5	0.0075	19.0	108.8	
18	0	0.0	0.0	0.0	Liquid UNH
	4.9	0.0015	0.3	6.0	
	6.0	0.0023	0.5	13.2	
	8.3	0.0042	0.8	24.3	
	15.3	0.0070	1.1	38.0	

(a) Heat flux = 23 kW/m^2 in all other runs.

(b) Airflow = 8 lpm in all other runs.

TABLE A.5. Results from Burning Mixed Fuels

Run #	Time, min	Cumulative % Release Uranium	 Smoke	Notes
40	0	0	0.0	Cellulose and PC
	1.0	0.016	0.7	DUO powder
	3.0	0.045	2.2	
	4.5	0.067	3.5	
	9.0	0.095	4.5	
41	0	0	0	Cellulose and PMMA
	5.0	0.012	0.2	DUO powder
	8.8	0.017	0.4	
	14.5	0.031	0.8	
	25.8	0.035	1.2	

TABLE A.6. Results from Burning 30% Kerosene/Tributylphosphate (TBP)

Run #	Time, min	Cumulative % Release		Notes(a)
		Uranium	Smoke	
44	0	0.0	0.0	Pure acid/organic with uranium
	13.2	0.009	0.10	
	16.3	0.040	0.53	
	22.5	0.132	1.32	
	27.5	0.404	1.89	
45	0.0	0.0	0.0	Pure acid/organic with uranium
	14.7	0.054	0.10	
	18.0	0.395	0.70	
	25.5	0.529	1.00	
	34.8	0.557	1.10	
46	0.0	0.0	0.0	Pure acid/organic with uranium
	19.0	0.006	0.04	
	25.5	0.081	0.17	
	35.7	0.031	0.33	
	53.3	0.434	0.76	
47	0.0	0.0	0.0	Acid with fission product/organic with uranium
	13.3	0.032	0.15	
	16.0	0.105	0.69	
	19.0	0.343	1.46	
	24.8	2.517	3.18	
48	0.0	0.0	0.0	Acid with fission product/organic with uranium
	17.0	0.034	0.10	
	22.0	0.335	0.69	
	26.3	2.054	2.28	
	34.0	2.697	2.88	
52	0.0	0.0	0.0	
	8.3	0.068	0.18	
	12.5	0.170	1.10	
	20.5	0.606	2.03	
	61.3	5.978	8.11	
53	0.0	0.0	0.0	Acid with uranium and fission product/pure organic
	9.3	0.097	0.22	
	14.2	0.206	1.1	
	37.0	2.620	4.64	
	65.0	7.088	8.94	

TABLE A.6. (contd)

		Cumulative % Release		
Run #	Time, min	Uranium	Smoke	Notes
56	0.0	0.0	0.0	Acid with uranium and fission product/organic with uranium 150 mL acid/50 mL organic
	24.8	0.032	0.18	
	31.5	0.060	1.15	
	39.3	0.330	2.45	
	57.3	1.563	6.51	
57	0.0	0.0	0.0	Acid with uranium and fission product/organic with uranium 100 mL acid/50 mL organic
	19.2	0.010	0.10	
	28.5	0.028	0.83	
	39.5	0.621	2.52	
	51.0	0.809	3.03	
58	0.0	0.0	0.0	Acid with uranium and fission product/pure organic 150 mL acid/50 mL organic
	16.0	0.003	0.20	
	23.0	0.006	2.04	
	30.0	0.012	4.55	
	40.0	0.170	6.44	
42	0		0	Pure acid/pure organic
	17.3		0.25	
	19.2		0.79	
	23.0		1.60	
	36.8		2.53	
43				No ignition. Slight boiling loss. Pure acid/pure organic
49	0		0	Acid with fission product/pure organic
	14.8		1.06	
	18.5		1.94	
	30.0		2.87	
50	0		0	Acid with fission product/Pure organic
	12.5		0.28	
	17.0		1.36	
	21.0		2.79	
	31.5		4.51	
54	0		0	Pure acid/pure organic
	12.3		0.16	
	16.8		1.10	
	27.0		2.74	
	32.0		3.81	

(a) 100 mL of acid/100 mL organic unless otherwise noted.

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Safety assessments and environmental impact statements for nuclear fuel cycle facilities require an estimate of potential airborne releases. Radioactive aerosols generated by fires were investigated in experiments in which combustible solids and liquids were contaminated with radioactive materials and burned. Uranium in powder and liquid form was used to contaminate five fuel types: polychloroprene, polystyrene, polymethylmethacrylate, cellulose, and a mixture of 30% tributylphosphate (TBP) in kerosene. Heat flux, oxygen concentration, air flow, contaminant concentration, and type of ignition were varied in the experiments. The highest release (7.1 wt%) came from burning TBP/kerosene over contaminated nitric acid. Burning cellulose contaminated with uranyl nitrate hexahydrate liquid gave the lowest release (0.01 wt%). Rate of release and particle size distribution of airborne radioactive particles were highly dependent on the type of fuel burned.

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