COMPARISON OF FISSION PRODUCT RELEASES FROM THE DESTRUCTIVE TESTS OF TWO SNAP 10A/2 REACTORS*

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Abstract—Retention of fission products by the water environment surrounding a water-moderated reactor has long been considered an effective measure for reducing the radiological consequences of a reactor accident. Little credit has been taken for this factor in reactor safety analyses, however, because experimental evidence as to the degree of this reduction has been lacking.

An unusual opportunity to study the retention effect of the water environment upon the fraction and type of fission products released during a reactor accident was provided as part of an experimental program in aerospace safety conducted by Phillips Petroleum Company for the United States Atomic Energy Commission. In separate experiments two modified SNAP 10A/2 flight system reactors containing fuel made of an alloy of uranium and zirconium-hydride were intentionally destroyed. One of these reactors was tested while immersed in water and the other was tested in air to determine the consequence of an uncontrolled self-limiting excursion in each of these environments.

This paper compares the fission product release information obtained from these experiments and evaluates the fission product retention effect of the water environment. The major conclusions reached regarding the retention effect of the water environment are that:

1. The water cooling effect on the hot fuel, which limited the destruction of the fuel and caused the fission products to remain trapped in the fuel matrix, is an important mechanism in suppressing the release of fission products.
2. The release fraction of noble gas fission products is greatly reduced by a water environment. Only 4% of the total noble gas fission products were released from the water-immersion test.
3. Halogen and "solid" fission products from an underwater destruction excursion are completely retained, provided the water environment exists following the excursion.

1. INTRODUCTION

In the safety analysis of many water-cooled reactors the coolant has been assumed to supply an additional barrier to the release of fission products. However, little credit has been taken for this factor because experimental evidence as to its effectiveness has been lacking. In the past, information concerning the effectiveness of such a barrier was studied by subjecting small, fueled capsule experiments to conditions simulating a reactor accident and observing the effect of an added water barrier upon the fraction of fission products released. [1] This approach has certain limitations since it is necessary to scale the results from the capsule experiments to full-scale integral-core reactors. Because the conditions in a full-scale reactor core vary from point to point and cannot be uniquely represented by a small isolated sample, the methods for scaling are approximate and not well known. This uncertainty in scaling generally results in an over-conservative estimate when applied to fission product releases.

An ideal experiment for determining the extent a water environment reduces the release of fission products from a reactor accident would consist of two full-scale reactor destructive tests,

with one reactor immersed in water and the other in air. This use of full-scale integral-core reactors would eliminate the necessity of scaling the results of smaller capsule-type experiments to actual reactor accident conditions.

An approximation to this ideal experiment occurred as part of an experimental program in aerospace safety conducted by Phillips Petroleum Company for the United States Atomic Energy Commission at the National Reactor Testing Station in Idaho. In this experiment, designated the SNAPTRAN program, two modified SNAP 10A/2 flight system reactors were intentionally subjected to severe reactivity insertions which resulted in their destruction. The first of these was an underwater test designed to determine the radiological consequences of an accidental immersion of one of these reactors in water, such as could occur from the aborting of an attempted launch of the reactor into orbit around the earth. The second test was conducted in an air atmosphere to gain information on reactor kinetics as well as to demonstrate the radiological consequences of an uncontrolled self-limiting excursion in an air environment.

The purpose of this paper is to compare the fission product release data from these two destructive tests in order to evaluate the extent to which the water environment reduced the release of fission products to the atmosphere. In addition, radiological instruments and sampling techniques are briefly described.

2. REACTOR DESCRIPTION

The two reactors used in the SNAPTRAN destructive tests were SNAP 10A/2 flight system reactors with modified control systems to provide for rapid insertion of reactivity. Each of the identical reactor cores contained thirty-seven fuel rods in a thin, cylindrical, stainless-steel vessel approximately 23 cm in diameter and 31 cm in height. The fuel rods were made from a homogeneous alloy of zirconium-hydride and uranium and were clad with Hastelloy-N. With this arrangement, each reactor contained 4.75 kg of U-235 and 464 g-moles of hydrogen. For normal flight system operation, an external, fixed beryllium reflector was placed around the core to provide sufficient neutron reflection for reactor operation and the interstitial spaces

in the core were filled with a sodium-potassium (NaK) liquid-metal coolant.

For the water-immersion test, the reactor was located in a concrete environmental tank which had an internal diameter of 4.25 m and a depth of 3.1 m. When filled, the water depth above the reactor was approximately 1 m. The size of the tank was selected to simulate a large body of water yet remain small enough to facilitate pressure measurements and evaluation of other phenomena. Because it had been shown that the normal beryllium reflector would be stripped from the reactor during entry into water following a launch abort, the beryllium reflector was not used in the water-immersion test. Instead, the water environment provided the necessary neutron reflection to achieve criticality, and the axial position of a 0.64-cm-thick, neutron-absorbing, boron-aluminum sleeve controlled the reactor. The NaK coolant was used, however, to provide for the possibility of an additional energy release through a NaK-water reaction. To initiate the test the boron-aluminum sleeve was rapidly withdrawn from around the reactor by a pyrotechnic actuator.

For the open-air destructive test, the reactor was mounted in the normal beryllium reflector assembly to simulate the space reactor package. Reactor control was accomplished by the position of beryllium drums which were rotated into rounded slots 90 degrees apart in the fixed beryllium reflector (Fig. 1). Rapid reactivity insertion was achieved by rapidly rotating the steel drive shafts of the beryllium drums with a rack-and-pinion gear train connected to a pneumatically driven cylinder and piston.

To facilitate movement from the test area to the assembly and examination area, both test reactor packages were mounted on railroad dollies. Reactor control and initiation of the destructive tests were accomplished from an underground shielded equipment and control building adjacent to the reactor test pad. The shielded roadway tunnel shown in Fig. 2 gave access to the underground area. The test cell building, also mounted on railroad-type wheels and steel tracks, was pulled back from the test pad prior to the initiation of both tests, leaving the test package essentially open to the environment.
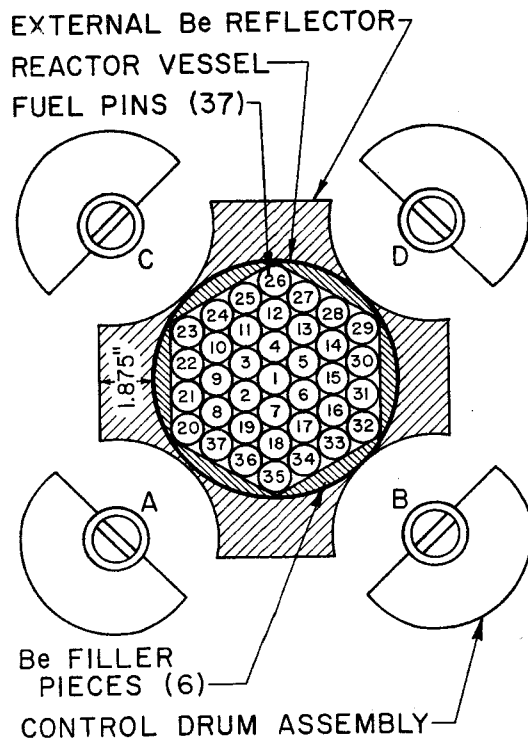
EXTERNAL Be REFLECTOR
REACTOR VESSEL
FUEL PINS (37)

Be FILLER
PIECES (6)

CONTROL DRUM ASSEMBLY

FIG. 1. Typical SNAP 10A/2 reactor and beryllium reflector cross-section.

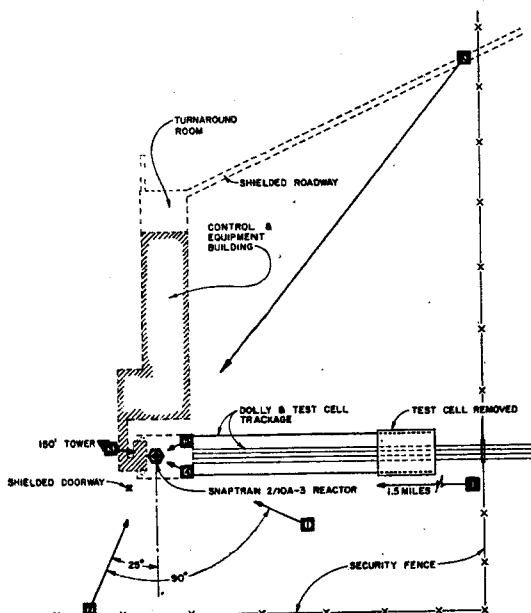

FIG. 2. Test pad layout.

3. TEST DESCRIPTION

The two destructive tests were conducted under similar weather conditions. The first test, which was the water-immersion test, was conducted on April 1, 1964, with a 10 meter per second wind speed and a slightly unstable vertical temperature profile (Pasquill class C). The second, or beryllium-reflected test, was conducted on January 11, 1966, with a 5 meter per second wind speed and a neutral (Pasquill class D) vertical temperature profile. Each test produced a single radioactive cloud which traveled downwind over an instrumented radiological grid.

The radioactive cloud from the water-immersion test seemed to be delayed for several seconds following the excursion, during which time the initial column of water and steam collapsed back into the environmental tank. Following the collapse of the water, the visible vapor-filled cloud moved downwind over the radiological grid. The short delay in release of the cloud was confirmed by a measured shift in the isotopic ratio of the daughters of krypton gases. For example, measured ratios agree with the theoretical ratios if a delayed release time of 3 sec is postulated.

Figure 3 shows the twin flashes from the pyrotechnic actuator which removed the control sleeve and initiated the water-immersion destructive test. In the next picture (Fig. 4) the reactor is at peak power, and the entire environmental tank is illuminated by the Cerenkov glow. The development of the water and steam cloud at 200 msec and 550 msec after peak power is shown in Figs. 5 and 6, respectively. Figure 7 shows the nearly collapsed water and steam cloud at 2.6 sec after peak power. Immediately following this, the visible cloud arose and moved downwind.

The beryllium-reflected test was somewhat more spectacular in that the entire test pad (some 20 m across) was covered with burning hydrogen and fuel. A visible cloud arose from this test without delay and continued to rise, reaching a height of approximately 50 m before leveling off at a distance of 100 m downwind. Some of the fuel pieces continued to burn for several seconds following the test and undoubtedly continued to evolve fission products. Measurements made with samplers located at

Fig. 3. Overhead view of the water-immersion test initiation.

intervals on a 46-m-high tower indicate that the amount of activity released by the burning fuel was small compared to that which followed the path of the visible cloud. Figure 8 shows the beryllium-reflected reactor on the test pad immediately prior to the test. The beginning and rise of the visible cloud are shown in Figs. 9 and 10 taken at 1.10 sec and 2.35 sec, respectively, after peak power.

4. RADIOLOGICAL MONITORING AND INSTRUMENTS

Both destructive tests were monitored to obtain radiological data from which the magnitude of the fission product releases and

radiological hazards could be determined. This monitoring was accomplished by a radiological monitoring grid located on a series of concentric arcs surrounding the reactor test pad. In a 60-degree sector centered on a line 30 degrees east of true north, which coincided with the movement of the prevailing wind, sampling stations were located out to a distance of 5 miles to provide downwind monitoring. The placement of the samplers used on the monitoring grid is shown in Figs. 11, 12, and 13. Figure 14 shows a typical grid station with a high volume air sampler, a fallout plate, a direct radiation monitor, and a balloon-type fission gas sampler. In addition to the downwind ground-level grid samplers, several sets of samplers were located

FIG. 4. Overhead view of the water-immersion test at peak power.

above the reactor test pad on a 46-m-high exhaust stack and a 46-m-high tower. These samplers provided information on the effective height of the radioactive releases and obtained samples through a vertical section of the radioactive cloud.

Figure 15 shows the primary sampler used on the grid. The sampler consisted of a Staplex high-volume air pump modified to hold a 4-in. diameter by 1-in. deep charcoal bed in series with a 4 in. diameter particulate filter (Microsorban). Noble gases were sampled using the balloon-type sampler shown in Fig. 16. The amount of noble gas drawn into the balloon through a one-way valve is determined by counting the radioactive daughters formed during a measured decay interval. Particle sizing of the debris from the destructive tests was attempted using the six-stage Anderson impactor samplers. Several Unico four-stage cascade impactors were also used during the water-immersion test. Horizontal fallout plates containing sticky paper sampled the deposition of radioactive debris. During the water-immersion test, water catch trays and cans were also used. Direct radiation measurements were made using film badge dosimeters, ionization chambers, and thermoluminescent dosimeters. For a more complete description of the reactors and the radiological sampling grid, the reader is referred to United States Atomic Energy Commission reports IDO 17019, IDO 17083, and IDO 17194. [2-4]

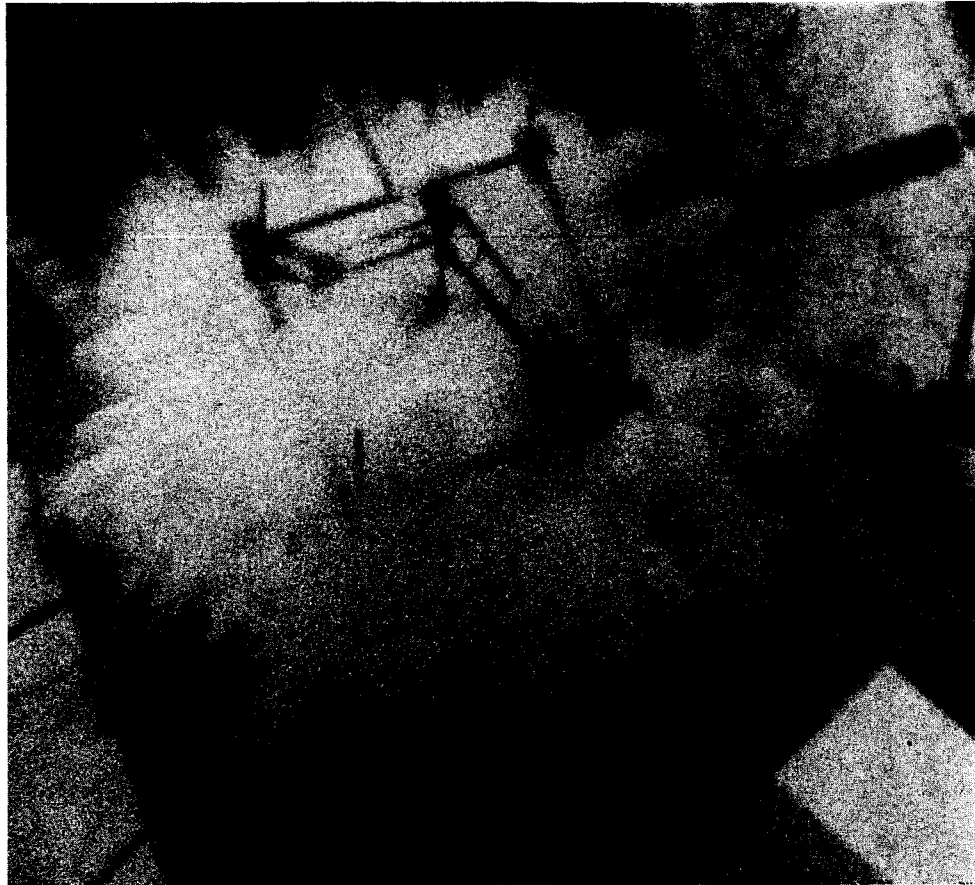FIG. 5. Overhead view of the developing cloud from the water-immersion test 200 msec after peak power.

5. DATA AND ANALYSIS

Since the magnitude of the nuclear energy release from each of the two tests was similar (45 MW-sec for the water-immersion test and 54 MW-sec for the beryllium-reflected test) the quantity of fission products available for release from each test was of the same order of magnitude. The quantity or fraction of the fission products actually released to the atmosphere differed between the two tests, however, indicating that the release of fission products from a reactor accident is a function of the immediate environment of the reactor.

Data from the radiological grid indicated that the only fission products released from the water-immersion test were noble gases. There were no indications of halogen or "solid" fission

products outside the water of the environmental tank other than those which were also the daughters of the short-lived radioactive gases. Only daughters of 3-sec Kr-92, 10-sec Kr-91, 16-sec Xe-140, and 41-sec Xe-139 were detected on the filters of the high-volume air samplers. In addition, the cesium daughter of 17-min Xe-138 was detected in the fission-gas sampler.

Because of this surprising lack of fission products, several lines of investigation were undertaken to verify and establish an upper limit for release fractions of the noble gases and the iodines.

Four independent methods were used to determine the release fraction for the noble gases. The first method, based on a generalized Gaussian dispersion formula[5] in which a source
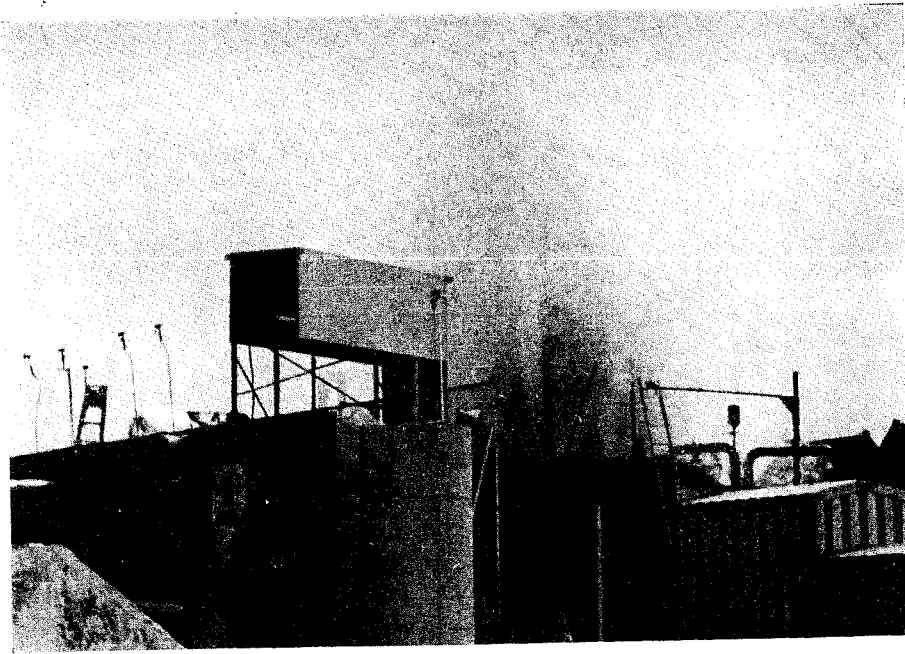
FIG. 6. Elevation view of the developing cloud from the water-immersion test 550 msec after peak power.
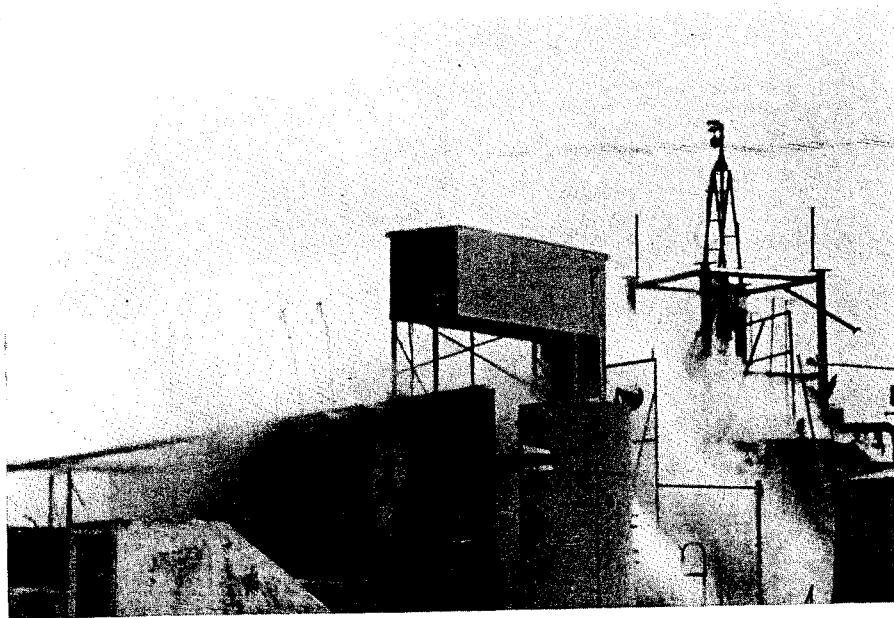

FIG. 7. Elevation view of the nearly collapsed cloud from the water-immersion test 2.6 sec after peak power.
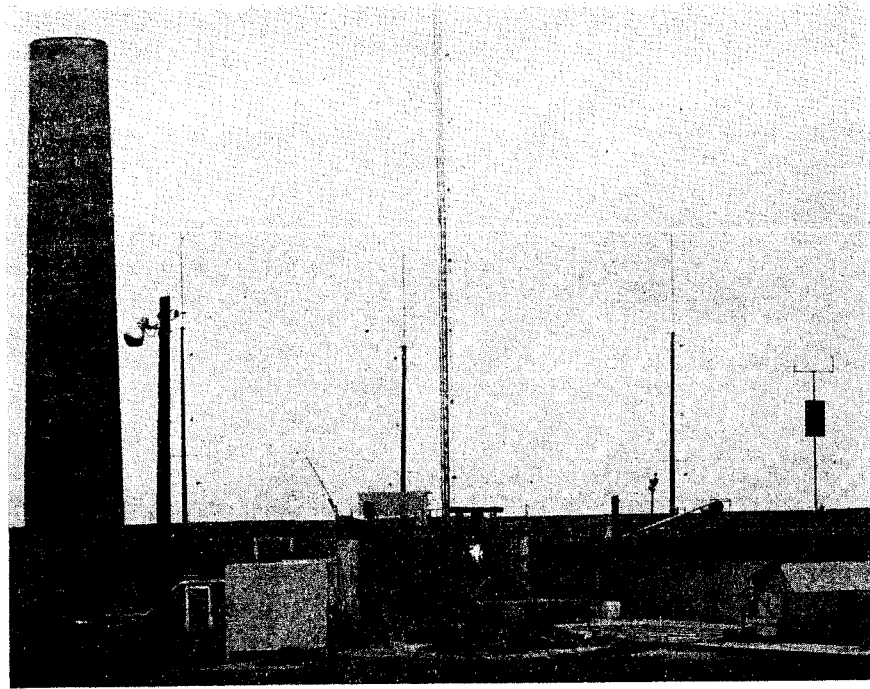
FIG. 8. Elevation view of the beryllium reflected test at 50 msec prior to reactor destruction.
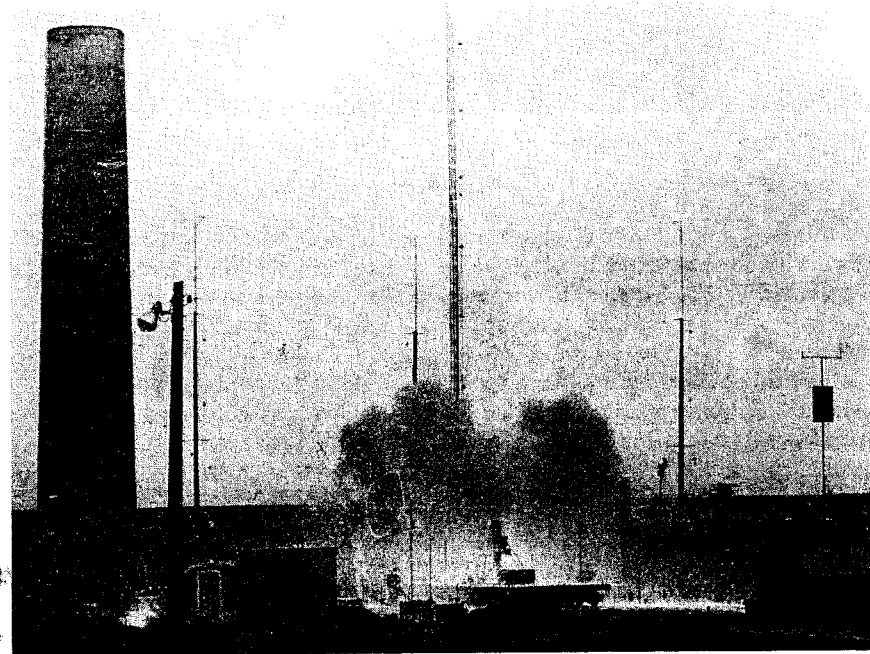

FIG. 9. Elevation view of the developing cloud from the beryllium-reflected test 1.10 sec after peak power.
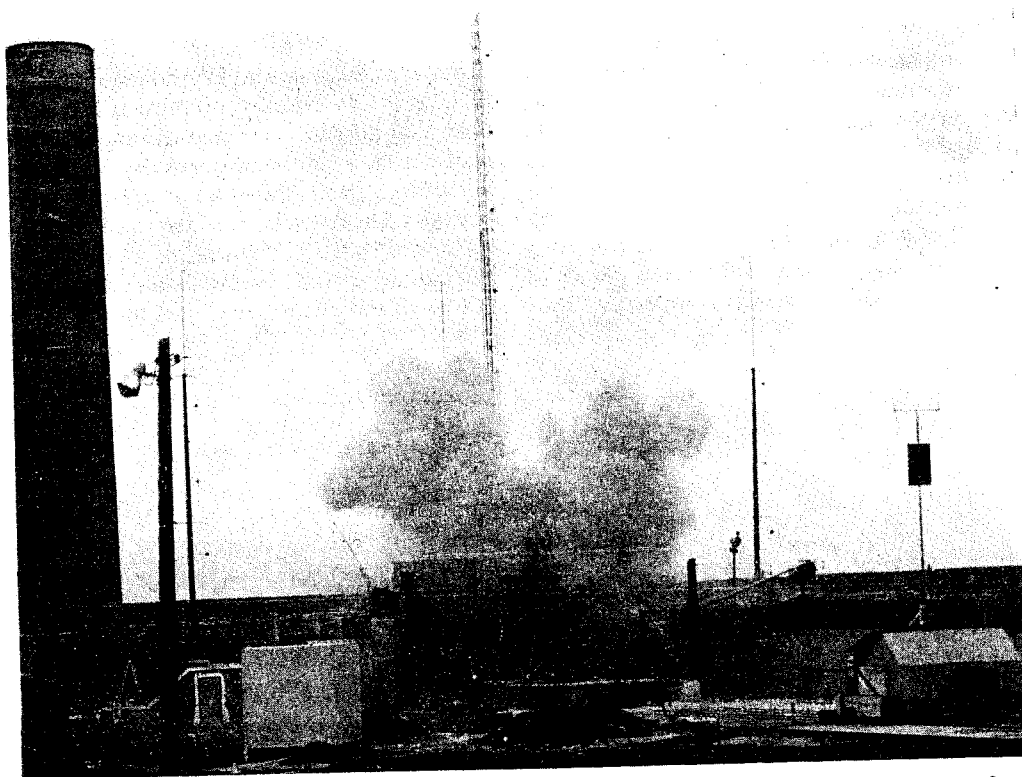
FIG. 10. Elevation view of the developing cloud from the beryllium-reflected test 2.35 sec after peak power.
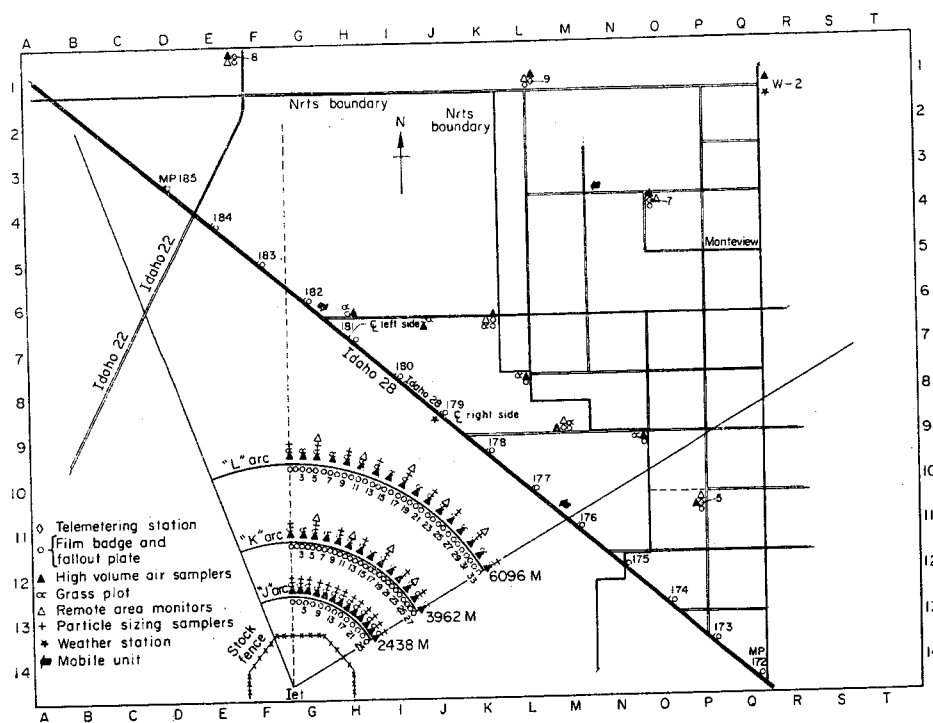

FIG. 11. Radiological monitoring grid beyond 1600 m.

term is calculated from field measurements, led to estimated releases of 4% for Kr-91, 3% for Kr-92, and 3% for Xe-139. In the second method, a physical estimate of the cloud volume was made from photographs and radiation measurements. This volume was assumed to be uniformly filled with the maximum concentration of Xe-138 detected in the cloud to estimate the total quantity released. This cated that 80% of the krypton remained in the fuel. It is felt that this amount is somewhat less than that actually retained during the excursion partly because of the opportunity for the krypton to diffuse from the fuel during the interval between the destructive test and the analysis time and partly because of the difficulty in extrapolating the results from one small piece of fuel to the entire core. All four independent
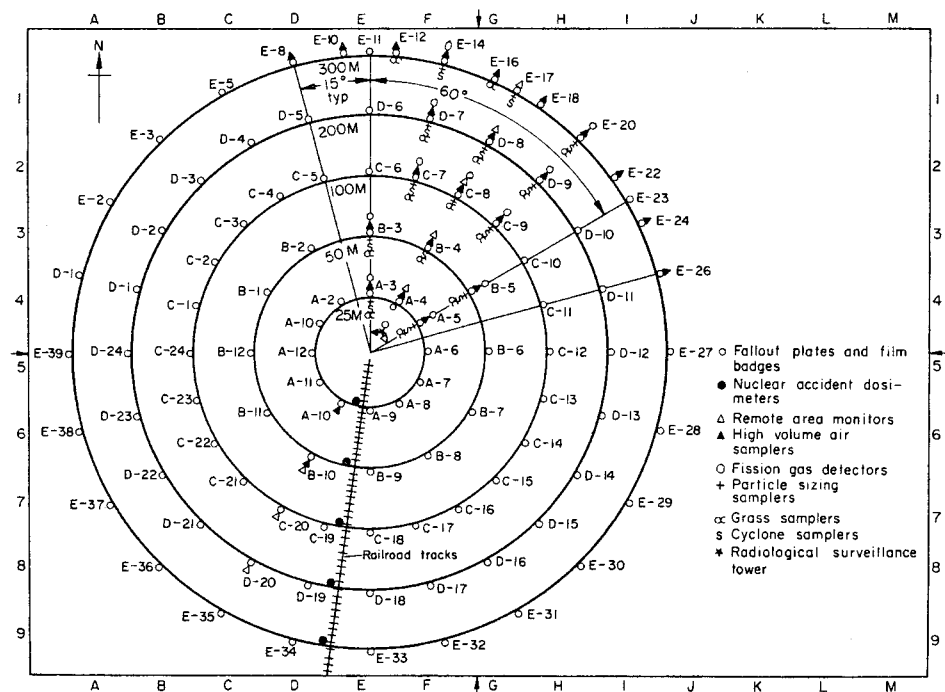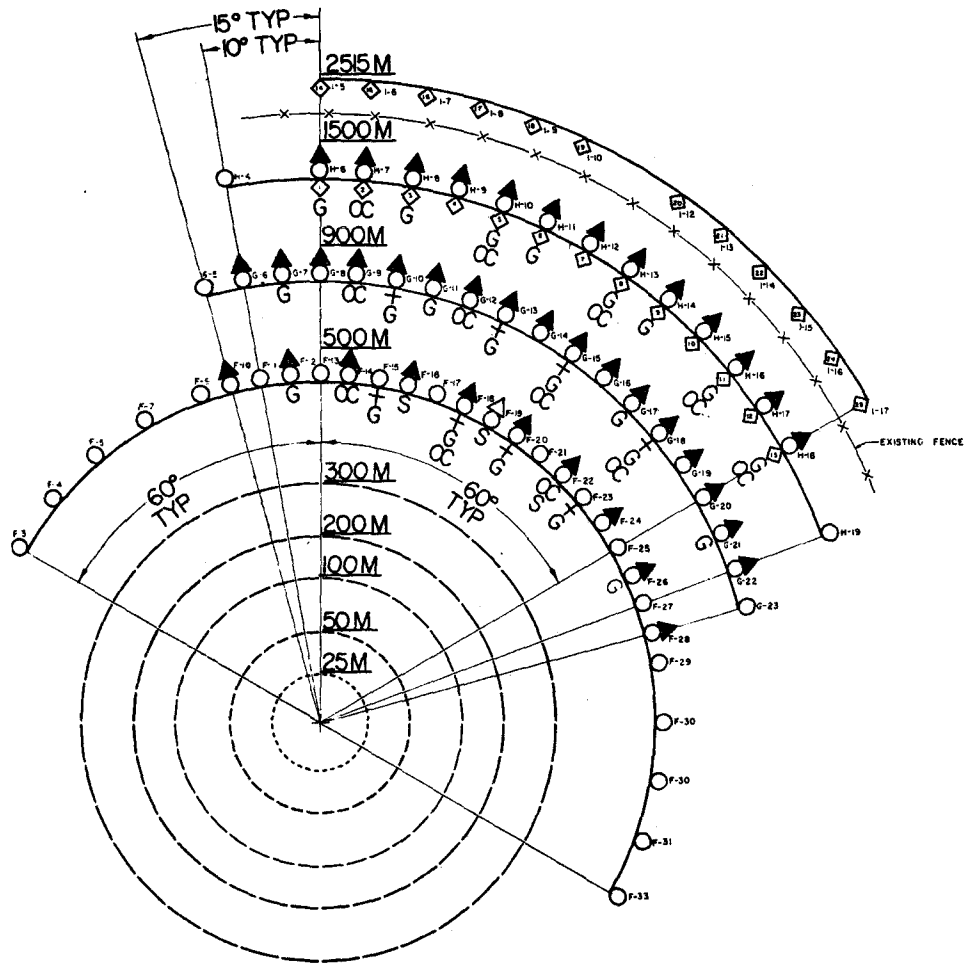

Fig. 12. Radiological monitoring grid to a radius of 300 m.

method indicated a release of 2% for Xe-138. In the third method, all (99%) of the recovered reactor fuel was dissolved and sampled for Cs-137, the daughter of I-137 and Xe-137. The difference between the calculated quantity produced in the reactor core and the measured amount would be that which was released as I-137 or Xe-137. This method indicated that 0% of the Xe-137 chain was released. The fourth method consisted of measuring the amount of Kr-85 remaining in a small piece of fuel and extrapolating this measured concentration to the entire core. This method indi-

methods indicate a very small release percentage of the noble gases. On the basis of these measurements and the assumption that all noble gases behave essentially the same as those measured, the estimate of 4% is taken to represent a reasonable upper limit of the noble gas release from the water-immersion test.

The lack of detectable quantities of iodine on the downwind grid samplers (specifically designed to be highly sensitive to airborne radioiodine) implied that the halogen fission products were completely retained in the fuel or in the water environment during the water-immersion

KEY

O – FALLOUT PLATES & FILM
    BADGES
G – GENERATORS
▲ – HIGH VOLUME AIR SAM –
    PLERS
◇ – TELEMETERING STATIONS
+ – PARTICLE SIZING SAM –
    PLERS
∝ – GRASS SAMPLERS
S – CYCLONE SAMPLERS
△ REMOTE AREA MONITORS

FIG. 13. Radiological monitoring grid to a radius of 2500 m.

test. To substantiate the downwind grid measurements, samples of the environmental tank water and the reactor fuel were analyzed for iodines. Significant quantities of I-131 and I-133 were detected in the samples of the environ-

FIG. 14. Typical grid station consisting of a fallout plate, a film dosimeter, a high volume air sampler, and a balloon fission gas sampler.

mental water taken several days after the test (after the water had been drained to a holding tank) indicating that iodines had been held up in the water. A quantitative balance was not possible because there was no way to distinguish between iodine produced directly by fission and that produced indirectly by tellurium decay. Tellurium strongly tends to plate out on piping and cold surfaces, thus reducing by some unknown quantity the total amount of iodine reaching the sampling point. In addition, the fuel analysis indicated that a significant amount of iodine had been retained in the fuel matrix. Radiochemical analysis of the fuel showed that approximately 87% of the expected I-131 was in the fuel at the time of analysis. This was anticipated because essentially all I-131 comes from telluriums with half-lives longer than 20 min, and tellurium would be expected to remain with the fuel material during the quick cooldown following the excursion. While a quantitative iodine balance was not achieved, the detection of significant quantities of iodine in the water and fuel tends to confirm the positive downwind measurement that halogens were not released to the atmosphere from the water-immersion test.

The test data from the beryllium-reflected test differed significantly from water-immersion test data with respect to the release of fission products. Large quantities of the noble gases, iodines, and telluriums were detected on the downwind radiological grid. In addition, small fuel particles were recovered as far as 200 m downwind, and significant quantities of the "solid" fission products were detected in the radioactive cloud.
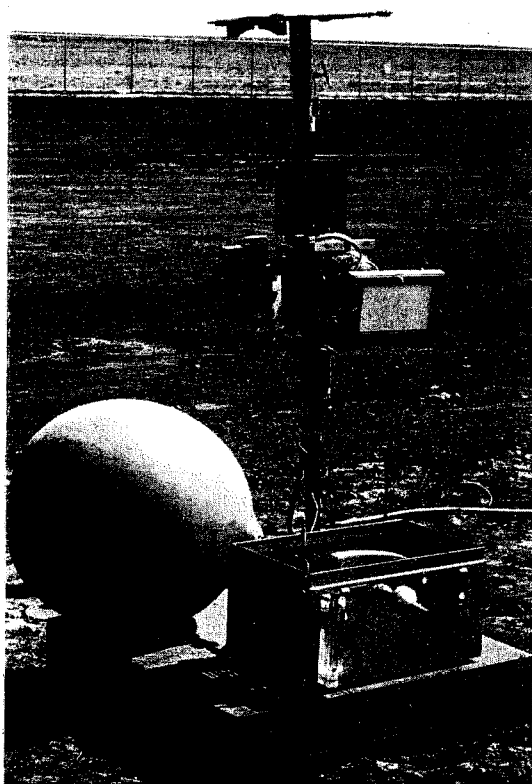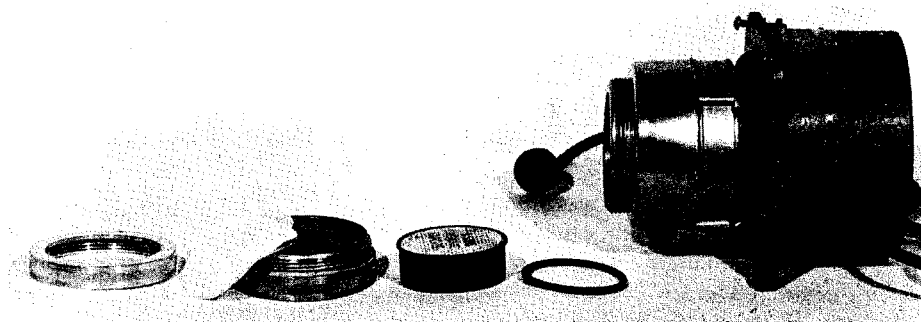

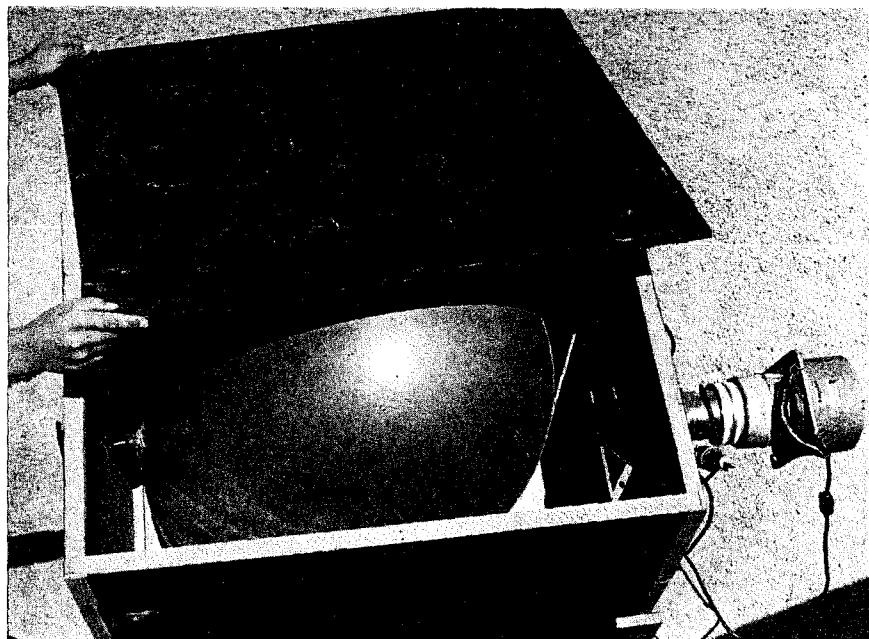FIG. 15. High volume air sampler with charcoal and dust filter.

FIG. 16. Balloon fission gas sampler.

The radioactive cloud from the beryllium-reflected test rose considerably higher than did the cloud from the water-immersion test and as a result could not be adequately sampled by the close-in ground-level samplers. This circumstance, plus the interference from the small fuel fragments, limited the release analysis to samples collected beyond 500 m downwind. Accordingly, all release estimates were made on the basis of dispersion calculations [6] in which a source term, developed from measured data, was compared with the calculated quantity available in the reactor.

Analysis of the activities of tellurium-132 and molybdenum- and technetium-99 collected directly by the samplers indicated that 45% of the tellurium-132 and 5% of the molybdenum- and technetium-99 had been released. These three isotopes were the only fission products other than halogens and daughters of noble gases detected on the grid. By assuming that the isotopes which are also daughters of noble gases were released with the same percentage as molybdenum, the activity on the samplers from the noble gas daughters could be calculated and the release percentage of the noble gases could be

determined. With this assumption the release values from the Kr-91, Kr-92, Xe-139, and Xe-140 chains indicated a release of 75%.

The iodine and tellurium release fractions were determined by solving simultaneous equations of the form:

$$D_n = F_0 D_{no} + \sum_{i=1}^{n} F_i D_{ni}$$

where D_n = detected activity of the nth isotope in a decay chain (corrected for diffusion);

D_{ni} = computed activity of the nth isotope from decay of the ith precursor;

D_{no} = computed activity of the nth isotope available for release;

F_i = fraction of the ith precursor released;

F_0 = fraction of the nth isotope of the decay chain released.

By comparing data from any two of the 131, 132, 133, and 135 decay chains and assuming all iodine detected on the grid was from either direct yield iodine or tellurium decay and that the samplers were 100% efficient, release fractions for both iodine and tellurium were obtained. This technique, when applied to all

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Table 1. *Summary of SNAPTRAN Destructive Test Data*

	Water-immersion test	Beryllium-reflected test
Noble gas release (%)	4	75
Iodine release (%)	0	70
Tellurium release (%)	0	45
Solid release (%)	0	5
Total inventory release (%)	<1	20
Maximum reactivity inserted ($)	3.8	5.1
Minimum period (msec)	0.64	0.20
Maximum power (GW)	18	75
Maximum energy release (MW-sec)	45	54
Maximum fuel temp. (°F)	2400	3000
	2000	2400
Highest avg. core temp. (°F)	1900 at Disassembly	1400 at Disassembly

available data from each of the decay chains, gave an estimated release of 70% for the iodines and 45% for the telluriums. The 45% release of tellurium agrees with the 45% release value determined from direct measurement of tellurium-132 collected on grid samplers.

Table 1 summarizes the fission product release data from the two tests and lists some of the reactor physics data as well.

From Table 1 it can be seen that the water environment was extremely effective in reducing the quantity of fission products released to the environs beyond the water. Ninety-six percent of the noble gases and essentially 100% of the halogen and "solid" fission products were retained in the fuel and in the water barrier. The reactor physics data indicates that there were differences between the two tests, especially in the amount of reactivity inserted and in the peak power attained; however, the total energy release and thus the quantity of fission products available for release were nearly the same. This similarity between the two tests indicates that the difference in fission product release to the environment was almost entirely due to the water environment.

6. DISCUSSION

Most surprising of the observed phenomena was the unexpectedly small release of noble gases from the water-immersion test. Since noble gases do not react with or absorb in water to any appreciable extent, it appears that the retention of the noble gases resulted from the rapid cooling action of the water on the disrupted fuel. The quickly cooled fuel apparently regained sufficient integrity to hold the gases within the fuel matrix. A comparison of the composite photographs of the core remains from both reactors shows that the cooling action of the water was effective in reducing the amount of fuel disruption. A significant portion of the thirty-seven fuel pins was identified and recovered from the water-immersion test (Fig. 17). However, in the beryllium-reflected test (Fig. 18) most of the fuel pins were completely shattered into small unidentifiable fragments.

The absence of the release of iodines from the water-immersion test was not unexpected, and the test served to confirm predicted results. In an excursion-type accident, iodine is produced in two ways: directly by fission and indirectly by tellurium decay. If the energy release is such that a water-environment exists over the core following a destructive excursion, the iodine produced from the tellurium which escaped from the fuel would be expected to form slowly enough to insure 100% absorption in the water. The iodine produced from tellurium remaining in the fuel would also be expected to remain trapped in the fuel. The direct-yield iodine which was available for
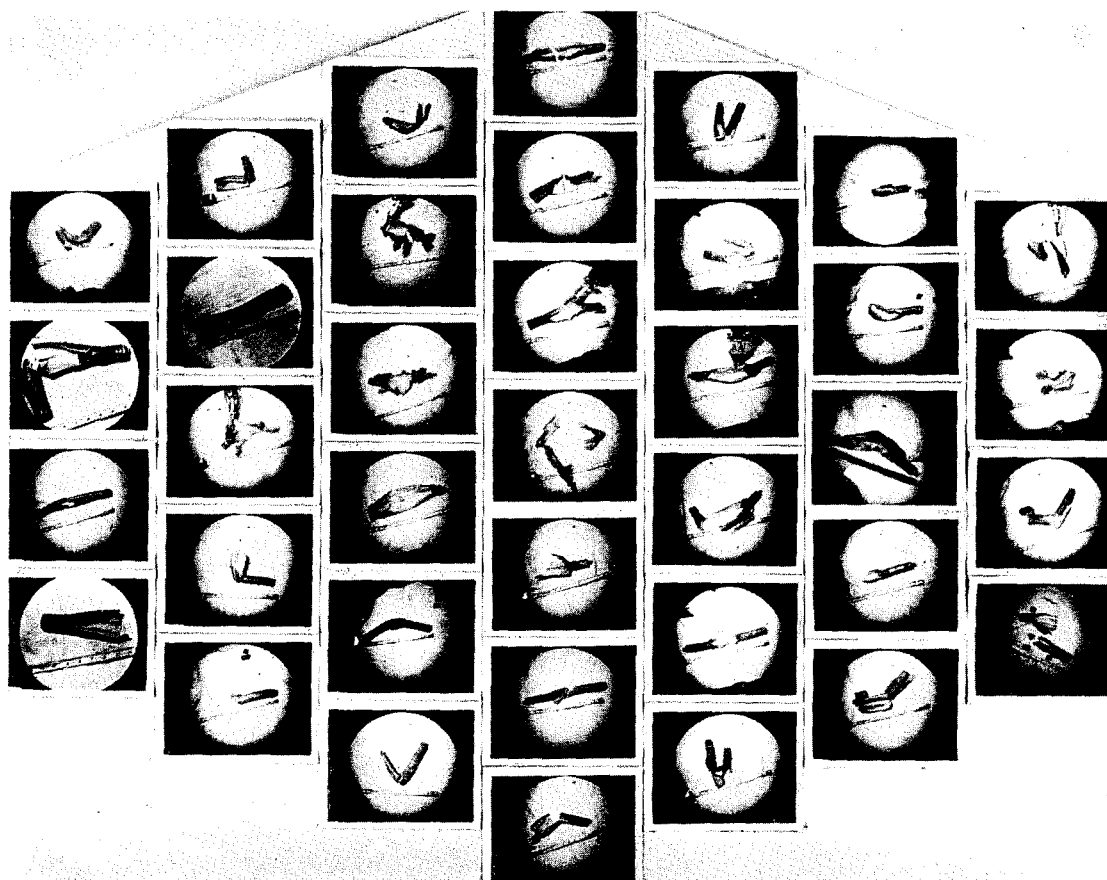
FIG. 17. Fuel rod remains from the water-immersion test arranged according to original core layout.

instantaneous release would have a high probability of reacting with and remaining in the environmental water since most forms of iodine readily dissolve in water, and the vigorous mixing of the fuel and water during the excursion should provide ample contact. In addition, the fuel from these particular reactors contained copious quantities of hydrogen which would be expected to dissociate from the zirconium-hydride lattice. The immediately available direct yield iodine could be expected to react with the hydrogen to form highly soluble hydrogen iodide, [7] once again keeping the iodine confined to the water environment. Also it is likely that the NaK coolant used during the water immersion test had some retention effect upon the iodines released from the fuel. The SNAPTRAN test did not demonstrate which mechanism was responsible for the retention of the direct yield iodine, only that the combination of the mechanisms was completely effective. Further experimentation with smaller capsule experiments could probably determine if the hydrogen and/or NaK were required for the observed iodine retention.

The release of a large fraction of the tellurium fission products from the beryllium-reflected test was another surprising result of these tests. While both tellurium and the other "solid" fission products were completely retained in the water-immersion test, 45% of the tellurium and 5% of the other "solid" fission products were released from the beryllium-reflected test. In most reactor safety analysis work, the telluriums
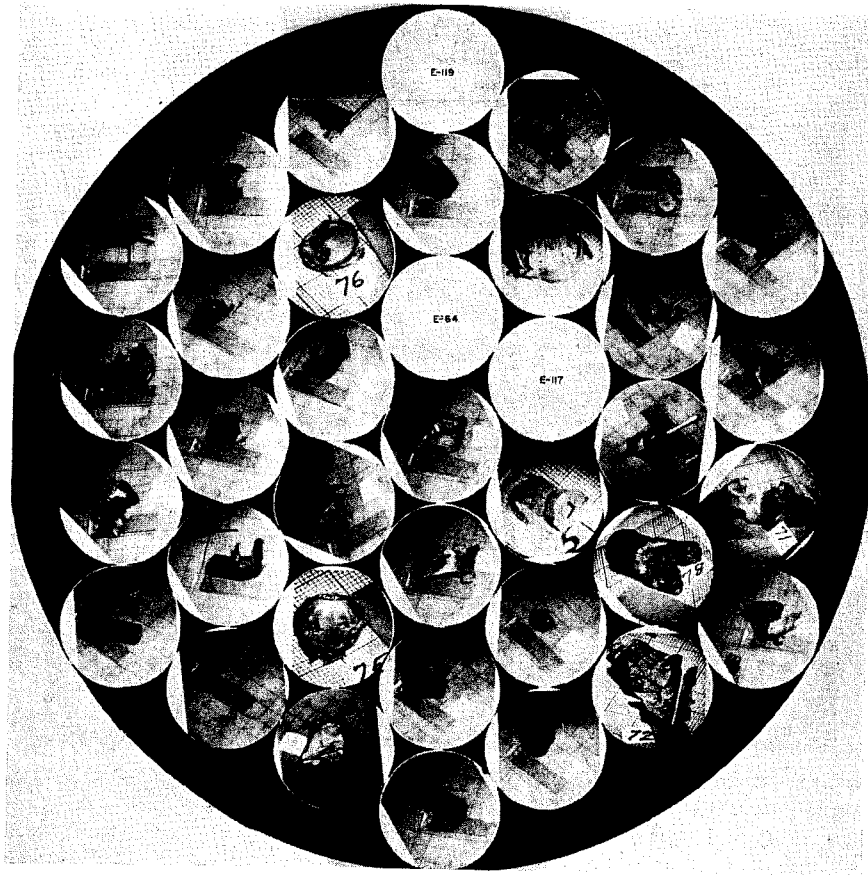
FIG. 18. Fuel rod remains from the beryllium-reflected test arranged according to origina core layout.

are grouped with the "solid" portion of the fission products. These SNAPTRAN tests indicate that this method of grouping is valid only for the water-immersed-type accident and that significantly larger release fractions of tellurium must be considered for reactor excursion accidents not immersed in water.

7. CONCLUSION

The use of full-scale integral-core reactors in these SNAPTRAN tests has demonstrated the effectiveness of a water environment in suppressing the release of fission products in a manner which is free of approximate and conservative scaling methods inherent with small scale experiments. Experimental results show that 96% of the noble gases and 100% of the

halogens, tellurium, and remaining "solids" were retained during the water-immersion test while only 25% of the noble gases, 30% of the halogens, 55% of the tellurium and 95% of the "solids" were retained during the beryllium-reflected test.

The extraordinarily high retention of the noble gases during the water-immersion test was apparently due to the quick cooling action of the water on the fuel, causing the individual gas atoms to be trapped into the remaining fuel matrix. The complete retention of iodine by the water-immersion test confirmed predictions that iodine produced from tellurium decay would be released sufficiently slow to insure complete absorption by the water, while iodine produced by direct yield would either combine

with free hydrogen or NaK and be absorbed in the water or would dissolve in the water directly. Further work is required, however, to determine if the large amount of hydrogen available during these tests is required to obtain the observed retention of iodine.

The surprisingly high release fraction of tellurium (45%) from the waterless beryllium-reflected test when compared with the 5% release fraction for "solids" demonstrates that tellurium can be classed as a "solid" only when the reactor is immersed in water. For the safety analysis reviews of reactors without a water environment, fission product release inventories will have to consider a higher release of the tellurium.

In summary, these SNAPTRAN tests, although conducted for different purposes, have successfully demonstrated some of the suspected conservatism in the assumptions used in our standard analytical models for performing safety analysis reviews and suggest that the full-scale, integral-core-type tests are the most effective way of obtaining the integrated effects of reactor accidents.

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