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SAFETY ANALYSIS OF PLUTONIUM LOADINGS IN ZPR-III

[Addendum to ANL-6408, Hazard Evaluation Report on the Fast Reactor Zero Power Experiment (ZPR-III)]

by

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INTRODUCTION

The recently revised ZPR-III hazards report[1] constitutes the basis for operations of the ZPR-III with uranium fuel.

There is at present considerable interest in extending the scope of ZPR-III studies to include plutonium-fueled critical assemblies, since there are no existing experimental data in the field of dilute plutonium critical studies for the fast power reactor program. A program of critical experiments for obtaining such information is planned for ZPR-III.

The ZPR-III machine is a framework of square stainless steel tubes in which critical masses can be assembled. Its dimensions and composition, together with its inventory of materials, are adaptable to fast reactor studies. It is built in two halves, one half being fixed and the other movable such that when assembled it forms a 1.7-m cube. About half the reactor under study is built in each of the halves of the machine. The reactor is then brought to criticality remotely by the travel of the movable half to a position adjacent to the fixed half, and subsequent adjustment of control rod positions. Figure 1 shows a view of the reactor with the halves separated.

An initial approach to criticality with a new composition or geometry is made in a series of loadings increasing stepwise toward the estimated critical mass from a conservative initial loading. Subcritical multiplication is checked with the halves together after each loading in the series, and this information determines the subsequent loading. After criticality has been reached, succeeding changes in the reactor loading are kept within the range of the control rods.

There are ten movable rods for control and safety, all loaded such that they have their maximum worth when fully inserted. Control and safety rods are identical mechanically, but differ in function in that the eight safety rods must be driven in before startup and thus they are in a position to

remove reactivity if required; the two control rods must be left out until the halves are driven together, at which time they are inserted the amount required to reach criticality.

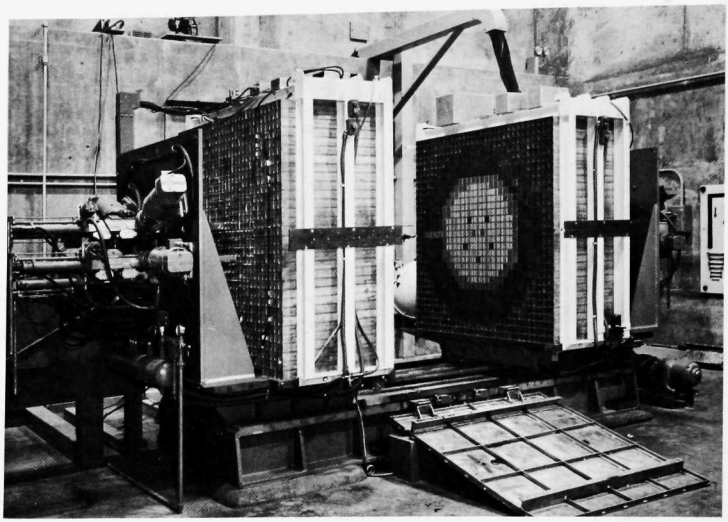

Fig. 1. Argonne Fast Critical Facility (ZPR-III)

A source has been used during startup with uranium loadings, and its use will be continued although it is probably redundant with plutonium loadings. The source is generally removed after criticality is achieved.

The system of interlocks and instrumentation is described in the ZPR-III hazards report.(1) Operating instruments for the plutonium program are to be the same as for the uranium studies, except that temperature instrumentation will be more extensive for plutonium cores, with more emphasis on alpha monitoring.

At present, 210 kg of plutonium are available in a form for use in ZPR-III. It is hoped that an additional 20 kg of plutonium, rich in the higher isotopes, may be obtained.

This report supplements the ZPR-III hazards report and specifies those revisions required for use of the above quantities of plutonium. Permission is desired to use the available inventory of enriched uranium fuel together with the plutonium, or to use either fuel separately. The rules for the handling and use of plutonium proposed in this report are in each case as restrictive or more restrictive than those for handling and using uranium. It therefore follows that any reactor system containing

both uranium and plutonium will be operated under the more restrictive plutonium rules, unless the plutonium is present only to the extent of a small reactivity coefficient sample (<1 kg) with the balance of the fuel enriched uranium.

The reactor is located at the National Reactor Testing Station, about 50 miles west of Idaho Falls, in the same area as the EBR-I, AFSR, and BORAX facilities. Outside of these facilities, which are within one-half mile, there are no other populated areas closer than four miles.

SUMMARY AND CONCLUSIONS

The ZPR-III fast zero-power reactor experiment has been operated by Argonne National Laboratory at the NRTS in Idaho since 1955. During this time the facility has been used for the study of about 40 distinct core configurations based on U^{235} fuel. Critical masses of these assemblies have ranged from 27 to 575 kg of U^{235}. The reactor has been started and brought up to operating level more than 5000 times. Personnel have been trained and qualified for ZPR-III operation in accordance with ANL policy.

The experience we have had tends to increase our confidence in the overall safety of this reactor system.

Plutonium loadings in ZPR-III were not discussed in the original hazards report, although there was no doubt that such loadings would eventually be contemplated. Since plutonium loadings were visualized in the conception of ZPR-III, the reactor was constructed in such a fashion that it could, with a few modifications, be adapted for such loadings.

The plutonium is to be put to use in the same sort of research programs as have been carried out for enriched uranium. Within the limitations of safe practices, it is proposed to take full advantage of the extreme flexibility of the ZPR-III machine for the construction of critical assemblies varying in shape, size, and composition. These assemblies will be fully reflected or otherwise protected from accidental increases in reactivity due to reflection. Core compositions will include simulations of proposed fast reactors as well as simplified compositions for validation of analytical methods. Critical mass is the basic experimental information obtained. The facility is also to be used for measurements of the worth of changes in composition and geometry. Fission rate traverses, activation measurements, spectral indices, heterogeneity effects, pulsed neutron experiments, oscillator experiments, Doppler experiments, and other experiments which can be performed within the safe limitations of the machine are to be included in the program. Metallic and ceramic versions of plutonium fuels are to be included. To simulate an oxide of plutonium, an oxide of an inert

substance will be added to the core composition. Carbides will be simulated by the separate addition of graphite to the core. Mixed uranium and plutonium cores are contemplated, as well as those using plutonium alone.

In undertaking experiments with over 200 kg of plutonium, Argonne is fully aware of the responsibilities to public safety and to its own personnel which are involved. Plutonium is one of the most toxic substances known, the maximum permissible amount in critical organs being 0.3 to 0.5 μg. Spread uniformly over the ground surface, this 200-kg quantity could contaminate more than 80 square miles to the unacceptable level of 1 mg/m^2. Careful consideration has therefore been given to the program details to insure that such a major release of plutonium is quite incredible.

It is believed that the considerations given in the following sections show that the design of the plutonium fuel is sound, that it can be handled without undue radiation hazard, and that it can be satisfactorily contained and monitored for containment during normal operations. Minor modifications of the ZPR-III machine will insure that reactivity additions take place at properly reduced rates.

A multitude of precautions has been taken against the possibility of a nuclear excursion, and this coupled with the successful history of safe operation with uranium indicates that a nuclear excursion with this facility is not credible. Because of limited experience with canned plutonium and with the pyrophoricity of plutonium and uranium, leaks, spills, and fires with plutonium are regarded as the most serious accidents which are even remotely credible. Even in this field, considerable evidence exists concerning the safety of plutonium, and this evidence is reviewed in this report. The maximum accident involving a remotely credible plutonium fire is estimated to release about 200 g of plutonium outside the ZPR-III building, with no excessive doses possible to areas beyond 11 miles.

Operations, management of the facility, maintenance, and all other features of the project will be the same as indicated in the hazards report, except where special problems associated with the use of plutonium are anticipated.

In view of the remoteness of the site and the high degree of protection, both administrative and built-in, and in view of the desirability of plutonium critical experiments to support the fast breeder reactor program, it is considered appropriate that Argonne proceed with the plutonium experiments under the conditions outlined in the following report.

The special considerations for plutonium fall into seven categories as follows:

I. Fuel Design and Fabrication

II. Fuel Neutron and Gamma Activity and Fuel Handling

III. Fuel Alpha Activity and Spontaneous Heating

IV. Plutonium Storage, Containment, Pyrophoricity and Air Monitoring

V. Operational Procedures Dictated by the High Reactivity and Low Delayed-Neutron Fraction

VI. Temperature Effects, Expansion and Doppler

VII. Accident Calculations

I. FUEL DESIGN AND FABRICATION

About 35 kg of plutonium fuel were received in 1960 and later in that year another 175 kg were ordered. The larger order was specified to match as closely as possible to the smaller order in total Pu^{239} content per piece and in the overall outside dimensions of the pieces, so that in operation all pieces would be interchangeable. The following paragraph summarizes the fabrication details.

The fuel consists of plutonium plates alloyed with 1.1 ± 0.1 w/o aluminum (9 a/o). The Pu^{240} content is 4.5 ± 0.3%, and thus may run as high as 4.8% of the total plutonium. Plutonium-241 is present to the extent of 0.5% and Pu^{242} is 0.04%. The balance of the plutonium, about 95%, is Pu^{239}. Total impurities other than plutonium and aluminum are limited to less than 0.3%. The dimensions of the Pu-Al core are approximately 0.085 x 1.765 in., with the length dimension in three sizes: 0.925, 1.925, and 2.925 in. They are clad with 1.5-mil nickel of sufficient integrity that no "wipe count" is obtained after coating. The nickel plating prevents contamination of jackets and equipment during assembly and welding operations. Midway in the manufacturing run it was determined that the nickel coating was not required for the prevention of contamination of the jackets and equipment. It was therefore not included on some of the later pieces. A record is kept of the weight of each piece, but in practice the pieces of any one size are usually considered interchangeable. The Pu-Al core plates are inclosed in a jacket of 0.012-in. Type 304-L stainless steel. The cans (jackets) are closed by end plugs of the same material, welded in place. Inside the can, at the end containing the welded end plug, a spring bears against the core slug, pushing it into contact with the opposite end of the can. The welds are tested for leak-tightness with 7-psia helium, using a mass spectrometer helium leak detector. The cans are welded under a 50/50 helium-argon mixture at $\frac{1}{2}$ atm and then buffed and polished and checked for contamination. Alpha activity from the surface of the cans is less than 1 count/min-cm^2. All pieces have been X-rayed to verify

the position of the spring. This is the fuel that is now available. Plutonium of different isotopic composition, or with slightly modified canning procedure may be used, but not to exceed approximately 230 kg total plutonium.

A summary of some of the physical and metallurgical properties of the Pu-Al alloy is given in Appendix B.

II. FUEL NEUTRON AND GAMMA ACTIVITY AND FUEL HANDLING

A. General Policy

Operating procedures are to be so arranged that personnel exposures to radiation are kept within the levels currently acceptable to the AEC and Argonne National Laboratory. In view of the fact that normal handling of the plutonium pieces may bring about more exposure to radioactivity than our personnel have encountered with uranium, exposures have been recalculated, and some rehearsals of the handling operations have been performed and monitored. Based on these calculations and rehearsals, procedures have been proposed with which personnel need not incur excessive exposure. The procedures will be subject to further monitoring and can be modified if this should prove necessary. The radiation estimates and handling procedures are outlined below.

B. Estimated Activities[2]

The spontaneous fission of the plutonium isotopes results in 2.70×10^{-13} c/g from Pu^{239} (1 c = 3.7×10^{10} d/sec), 1.22×10^{-8} c/g from Pu^{240}, and 2.11×10^{-8} c/g from Pu^{242}. The spontaneous fission of even-odd* Pu^{241} is expected to be of the magnitude of Pu^{239}. Therefore, considering the analysis of the plutonium used, the Pu^{240} with two neutrons per spontaneous fission is the principal source of neutrons. An assembly containing 200 kg plutonium may contain about 10 kg Pu^{240}, which would comprise a distributed source of 10^7 neutrons/sec. This could amount to as much as 500 n/sec/cm^2 over a large fraction of the body area of a person standing between the halves, depending somewhat, of course, on the geometry involved. This is about ten times the tolerance level for a 40-hr/week exposure. The calculation neglects the effects of absorption and moderation within the assembly, but it also neglects multiplication. The problem is considered serious enough that the neutron field between the halves and near the assembly will be monitored each time a new size of assembly is constructed, and personnel exposure limited accordingly. The handling of individual pieces and drawers containing up to 2 kg of plutonium will be monitored in a similar fashion.

*In the absence of reliable experimental data, the even-odd characteristics of the proton and neutron numbers of the isotope are taken as a rough indication of the tendency to spontaneous fission.

The X-ray and gamma activities of plutonium may also present a problem. These activities are associated with the alpha or beta activities of the isotopes and are generally much shorter than the spontaneous fission half-lives. Plutonium-239 has 0.0611-c/g alpha activity. About 0.3% of the disintegrations are accompanied by gamma photons of 40 to 120 kev, and about 3% are accompanied by X rays of energy around 20 kev. Although in a large assembly this may come to a total source equivalent to some hundreds of curies of pure gamma emitter, it is expected that absorption in the assembly and in the air will take care of most of this. For example, 95% of the X rays are attenuated just in passing through the stainless steel jackets of the fuel pieces. Because of its shorter half-life, Pu^{240} has a specific activity of 0.226 c/g of alpha activity. Moreover, about 24% of the Pu^{240} disintegrations are accompanied by the 0.043-Mev gamma of U^{236}. Therefore 10 kg of Pu^{240} could result in the equivalent of 600 c of a single gamma emitter, but again because of the softness, much of this is absorbed internally. Finally, consideration must be given to Pu^{241}, whose beta activity amounts to 110 c/g, and although this is not accompanied by any observed gamma, the 470-yr daughter, Am^{241}, which builds up as the 12.5-yr Pu^{241} decays, does emit gammas. In fact, 20 months after production, the 1.2 kg of Pu^{241} present in the original inventory would have already produced 100 g of Am^{241} (324 c). About 40% of the Am^{241} decays are accompanied by gammas, mostly at 0.059 Mev, and by 18-kev X rays. Again the X rays are rather soft, but because the gammas can build up to a considerable amount in terms of total emissions, the vigilance of monitoring must be maintained for many years.

It is not anticipated that operation of the critical facility with plutonium will build up any significant quantity of long-lived fission products. Seven years of operation with uranium has not built up long-lived activities to the extent that any special remote handling procedures have had to be established. The natural activity of the plutonium is such that the superposition of the long-lived fission products on top of it will not cause additional handling problems. The activity from short-lived fission products, both between the halves and in the neighborhood of individual drawers and pieces is discussed for uranium in the hazards report. In general, the same figures will apply for plutonium, except that plutonium critical masses for a given volume will be expected to be about 0.6 the corresponding uranium critical masses. Consequently, the fission products per gram of fuel for a given integrated power of operation will be about 1.7 times as great in the case of plutonium. In the hazards report, the values listed in the table headed "Radiation Intensity" are calculated for a 60-liter core with 14 v/o U^{235}. A plutonium core of this size would have about 0.6 this fuel density. Because of the canning material, an individual plutonium plate would contain only about two-thirds as much fissile material as the same nominal size uranium plate. The fission product activity at the surface of the plutonium plate would therefore be increased by a factor of about 1.2 over the corresponding values listed for uranium. In normal operation this would still be small compared to the plutonium activity.

C. Time-and-exposure Study for Drawer Loading

Measurements have been made of the time and exposure for operations involving individual drawers loaded with typical quantities, 0.5-1.0 kg of plutonium. Starting from storage in the vault, the following are the operations:

1. Take the can from the vault and unbolt the top (40 sec).

During this operation the plutonium is still in the container and partially shielded. The hands are exposed to radiation from the plutonium cans at a distance of about 2 in. through the container lid in a 15-mr/hr gamma field. Body exposure is below measurable levels.

2. Monitor the cans for alphas (15 sec).

The lid is off the container; hands are 6 in. from the plutonium in a 80-mr/hr field of betas, and soft and medium gammas. The nearest parts of the body are about 24 in. from the plutonium in a 2-mr/hr field.

3. Carry the container from monitoring hood to loading hood (5 sec).

The top is replaced on the container but not bolted. Hands are 2 in. from the fuel in a 15-20 mr/hr gamma field. Body exposure is negligible.

4. Load plutonium from container into ZPR-III drawer (25 sec).

Twenty-five seconds were required to load six pieces (about 0.5 kg) into a prepared drawer. Fingers are in contact with the pieces in a field of about 262 mrem/hr (12 mrem neutrons, 250 mrem gammas). The body is exposed to only 2 mr/hr at 24 in.

5. Bolt the top back on the container and return it (partially filled with fuel) to the vault (40 sec).

The hands are in a 2-mr/hr field 2 in. from the fuel and shielded from it by the container lid and walls.

6. Place the loaded drawer on the cart (5 sec).

The hands are 2 in. from the fuel in a 20-mr/hr field. Body is 24 in. from the fuel in about a 2-mr/hr field.

7. Push the cart into the assembly room near the reactor (30 sec).

The hands are at least 6 in. from the fuel in a 5-mr/hr field. The body is 24 in. away in a 2-mr/hr field.

8. Take drawer off cart and place in ZPR-III matrix (15 sec).

Hands are at 2 in. in a 20-mr/hr field, body at 18 in. in about a 5-mr/hr field.

The distances given above are the points of nearest approach. Measurements were taken with a Juno meter calibrated for 1-Mev gammas, although the plutonium radiations are less energetic. The sensitive volume of the Juno meter can only be moved to within 2 in. of the sample, and therefore the surface contact field is from an additional measurement made with smaller probes.

Although neutron measurements were not taken in all phases of this study, an exposure estimate can be made from the times and distances. An overconservative assumption would be that loading a 1-kg drawer involved a 3-min exposure at a total body distance of 10 cm. The dose rate from a 1-kg point source (containing 60 g Pu^{240}) at 10 cm is 45 $n/cm^2/sec$. Allowable exposure per week at this dose rate is 2/5 of 40 hr, or 16 hr. Since there may be as many as 150-200 drawers involved in large loadings, the time required to load all drawers would be 450-600 min, or 8-10 hr. Thus, as far as handling individual drawers is concerned, one individual could make essentially a complete loading and unloading once a week.

9. Exposure between matrix halves.

Exposure may be incurred while loading drawers into the assembly from drawers that have already been loaded. A subcritical plutonium assembly which was constructed in July 1961, provides some data on the radiation fields developed between the halves.[26] This had 20 kg of plutonium in a 24-liter volume in one half of the machine, with the fuel density corresponding to an EBR-II mockup - about 0.83 kg/liter. The gamma level was 6 mr/hr midway between the halves, and 18 mr/hr at 14 in. from the interface (measurements were taken with a Juno meter and are corrected for U^{238} betas). The fast neutron field midway between the halves was about 70 n_f/cm^2-sec. Thus, even with this small quantity of plutonium, neutron fields are appreciable. Personnel are monitored and exposure times limited in accordance with established Health Physics procedures.

III. FUEL ALPHA ACTIVITY AND SPONTANEOUS HEATING

The spontaneous alpha decay of Pu^{239} generates about 1.9 w/kg, and of Pu^{240}, 6.9 w/kg. Americium-241 generates 106 w/kg. The buildup of Am^{241} from the 12.5-yr Pu^{241} would add, after 12 yr, about 50 w to the 450 w generated by the total plutonium inventory. These heat generation rates are sufficient to produce sensible heating of large masses of plutonium which are left assembled for a long time. It is difficult to calculate the magnitude of the temperature rise in ZPR-III because of the many interfaces between pieces, making it difficult to estimate an equivalent thermal conductivity for the assembly.

Measurements were therefore made on the subcritical assembly mentioned in the previous section to determine the temperature rise in a source of 20 kg plutonium, distributed in 24 liters, blanketed with full-density depleted uranium, in one-half of the ZPR-III machine. Other diluents with the plutonium simulated the EBR-II composition. The temperature effect of having the two halves together was simulated by placing 3 in. of Fiberglas insulation over the interface. Equilibrium temperature profiles measured in the core indicated an equivalent thermal conductivity of 0.01 cal/sec-cm-°C in the axial direction and of about 0.0015 cal/sec-cm-°C in the radial direction transverse to the plates. In the blanket, conductivities were larger by a factor of about two. (The thermal conductivities of pure stainless steel, uranium, and aluminum are 0.05, 0.06, and 0.5 cal/sec-cm-°C, respectively.) If an average conductivity, 0.004 cal/sec-cm-°C, is assumed to apply isotropically in the core and 0.01 cal/sec-cm-°C in the blanket, the temperature rise in a 210-kg core, from outer edge of blanket to core center, is calculated to be about 36°C.

With ambient temperatures of 25°C, this results in 61°C temperatures at the center of a 210-kg plutonium core, which would not cause phase changes or structural damage to the fuel. Consequently, no external cooling system is considered necessary. Smaller ZPR-III cores, in which the fuel is more concentrated, have a smaller temperature rise.

Measurements were also made on the length of time required to attain temperature equilibrium in the mockup. At least four days were required to establish an equilibrium pattern of temperatures throughout the assembly.

IV. PLUTONIUM STORAGE, CONTAINMENT, PYROPHORICITY, AND AIR MONITORING

The low biological tolerance level for plutonium together with the pyrophoric nature of some of its forms dictate an extremely dependable degree of containment. Monitoring for alpha activity is the best way of

checking on the presence of plutonium, but this is complicated because working areas must also be used for uranium. The uranium is an alpha emitter also, with most of its alpha activity coming from U^{234}, generally present in about one percent of the U^{235} concentration. This increases the general background of alpha activity, making the presence of plutonium more difficult to detect than otherwise. The maximum energy of Pu^{240} alphas is 5.16 Mev, compared with the maximum energy of the U^{234} alphas of 4.77 Mev. This distinction offers some possibility of detecting the plutonium with a carefully biased alpha proportional detector. Principal reliance, however, must be placed on monitoring for any sudden or large increase in alpha background such as would indicate air-borne plutonium or surface contamination emanating from a ruptured can.

A. Monitoring and Handling Procedures

Two 150-cfm hoods have been designed and built for use in the loading room. These hoods are approximately 34 in. wide x 48 in. long x 36 in. high. One of these is portable and is placed on the loading room table during plutonium operations or removed for uranium operations. The other hood is permanently installed in the loading room near the vault door. Each hood has its own filtered exhaust.

The procedure for examining and handling the plutonium has been worked out in some detail, and the following steps are specified:

1. The plutonium in the vault is stored in flanged and bolted containers centered in birdcages. These containers are carried into the first (permanent) hood where they are unbolted and opened. Only 3.5 kg of fissionable material are permitted in this (or the second) hood. The inside of the containers above the plutonium pieces is monitored with an alpha surface probe. Any rupture of a plutonium can which has occurred in storage is expected to emit sufficient plutonium to be easily detected in this fashion. This first hood is monitored with an air-sampling monitor.

2. The containers are then moved to the portable hood on the loading table. A plutonium piece counter (2π proportional counter) will be located inside this second hood and will be used to check individual pieces. A plutonium impactor monitor (for air-borne plutonium detection) is connected to the second hood at all times when plutonium is being handled in the hood. The monitor will indicate 0.5 MPC in 5 min.

3. A plutonium impactor monitor is used in the reactor room and started prior to reactor operation.

4. Upon completion of reactor operations, a waiting period of 10 to 15 min is allowed for a complete cycle of the monitor. Upon completion of the monitoring cycle, if the air activity is normal, entry is made into the reactor room.

5. If plutonium is to be removed from the reactor, an impactor monitor is connected to the appropriate hood for monitoring. All temporary storage and drawer unloading must take place in the second hood. Plutonium materials unloaded in the hood are sealed in metal containers prior to their removal from the hood.

6. All handling of the plutonium impactor monitors and backup monitoring for plutonium is done by Health Physics personnel unless in a specific instance such work is delegated to the Operating Supervisor on duty.

7. The use of protective clothing, specifically of rubber gloves, will be rigidly enforced. Other protective measures, such as the use of tongs, lead gloves, and portable shields, will be adopted as monitoring indicates they are necessary.

B. Pyrophoricity

Both uranium and plutonium are known to be combustible in air. The rate of burning and the extent to which a fire propagates itself throughout a large mass are related to the ventilation rate, the rate of heat loss, and the formation of protective coatings. In large samples of relatively pure solid materials, the heat conduction to the bulk of the material is such that a fire initiated at a point will not ordinarily propagate throughout the mass. Excerpts from recent experimental studies on this subject are the following:

"Studies of various thicknesses of uranium foils have shown that strips greater than 1.5 mm thick will not continue to burn in air when the heat from the igniter is removed";(3) and "several attempts were made to burn 60 mil (1.58 mm) diameter uranium wire and 0.13 x 1.0 mm zirconium foil strips at an ambient temperature of 400°C. Neither of these two samples would burn in air with the surroundings at room temperature, although 30 mil uranium wire and 0.13 x 0.6 mm zirconium foil will burn in room temperature air. Heating the surroundings to 400°C, however, would not produce a self-sustaining combustion."(4)

In regard to plutonium:

"The oxidation behavior of molten plutonium held at 900°C in an air atmosphere has been reported by Hanford. Two 800 g pieces, contained in a magnesium oxide crucible, oxidized slowly, resembling burning charcoal. In contrast, plutonium turnings burn brightly. A plutonium oxide coating formed over the molten metal, protecting the metal from further oxidation. Even with intermittent stirring, it was not possible to oxidize completely the massive plutonium. It was necessary to cool, remove the oxide, and reheat the metal twice before oxidation was complete."(5)

Hilliard also reports that burning of uranium samples, 0.127 to 0.515 in. minimum dimension, was not sustained in air when the external heat supply was removed.(6)

It cannot be said that instances of self-sustaining combustion of massive uranium pieces are unknown, however. Schnizlein et al. cite the example of a truck loaded with half-inch-thick pieces of uranium, which caught fire after a motor accident. The fire lasted nearly 24 hr and could not be extinguished.(7) In the light of the more recent laboratory experiments, it seems likely that these instances of combustion of massive uranium must have been influenced by unusual grain size or irregularity, metallurgical history, porosity, the presence of external heat sources, or some of the other special mechanisms which are known to promote metal fires.

In the event that the core was raised to a high temperature, alloying between the plutonium and other materials would be a possibility. Plutonium forms a eutectic with iron at about 410°C, for example. Some information on the oxidation rate of various alloys of plutonium with iron, thorium, zirconium, uranium, and uranium-molybdenum, has been reported from Harwell.(8) The various alloys were heated to 450°C or more in dry air and oxidation rates determined. In no case were the rates greater than for pure uranium. The alloys tested were 15 and 25 a/o Pu in Fe; 15 and 30 a/o Pu in Th; 10, 20, and 40 a/o Pu in Zr; 5 a/o Pu in U; and 20 a/o Pu-25 a/o Mo in U. It was also found that the oxidation of the Pu-Zr alloys at 700°C in CO_2 proceeded at rates comparable to 450°C in air. Many of the alloys normally expected to be formed in plutonium cores are therefore not any greater a combustion hazard than the materials heretofore present in the uranium cores.

Most of the non-fuel materials used in ZPR-III are noncombustible. However, for cores containing sodium it must be assumed that if combustion were started it would more likely proceed to completion because of the self-sustaining properties of the sodium. Graphite is relatively difficult to burn and is not expected to add to the combustion hazard.(9)

C. Dispersion Accompanying Combustion

There exists a miscellany of results on the dispersion of plutonium and uranium after fires and explosions. Although none of the data exactly duplicate the ZPR-III situation, general patterns are apparent which may have some bearing on the proposed operation. In the following paragraphs, summaries of a few dispersion studies are presented.

Carter et al.,(10) have investigated the combustion of plutonium cylinders and turnings, and the dispersion of the products into the atmosphere. The combustions took place in a light draft (chimney) of dry air,

and the air-borne combustion products were collected on filters. They estimated that only the particulate matter smaller than 3μ in diameter need be considered an inhalation hazard in the lungs, because of both the rapid fallout and low retention of larger particles. They determined that only about 0.05% of the residue from the cylinders (0.01% from the turnings) was in the hazardous size range. They concluded that with no dispersal agent present, only 10^{-4} of the original plutonium burned would constitute an inhalation hazard. Even in a turbulent air stream, the fraction which constituted an inhalation hazard could not exceed 5×10^{-4}, since this was the maximum amount in the size range below 3μ in diameter.

The 3-μ upper limit for the biologically objectionable particles has been disputed by more recent studies,[11] which indicate that 10μ is a safer limit to use.

Burning tests have been reported for plutonium by Cheever,[12] with the object, among others, of determining the percent of plutonium which became air-borne. Tests were conducted in a plutonium glovebox system. The box atmosphere was filtered room air, with moisture content 30 to 70 grains per pound of dry air. One to two grams of plutonium in the form of slab, cylinder, or turnings were ignited by a resistance-heated Nichrome wire. Diffraction studies indicate that PuO_2 was the oxide formed. The fraction of the burned plutonium collected on filters amounted to only 1.2×10^{-5} in the maximum case observed. Air velocity through the filters was given as 5 fpm, but no estimate was made of air velocity in the combustion region.

Studies of burning uranium have been made by Conners and O'Neil,[13] who believe that the physical characteristics of uranium oxide fumes are similar to those of plutonium. Uranium turnings were ignited by means of a Nichrome heater in a porcelain dish in a combustion hood. Dimensions of the hood indicated average air velocities up to 30 fpm in the combustion area. The fume appeared to be the black oxide, U_3O_8, of specific gravity 7.31. The order of magnitude of the fraction air-borne was estimated by taking air samples near the entrance to the exhaust system. Values of this fraction ranged from 4×10^{-6} to 7×10^{-5}, with a single observation of 2×10^{-4}. The weighted mean was 2.6×10^{-5}.

Hilliard has summarized a number of references concerned with the dispersion of plutonium following combustion.[14] Although he is particularly interested in the case in which irradiated uranium, containing some converted plutonium, undergoes combustion, his studies also include the case of pure plutonium. After showing that the evaporation of plutonium in a vacuum from mixtures with uranium correlates well with theory, he then extends the theory to cover the evaporation into air at various velocities. His conclusion from these studies is that evaporation cannot make a significant contribution to the amount of plutonium dispersed at the temperatures prevalent in self-sustaining fires, i.e., up to 1350°C.

He thus reasons that the fine particles, $<3\mu$ in diameter, of non-volatile oxide are the chief means of dispersal. Noting that Carter et al.[10] have already set an upper limit on the amount of plutonium which forms fine particles, and permitting a slight extension of this amount in order to include other piece sizes, he arrives at 0.08% as the maximum fraction of the plutonium which might result in an inhalation hazard following a fire.

When the oxidation is accompanied by an explosion, contamination is likely to be much more widespread. The Oak Ridge explosion of November 1959[15] was not a complete dispersion however. In this instance, an evaporator containing about 1300 g plutonium in solution exploded inside a shielded cell. Although the heavy walls of the cell prevented extensive damage from the explosion, a door was blown open by the force of the blast, permitting direct escape of some contamination.

No nuclear explosion was involved. The evaporator was estimated to have contained about 3 kg picric acid, some alkali nitrates, and about 18 liters of nitration products which had accumulated from an organic decontaminating agent. Peak explosion pressure was assumed to be about 1100 psi.

Of the 1300 g plutonium originally present, 1100 g was recovered from the damaged equipment, 150 g was flushed and recovered from the interior walls and apparatus of the cell, and smaller amounts from other locations. It was estimated from smear samples that about 600 mg escaped from the building through the door which was forced open. Fallout was rapid and was confined to a small fraction of the site area.

This would seem to be an example of a partially contained blast. The heavy shielding walls of the cell absorbed some of the force of the explosion, but a large portion of the energy was certainly directed out the opened door. The fraction released to the outside, 5×10^{-4}, was no doubt limited by the partial containment. Although an open explosion would certainly be expected to spread a larger fraction of its material over a wider area, the example indicates that even partial containment can be very effective in confining a large fraction of the explosion products.

Additional pertinent references to plutonium and uranium dispersal are found in the classified literature.

In summary, it seems reasonable to expect that the fraction of plutonium dispersed following a fire which would actually be an inhalation hazard would not be greater than 10^{-3} of the amount oxidized.

V. OPERATIONAL PROCEDURES DICTATED BY THE HIGH REACTIVITY AND LOW DELAYED-NEUTRON FRACTION

A. Delayed Neutrons - Startup Considerations

Depending somewhat on the energy of the neutron which causes fission, there is about 0.0022 delayed neutron per prompt neutron in the case of Pu^{239} compared with 0.0069 for U^{235}.(16) The corresponding number for U^{238} is about 0.018, but the higher fraction of delayed neutrons present in cores containing U^{238} is somewhat offset by the fact that the delayed neutrons are emitted at an energy too low to cause fission in U^{238} and, hence, are less effective than the prompt neutrons. The net result has been that over the wide range of compositions studied so far in the ZPR-III, the "effective beta" has varied over a relatively narrow range of about 0.007 ± 0.0003.

"Effective beta" is an important criterion in startup accidents, representing the difference in reactivity between delayed and prompt critical. Calculations of startup accidents generally assume that the length of time required to add an amount of reactivity equal to the effective beta is the maximum time available for shutdown instruments to operate. Actually, ZPR-III experience has shown that there is more time available than this would indicate, because, with an adequate source in the reactor, the instruments are operating on subcritical multiplied neutrons considerably before criticality is achieved. Thus, period scrams are possible while the reactor is still subcritical.

For plutonium loadings, the minimum "effective beta" would be about 0.0022, with higher values for loadings containing some U^{235} or U^{238}. Nevertheless, in order to be conservative, the criteria for rate of addition of reactivity in plutonium cores will be adjusted from the U^{235} cores by the ratio of the "effective beta" in these two types. Thus, if $(dk/dt)_{U^{235}}$ is the maximum permissible rate of addition of reactivity in a uranium core, then $(dk/dt)_{U^{235}} (\beta_{Pu}/\beta_{U^{235}})$ will be the maximum permissible rate in a plutonium core, or mixed plutonium-uranium core.

1. Carriage Drive

The ZPR-III hazards report(1) specifies that reactivity additions due to closure of the halves will be not greater than dk/dt equal to 5×10^{-4} per second for the final 12% of reactivity addition. For plutonium, the corresponding figure would be $(5)(0.22/0.7) \times 10^{-4}$ or 1.6×10^{-4} per second. For the geometries of fast power reactors, 12% has been taken to represent the final 3 in. of carriage travel, and this is not expected to change appreciably with plutonium loadings up to 210 kg. This last 3 in. of closure should occupy at least 750 sec (12.5 min) in order not to exceed

dk/dt of 1.6×10^{-4} per second. For the plutonium loadings, a carriage drive with a speed of $\frac{1}{6}$-in./min is used, so that the final 3 in. of closure will require 18 min.

The intermediate and high carriage speeds are based on convenience of startup operation rather than on rate of reactivity addition, since the machine is not intended to be critical until the halves are completely closed. In the unlikely event that criticality occurred before the slow-speed drive was in operation, the instruments and safety rods are fast enough to shut the reaction down, as shown in Section VII, A. Therefore the high and intermediate speeds are not reduced for the plutonium loadings.

2. Control Rod Drives

The ZPR-III has 10 movable rods, 5 in each half. Any one rod in each half may be designated as a control rod, the others being considered safety rods. Safety rods are cocked (driven into the reactor) before startup, and consequently their speed is not relevant. Control rods are driven into the reactor one at a time after the carriage is driven in. The hazards report for U^{235} loading specifies that the speed and the worth of control rods shall be such that dk/dt is not greater than 5×10^{-4} per second due to control rod insertion. The corresponding figure for plutonium then is 0.00016 per second. The control rod speed is 4 in./min or 0.07 in./sec. Control rods must therefore be loaded in such a pattern that their worth is not over 0.002 Δk/in. for plutonium loadings. In any case for which this is not possible, the control rod drive speed will have to be reduced to stay within the permissible rate of reactivity addition.

In other respects, the use of control and safety rods is the same as described in the U^{235} hazards report.

B. Reactivity of Plutonium - Worth of Loading Changes

The hazards report[1] for uranium loadings in ZPR-III contains no restriction on the increments of fuel or reactivity that may be added between runs, or on the total excess reactivity in the reactor. Nevertheless certain practices have been established which limit these activities. It is prudent at this time to review these practices for their applicability to plutonium loadings. Some of these practices are the following:

No loading plan of the ZPR-III will be approved unless estimates show that the reactor will be still subcritical when the halves are fully closed with two control rods out. If the reactor is found to be critical before the halves are fully together, it is shut down immediately. Deviations from the rule for special experiments can be carried out, as regulated by the ANL Policy and Practice Guide.[17]

During an approach to criticality with a new composition or geometry, the first loading has one-third to one-half of the estimated critical mass, depending on how much previous experience is available to verify the estimate. The second loading adds not more than an additional 20% (of the estimated total). Increments of this size are continued until subcritical multiplication indicates that this size increment might make the reactor critical with the control rods withdrawn (a safe allowance is made for possible curvature in the subcritical multiplication plot). Increments are then reduced so that criticality with the control rods less than fully out can be estimated. With conservative estimates, this practice has led to the achievement of criticality in five to ten steps. Until considerable experience has been gained with plutonium, approaches to criticality will be made with increments not over 10% of the estimated total critical mass.

It is of interest to estimate whether or not the same sizes of loading changes which have become customary for uranium-filled reactors are still safe for the equivalent plutonium reactors. The critical mass of a solid plutonium sphere is about one-third that of a solid enriched uranium sphere. The number of neutrons per fission is higher for plutonium than for uranium. In comparing assemblies of plutonium with uranium, one would probably consider as being equivalent those assemblies which have the same ratio of fissile to fertile atoms, and the same radius. Thus the fuel density in the plutonium assembly would be less than in the uranium assembly. In the range of the fast breeder reactors a ratio of fissile to fertile atoms in the core of 0.2 might be typical. Calculations[18] indicate that to have the same buckling, a plutonium assembly with this dilution would have about 0.7 the fuel density of a uranium assembly with the same dilution. It has been shown that for two reactors having the same radius and reflector savings, but different core composition, the worth of an increment of mass at the edge of the core can be expressed as $\frac{dk}{k} = \frac{1}{f}\frac{dM}{M}$, with f proportional to $\frac{\nu\Sigma_f}{(\nu - 1)\Sigma_f - \Sigma_c}$ (one-group approximation).[19] Here ν is the number of neutrons per fission, and Σ_f and Σ_c are the macroscopic cross sections for fission and capture respectively. According to currently used fast reactor cross-section sets, f would be approximately the same for a dilute uranium reactor as for a dilute plutonium reactor with 0.7 the fuel density of the uranium reactor. Thus, substitution experiments in a plutonium reactor will produce about the same reactivity change as in the equivalent uranium reactor if the experiments involve the same volume of core material in each case. It can also be seen that if the proportion of U^{238} in a core is reduced, thus reducing Σ_c in the denominator, f will be reduced (substitutions worth more), as has been observed in the uranium assemblies.

It has been the practice to avoid loadings which would incorporate more than 0.5% reactivity in the core with halves together and all rods in.

The corresponding figure will be held to 0.15% with the plutonium loadings, and the preceding observations on the value of f indicate then that the maximum volume of excess core material that can be added to a plutonium core will be about one-third what it was for the corresponding uranium core.

VI. TEMPERATURE EFFECTS

A. Expansion of Columns of Fuel in Reactor Drawers

When ZPR-III drawers are loaded with uranium fuel, the thin plates of fuel are placed end to end, starting from the center of the reactor. Thus, any temperature rise of the fuel produces expansion of the individual fuel pieces which cumulates in the expansion of the entire column of fuel. The situation is different for plutonium. The plutonium pieces are canned in individual containers each with its own internal gap. Thus expansion of the individual pieces would not be cumulative.

Each can is marked with an engraved number and with a colored mark on the end which contains the spring. The cans have been radiographed after assembly to insure that the spring is properly placed, and to locate other possible internal defects.

Each individual can of plutonium is loaded into the drawer with its spring and gap toward the back of the drawer. Individual pieces then always expand outward from the core center. Obviously, however, the reactivity reduction associated with the expansion is considerably less than when the expansions of a whole column are cumulative. In order to estimate the effect of expansion in a typical 260-liter, 227-kg plutonium ZPR-III loading, the following calculations were performed.[20] (See Appendix C.)

An SNG calculation in slab geometry (with corrections for radial leakage) with plutonium distributed uniformly over the whole core was the reference calculation. Another SNG calculation was then performed with the plutonium contracted by 4% to make 0.2-cm plutonium-free gaps 5 cm apart. There was, of course, no central gap. The assumed radial buckling was the same for both calculations. From the change in k of the SNG calculations, it was found that

$$\frac{\Delta k}{k} = -0.045 \frac{\Delta l}{l} \approx -0.5 \times 10^{-6} \text{ per } ^\circ\text{C} \quad ,$$

where $\Delta l/l$ is the fractional change in length. This procedure might overestimate the expansion due to internal heat generation by a factor of two, since the actual heating would be greatest at the center where the expansion

is least effective. Since the coefficient for expansion of full columns of fuel expelling the blanket is about

$$\frac{\Delta k}{k} = -0.34 \frac{\Delta l}{l}$$

for uniform expansion (and half of this amount for actual expansion), the plutonium loadings can be expected to have a shutdown coefficient that is 13% of the uranium loadings.

Note that as canned for these critical experiments there is no longer any compelling reason to load the plutonium fuel end to end.

B. Doppler Coefficient

The Doppler coefficient in ZPR-III cores can be positive and an order of magnitude greater than the small shutdown coefficient due to expansion. Recent calculations of the Doppler effect in Pu^{239} indicate[21] that, for a given spectrum it is 2 to 3 times as large as the corresponding effect in U^{235} (or the negative effect in U^{238}). The Doppler coefficient in core C described by Baker,[20] a typical ZPR-III plutonium assembly, is estimated at almost $+10^{-6}\Delta k/°C$, based on the recent data but ignoring the effect of heating in the U^{238}, and using Nicholsen's method of calculation.[22] Comparison of the spectra of carbide cores with metallic cores indicates that the carbides might have Doppler coefficients as large as $+5 \times 10^{-6}\Delta k/°C$.

On the other hand, if an assembly contains appreciable quantities of U^{238}, this would provide an effect at least partially compensating that of the plutonium. The temperature rise of the U^{238} would be related to the temperature rise in the plutonium by

$$\frac{T_{U-8}}{T_{Pu}} = \frac{\sigma_{f\ U-8}}{\sigma_{f\ Pu}} \frac{c_{p\ Pu}}{c_{p\ U-8}} ,$$

where the ratio of heat capacities, c_p, is 1.23 and the fission ratio is not less than 0.03 for the assemblies contemplated. Thus the ratio of temperature rise in the two materials is not less than 0.037. The Doppler coefficient of both U^{238} and Pu^{239} drops approximately as T^{-1} or $T^{-1.5}$, but since the U^{238} temperature rise is rather slight, its Doppler effect remains nearly constant at a slight negative value, whereas the positive plutonium coefficient decreases as its temperature rises. On the other hand, the Doppler importance of a given isotope is proportional to the square of its isotopic ratio. Therefore, for compositions like core C, in which the U^{238}/Pu^{239} isotopic ratio is greater than 6, the effect of the Doppler coefficient in the U^{238} is nearly that of the plutonium, and certainly the U^{238} negative effect would predominate at fuel temperatures above 400°C.

Both the plutonium and uranium effects increase in softer spectra, and in roughly the same proportion. The worst Doppler situation visualized is a plutonium carbide core with no U^{238} present. This would produce a Doppler coefficient of about $5 \times 10^{-6} \Delta k/°C$ at room temperature, and the integrated Doppler effect from room temperature to vaporization would amount to 0.0026 to 0.004 Δk.

VII. ACCIDENT CALCULATIONS

A. Excursion Kinetics

A parametric study of excursions has been made with a view to determining whether the positive Doppler effect increases the probability of an uncontrollable excursion. The initiation of any of these excursions requires some sequence of highly improbable events. It will be shown that the presence of the positive Doppler effect does not substantially increase the probability of an uncontrollable excursion. If such an excursion could be initiated, the positive Doppler effect would cause a substantial increase in the magnitude of the excursion, but not so great that the fission products represent a greater long-term hazard than the plutonium.

None of the explosions listed in this section could possibly run its complete course to self-shutdown under the operating conditions of ZPR-III. All would be stopped at an early stage by removal of reactivity initiated by period or power-level trips. The calculations permit estimates of the time available for various instruments to function and the power level at which they would become operative under various conditions of reactivity addition. At all rates of reactivity addition up to the maximum which can be added by closure of the halves at high speed, there is sufficient time and a large enough instrument signal so that the safety rods will shut the reactor down without damage.

Details of the calculations of possible explosions are given in Appendix A; a summary and analysis are given here.

The principal tool used in the analysis of possible ZPR-III accidents is the space-independent RE 129 J code for IBM 704. This code follows the course of an incident in which the reactivity of the assembly increases as

$$\frac{dk}{dt} = A + Bn, \quad 0 < t < t_0 \quad .$$

Here A is a ramp rate of reactivity addition (which may be zero), B is a temperature-dependent coefficient, and t is time. The relationship between k and n is computed from the usual kinetics equations as in other versions

of the RE 129 codes which solve the same equation. Appropriate delayed-neutron data can be inserted in the input for the particular composition investigated. Having computed the increase of n with time, the code then determines $\int n\, dt$, which is proportional to the enthalpy increase of the fuel. For the purposes of ZPR-III calculations, heat generated in the fuel is assumed to remain in the fuel, which is the worst situation as far as positive Doppler effect is concerned, and is overly pessimistic for excursions that start slowly. The time constant for the transfer of heat from the fissile to nonfissile materials is actually of the order of 20 sec, but the computation neglects this as though it were infinite.

As the computed $\int n\, dt$ increases and, consequently, fuel temperature rises, the coefficient B is readjusted to take account of the changes in fuel dimensions and Doppler coefficient with temperature. This procedure continues until the fuel at the center of the reactor reaches its melting point, after which expansion ceases, but Doppler effects continue. Finally, when fuel at the center of the reactor reaches its vaporization temperature ($\int n\, dt = 2.8 \times 10^6$ for core C), the method of computing reactivity variations is altered. From this point on, the plutonium vapor is permitted to behave as an ideal gas confined to the void fraction of the region in which vaporization has taken place. This region grows as vaporization continues. At the same time, the dimensions of the core are enlarged according to the acceleration of the core and blanket mass due to the gas pressure at the center. Core and blanket pieces are assumed to accelerate like free bodies initially at rest. Eventually this enlargement of the core reduces reactivity to the point where the reaction is shut down.

This simple approach neglects the details of some of the thermodynamic and shock effects of an explosion, but it provides a means of comparing the course of different excursions and permits an estimate of their relative severity. The final estimate of the total fissions involved in the burst must be considered semiquantitative at best.

As initial condition, the reactor is presumed to contain a density of one neutron/cc. The excess multiplication can be either zero or some finite amount. The excursion calculations have been based on a reactor similar to core C,[20] and containing 0.66 g Pu^{239}/cc, 5.7 g U^{238}/cc, plus nonfissile materials. The presence of the U^{238} was taken into consideration in computing the effective β of 0.004166. Core C has a critical volume of 258 liters and requires essentially all our inventory of plutonium.

Calculations were performed with various ramp rates of reactivity insertion and various initial values of k-excess. Doppler variations as T^{-1} and $T^{-1.5}$ were both investigated. The initial room-temperature value of the Doppler coefficient was taken to be either 5×10^{-7}, 5×10^{-6}, or $10^{-5}\ \Delta k/°C$ in various groups of problems. The first value corresponds to

Doppler effects that would be present in a core with considerable U^{238} present, although due to limitations in the problem capacity, the pessimistic assumption was made that the negative U^{238} Doppler coefficient declined just as rapidly as the positive plutonium Doppler decreased with increase in fuel temperature. The second value of Doppler coefficient, 5×10^{-6} $\Delta k/°C$, would represent the situation in a plutonium carbide spectrum, with U^{238} present to increase the effective β, but with no consideration given to the heating of the U^{238} as a Doppler shutdown mechanism. The problems with a Doppler coefficient of 10^{-5} $\Delta k/°C$ were merely introduced to provide another factor of two in the Doppler effect due to the possibility of having a lower spectrum.

Some of the results of these problems are summarized in Table I. In Table I the first six columns on the left give the problem number and input specifications. Following this are three columns of figures specifying the characteristics of the burst at the moment when the accumulated heat produced in the fuel is sufficient to raise its temperature 20°C. (Throughout each problem, the assumption is made that all heat produced in the fuel remains in the fuel.) Following this is information giving the situation when the fuel temperature reaches successively 100 and 400°C temperature rises. These three points give an indication of the time scale of the first phase of the excursion. The final two columns of Table I give the final total fissions when the pressure has disassembled the reactor and the maximum value of k-excess during the course of the excursion. This maximum of k-excess occurs just before the peak power is reached.

Examination of Table I shows without question that when large amounts of reactivity are added to the reactor at high rates, the resulting excursion can be very large if no mechanical shutdown device is invoked. Any combination of reactivity additions which total a dollar or more before vaporization begins leads to large bursts. This result was anticipated even with the uranium loadings for ZPR-III and is discussed in the summary of the ZPR-III hazards report.

Certain of these problems, however, shed some light on the suitability of the mechanical shutdown devices built into ZPR-III for terminating an autocatalytic (positive Doppler) excursion. In particular, examination of problem Nos. 15-18 is instructive. In problem 15, the reactor is presumed to have been put on a positive period corresponding to $\Delta k = 0.0006$ (asymptotic period would be about 50 sec) at a flux level of 2×10^{8} nv. This power level is high enough for the instruments to see the entire excursion. By the time the fuel temperature had risen 100°C, the period would be short enough to cause the scram devices to be actuated, and furthermore, there would be ample time (16 sec) for the instruments to operate before the fuel temperature reached disastrously high levels. (400°C is somewhat arbitrarily considered to be the upper limit of safe fuel temperature.) Problem 16 illustrates the course of a similar

Table I

EXCURSION CALCULATION SUMMARY

No.	β_{eff}	Initial k_{ex}	Ramp, k/sec	Doppler at Room Temp, $\Delta k/°C$	Doppler Variation	Rise of Fuel Temperature									Total No. of Fissions	Max $k_{ex} \times 10^3$
						20°C ($\int n\,dt = 2.5 \times 10^4$)			100°C ($\int n\,dt = 1.25 \times 10^5$)			400°C ($\int n\,dt = 5 \times 10^5$)				
						Time, sec	Power, w	Period, sec	Time, sec	Power, w	Period, sec	Time, sec	Power, w	Period, sec		
1	0.00417	0	0.02	5×10^{-7}	$T^{-1.5}$	0.22	1.0×10^9	4.6×10^{-4}	0.2207	6.4×10^9	4.4×10^{-4}	0.2213	20.8×10^9	5.7×10^{-4}	5.9×10^{18}	4.41
4	0.00417	0	0.002	5×10^{-6}	$T^{-1.5}$	2.1	0.43×10^9	7.2×10^{-4}	2.107	8.7×10^9	2.0×10^{-4}	2.1076	70.8×10^9	7.33×10^{-5}	1.1×10^{21}	6.64
5	0.00417	0	0.02	5×10^{-6}	$T^{-1.5}$	0.219	1.2×10^9	3.13×10^{-4}	0.2203	13.1×10^9	1.3×10^{-4}	0.2205	103×10^9	6.25×10^{-5}	1.4×10^{21}	6.8
9	0.00417	0	0.002	10^{-5}	$T^{-1.5}$	2.10	0.2×10^9	7.44×10^{-4}	2.104	12.8×10^9	9.99×10^{-5}	2.105	154×10^9	3.30×10^{-5}	9.3×10^{21}	9.28
10	0.00417	0	0.02	10^{-5}	$T^{-1.5}$	0.219	1.6×10^9	2.34×10^{-4}	0.220	18×10^9	8.28×10^{-5}	0.2202	176×10^9	3.26×10^{-5}	10.3×10^{21}	9.47
12	0.00417	0	0.002	5×10^{-6}	T^{-1}	2.106	0.44×10^9	6.74×10^{-4}	2.107	8.1×10^9	1.68×10^{-4}	2.1072	102×10^9	5.50×10^{-5}	5.1×10^{21}	8.34
13	0.00417	0	0.02	5×10^{-6}	T^{-1}	0.2198	1.2×10^9	3.03×10^{-4}	0.220	13.4×10^9	1.24×10^{-4}	0.2205	126×10^9	4.81×10^{-5}	5.8×10^{21}	8.53
15	0.00417	0.0006	0	5×10^{-6}	T^{-1}	271	1.24×10^4	33.2	321	1.6×10^5	12.4	337	2.2×10^6	9.39×10^{-1}	2.66×10^{19}	4.74
16	0.00417	0.0012	0	5×10^{-6}	T^{-1}	97.1	3.9×10^6	11.4	114.5	3.2×10^6	5.109	121	5.7×10^6	6.18×10^{-1}	1.57×10^{20}	5.38
17	0.00417	0.002	0	5×10^{-6}	T^{-1}	34.26	1.24×10^5	3.64	39.8	8×10^5	1.617	41.7	3.2×10^7	6.29×10^{-2}	5.87×10^{20}	6.12
18	0.00417	0.004	0	5×10^{-6}	T^{-1}	0.74	1.5×10^7	1.39×10^{-2}	0.75	4.1×10^9	2.4×10^{-4}	0.75	96×10^9	5.72×10^{-5}	4.26×10^{21}	8.09
21	0.00215	0	0.004	5×10^{-6}	$T^{-1.5}$	0.559	7×10^8	5.13×10^{-4}	0.56	9.2×10^9	1.7×10^{-4}	0.561	102×10^9	6.0×10^{-5}	1.26×10^{21}	4.7
22	0.00215	0	0.02	5×10^{-6}	$T^{-1.5}$	0.118	1.2×10^9	3.06×10^{-4}	0.119	13.6×10^9	1.3×10^{-4}	0.1195	141×10^9	5.6×10^{-5}	1.49×10^{21}	4.85
23	0.00215	0.0004	0	5×10^{-6}	$T^{-1.5}$	270.8	1.4×10^4	29.1	312.6	1.6×10^5	7.85	322.3	8.8×10^6	2.7×10^{-1}	5.66×10^{19}	2.88

excursion with twice this amount of reactivity, corresponding to a 14-sec period, and although this is twice the amount normally handled in experiments, there is still ample time (24 sec) for the functioning of instruments and scram devices between the onset of sensible heating (fuel-temperature rise of 20°C) and the upper limit of safe fuel temperatures. The time available for instrument operation is greater in problem 16 than in 15 because the period has decreased to the trip level when the fuel is at a lower temperature, i.e., with only a 20°C temperature rise. The same sequence occurs in problems 17 and 18. In problem 17, the initial reactivity is increased to about half a dollar, considerably more than is available in any planned experiment. Even under this extreme condition, there is more than 7 sec available for the instruments to operate within the period of fuel-temperature rise from 20 to 400(°C) above room temperature, and the instruments would, therefore, terminate the excursion. In problem 18 the initial reactivity is almost a dollar, the excursion proceeds very quickly, and the instruments would not function in time to prevent a serious excursion. In summary, with or without the autocatalytic Doppler effect, accidental reactivity additions of about a dollar will cause serious excursions, but at least half a dollar can be handled by the shutdown instruments.

This range can be narrowed somewhat. The maximum k_{ex} which could be introduced and still allow sufficient time (one second is more than enough) for the control rods to operate is about 0.0030, as shown by interpolation within the range covered by problems 15-18. If no Doppler effect were present, a k_{ex} of 0.0038 could be tolerated with the same time allowance. The Doppler effect thus reduces the excess reactivity which the machine can tolerate from 0.0038 to 0.0030 k_{ex} (90 to 70 cents). Introduction of neither of these amounts of excess reactivity is permissible under normal operating planning and procedures. Such amounts of excess reactivity could only be introduced through errors in planning and loading, and would lead to an excursion only if they were combined with instrumentation or interlock failures. The Doppler effect increases the probability of a disastrous excursion only to the extent that the errors leading to 0.0030 k_{ex} may be more probable than those leading to 0.0038 k_{ex}. Since the ZPR-III is well protected, by instruments, interlocks, and procedures, against either of these errors, it is not believed that the Doppler effect unduly increases the probability of an excursion.

Comparison of ramp rates of addition of reactivity with different assumed Doppler effects is possible by inspecting problems 1, 5, 10, and 13. In these, about five dollars per second reactivity was being added. This is actually about the maximum rate of reactivity which can be realistically assumed. It corresponds to the rate of addition associated with closure of the halves at high speed. In problem 1 the positive Doppler effect was small, of the order of the negative expansion coefficient. In problem 5 this is increased to a value corresponding to what might be expected with a carbide assembly with no contribution from U^{238}.

Problem 10 arbitrarily doubles this amount. Problem 13 approximates a Doppler variation with T^{-1} rather than $T^{-1.5}$. The time scale is very nearly the same in each case for the first stages of the reaction, that is, up to 400°C, about 220 ms in each case. Control rods or other devices might not act fast enough to shut down such a burst. On the other hand, if the source were in position (a startup requirement), and if the excursion started from subcritical (as on assembly of the halves), multiplication would be sufficiently high for the instruments to register an apparent period when the reactor was still at least 2% ($5) subcritical. This would provide an additional second for the safety rods to operate, sufficient time for them to take effect before criticality is actually reached.

Problems 4, 9, and 12 show that if the reactivity addition rate is reduced by a factor of 10, to 0.002 $\Delta k/sec$, the situation is not substantially changed. The time between the onset of sensible heating and the exceeding of safe temperature limits is just a few milliseconds, not enough for safety rods to operate. In these cases, however, more than 2 sec elapse between criticality and sensible heating. This is enough time for the control rods to operate. Note that the rate of insertion of reactivity with the intermediate-speed motor is 0.004 $\Delta k/sec$, and thus lies between the cases discussed in this and the previous paragraph.

Thus on all carriage speeds, the instruments would function in time to shut down an excursion.

Finally, problems 21, 22, and 23 were calculated with the reduced delayed-neutron fractions characteristic of Pu^{239}, without any contribution from U^{238}. The value of β_{eff} was taken as 0.00215. The time scale for these excursions is quite similar to that of the other excursions if all reactivity insertions are reduced to a dollar basis. The values of λ_i and β_i in all of the above calculations are taken from Keepin.(16)

Very little can be expected in the way of avoiding a disastrous excursion if after the safety rods are ejected the carriage in a non-normal fashion continues to move together, adding more reactivity than the safety rods have been able to remove. The possibility of achieving criticality before the halves are together is therefore kept to an absolute minimum by conservative planning of each loading and strict administrative control on checking and inspection procedures. Since the safety rods normally contain at least 2% reactivity, the loading error would have to be larger than this amount at the same time the halves were driven together in order to cause this uncontrolled excursion.

B. Maximum Credible Accident

Courses of events which might lead to an excursion have been examined many times, by people within and outside the ZPR-III staff. Some

of the excursions which have been proposed for examination do not lend themselves readily to any quantitative estimate of probability, but all possible measures have been taken to keep these probabilities low. Among such excursions are those due to:

1. Sabotage

   Personnel screening procedures, the system of loading checks by independent physicists, the use of key control for the reactor requiring two responsible people for operation, and the detailed records kept of the reactor loadings, all help to reduce the possibility of sabotage.(1)

2. Natural Catastrophes, Such as Earthquakes and Floods

   These contingencies are taken into consideration and guarded against in the design of the building.(1)

3. Collapse of the Matrix

   This is an unrealistic proposal, since the matrix tubes are conservatively stressed. Calculations indicate that if the lower 40% of the assembly collapsed, including 20% of the core, and filled the 3% of vertical clearance in that region, the total gain in reactivity for core C would be only about half a dollar.

Reliability of equipment and supervisory procedures designed to avoid nuclear excursions during operation can be examined in more detail, against the background of the long history of reliable operation of the ZPR-III. Concerning operational sequences which are not normal but have been proposed as potentially leading to an excursion, the following cases deserve consideration:

1. Failure of Nuclear Instrumentation

   To lead to an excursion, a failure of nuclear instrumentation would have to involve all five independent instrument channels which are connected to scram circuits. These channels are truly independent of each other, each with its own detector, preamp, amplifier, power supply, monitor circuits, and scram relays. Three of these channels trip the scram circuit on high power level and two trip on short period. Operating and inspection procedures have been well established, and the operating reliability of these instruments has been very well demonstrated. The instruments are fail-safe in the sense that loss of power to the instrument circuits scrams the reactor.

Simultaneous failure of all five channels in a non-safe fashion, so that an excursion would not be detected and terminated in an early phase, is not considered credible.

2. A Startup Accident Resulting from Mechanical Failures

Although many different hypothetical sequences have been examined, no operating situation has ever been conceived in which less than three independent and highly improbable events were required to precede the excursion. For example, it has often been proposed that the machine might be loaded in such a way that criticality might be reached before the carriage was fully closed, that the carriage might continue to drive together in spite of this, and that excess reactivity so added could result in an excursion. Although there are many variations to this accident, the following three events are prerequisites for such an excursion:

a. An error in planning or loading must occur. Normal practice is to plan each loading so that criticality will be reached by insertion of one or two control rods after the carriage is closed. The error must be large enough to make the reactor critical before closure; otherwise the operation proceeds normally.

During the seven years in which the ZPR-III has been safely operated, including over 5000 different loadings, there have been only two occasions when the plans for a reactor loading were sufficiently in error to cause the reactor to go critical before closure. Since these two planning errors were not coincident with the other extremely unlikely events required to cause an excursion, no serious consequences resulted. The plans for these loading changes are always based insofar as possible on previous experimental information. The first error was due to extrapolation of the reactivity effect due to fuel bunching (a linear extrapolation of the results of one- and two-fuel-plate bunchings was not applicable to three-plate bunchings). Criticality was reached with the halves separated by about 0.12 in., and a normal scram was set off from a period signal trip. After this incident, an interlock was added which stops closure of the carriage when the multiplication is rising on an apparent 30-sec or shorter period. Several years later, when the second error occurred, this device disclosed the error well before criticality was achieved and scram circuits were not even required. In this second case, criticality was reached

with the halves separated by about 0.18 in. The error was due to the misinterpretation of a previous experiment measuring the worth of the addition of core material at the edge. The change was wrongly attributed to a volume equal to twice its actual volume. In extrapolation to the next experiment, the volume chosen was then too large for the worth of the control rods.

No errors in loading performance of any significant magnitude have ever been observed. This is attributed to the fact that the loading of each drawer is carefully checked piece by piece, and that no single piece incorporates a substantial amount of reactivity. The probability of significant loading errors is therefore very low. A loading error large enough to make one half critical by itself is inconceivable because of the large amounts of material required and independent checks by supervisors. Even a loading error large enough to make the reactor critical on its intermediate or high-speed ranges is practically inconceivable, as this would require from 20 to 50% excess mass, complete disregard for stepwise approach to criticality, and complete breakdown of administrative checking procedures.

b. The carriage must fail to stop when a 30-sec period is reached and must fail to reverse itself when a 15-sec scram signal is reached. In normal operation these functions are automatic and controlled by independent relays. The relay-controlled logic curcuits are subject to frequent operating tests and have been fully reliable over the years. The carriage has never failed to reverse itself during its forward travel when called upon to do so; this has been verified over 100 times during seven years.

c. The safety rods must fail to shut the reactor down. This might occur if a large number of rods failed to operate. However, in over 700 operational checks of the safety rods, performed at least twice each week, no single rod has ever failed to scram. This has been checked at normal and reduced air pressure, under simulated conditions of air failure, and with a great variety of rod loadings. An error large enough to override the safety rods, generally worth at least 2% Δk, has never occurred.

Furthermore, the excursion kinetics calculations of the previous section show that if the safety rods operate in their normal fashion, they are fast enough to handle any ramp rate of reactivity up to that of the high-speed carriage movement.

The preceding three areas of safeguards therefore all have an exceedingly low probability of breakdown. They are all quite independent and their simultaneous failure is required to produce an excursion. In addition, a fourth should be cited: The operating crew must be inattentive to the instrumentation for a considerable period prior to the excursion, since with any degree of attentiveness, manual action to shut the reactor down can be taken.

The above considerations apply to a wide variety of startup accidents involving many different possible circuit failures, or different loading errors. Analysis of these required sequences of events, as discussed above, has in every case resulted in the conclusion that the probability of such a startup accident is incredibly low.

In general, considerable precautions have been taken against all conceivable methods of adding sufficient reactivity to produce a nuclear excursion. The considerable body of safe operating experience obtained with uranium loadings and the fact that the plutonium operations are not basically different provide justification for considering uncontrollable nuclear excursion to be beyond the area of credibility.

On the other hand, there are potential hazards associated with the plutonium operations with which we have little previous experience. These hazards are associated with the rupture of the canning materials and with plutonium fires. Previous experience with the particular materials involved is insufficient to predict the probability of these accidents.

It has been an infrequent but occasional experience in the uranium operations to drop a piece of the uranium to the floor from heights of several feet. Occasionally full drawers have been accidentally dropped. Plutonium operations are designed so that fuel handling is at a minimum, and extreme caution is emphasized, but it is still possible that pieces or even drawers may be dropped. Plutonium cans are designed to resist a moderate impact, but conceivably they could be ruptured by such a fall, permitting the exposure of plutonium to air with possible oxidation and dispersion. The drawers are transferred from the room where they are loaded to the reactor by means of carts. These carts may contain two drawers with a total of 2.6 kg plutonium. In the unlikely event that a cart should be upset and spill its contents on the floor, as much as 2.6 kg plutonium might become exposed to the air as a result of ruptured cans. Although, as indicated in the section on pyrophoricity, plutonium pieces of the size used here are not expected to burn easily at room temperature, nevertheless, imperfections and finely divided particles can initiate a fire which could conceivably involve the whole 2.6 kg. Because of physical separation from the reactor, it is not believed that such a fire could

spread throughout the assembly. Consistent with the experience with plutonium and uranium fires, the fraction of the plutonium which would become air-borne can be expected to be about 10^{-3}, or 2.6 g. This amount can be collected on the air filters without difficulty unless the filters should become plugged by other smokes. If the filters should become plugged, the 2.6 g of the plutonium could escape through leaks in the ZPR-III building, presenting some contamination problem to the surrounding area.

A fire in the assembly might be conceived as starting from a leaky can which caught fire spontaneously while in the reactor. There is ample evidence to indicate that a fire would not propagate throughout the entire inventory of plutonium and uranium, but here again experience with a close counterpart of the ZPR-III situation is lacking. With sodium present in the core, as it may be for some compositions, it is hard to rule out the possibility that a fire initiated by damage to a single fuel piece might propagate throughout the assembly. In the worst possible case, in which the entire inventory of plutonium might burn with associated quantities of sodium and uranium, intense temperatures would be developed in the assembly room, door gaskets and sealing devices would probably be destroyed, air filters could be damaged, and air-borne plutonium oxide might escape from the building. Based again on experience with fires, the amount of plutonium escaping from the immediate vicinity would be less than 10^{-3} of the 210-kg inventory, or about 200 g.

Accidents of this nature, although contrary to expectations based on laboratory tests, are not the subject of a large background of experience to aid in judging their credibility.

It is therefore believed reasonable to assume that a maximum credible accident might consist of a fire in which a large part of the inventory of plutonium was oxidized, damage to the sealing of the building occurred, and plutonium in a quantity of about 200 g was released from the building.

C. Contamination Levels

Dispersal of 200 g of plutonium following the maximum credible accident has been estimated in Tables II and III for different meteorological conditions. The release of 200 g of plutonium of the composition available to ZPR-III would include release of 11.5 c Pu^{239} and 2.3 c Pu^{240}, a total of about 14 c of plutonium alpha activity. The calculation was made for the cases in which the plutonium was released from the fire at a height of 2 or 25 m. Dispersion was then computed by the Sutton equation with no attenuation due to fallout. Cases are considered in which the plutonium may be either in a soluble or insoluble form, retention in the critical organ differing by about a factor of 1.5 in the two cases.

Table II

MICROCURIES OF PLUTONIUM WHICH COULD BE ACCUMULATED IN CRITICAL ORGANS BY INHALATION UNDER DAYTIME CONDITIONS

Values are based on the Sutton diffusion equation from a point source with no attenuation due to fallout, for a total release of 14 c (200 g). Numbers in parentheses are the powers of ten by which the values are to be multiplied.

Material	$n = 0.2$, $h = 2$, $\bar{u} = 16$, $C = 0.3$		$n = 0.2$, $h = 25$, $\bar{u} = 16$, $C = 0.289$	
	Soluble Pu	Insoluble PuO	Soluble Pu	Insoluble PuO
Critical Organ	Bone	Lungs	Bone	Lungs
Max permissible amount in critical organ, μc	3(-2)	2(-2)	3(-2)	2(-2)
Distance Downwind, m				
560	6.5(-3)	4.3(-3)	6.5(-3)	4.3(-3)
1000	2.3(-3)	1.5(-3)	2.4(-3)	1.6(-3)
1800	8.0(-4)	5.3(-4)	8.5(-4)	5.7(-4)
3200	2.8(-4)	1.9(-4)	3.0(-4)	2.0(-4)
5600	1.0(-4)	6.9(-5)	1.1(-4)	7.4(-5)
10000	3.6(-5)	2.4(-5)	3.9(-5)	2.6(-5)
18000	1.3(-5)	8.5(-6)	1.4(-5)	9.1(-6)
32000	4.5(-6)	3.0(-6)	4.8(-6)	3.2(-6)

Table III

MICROCURIES OF PLUTONIUM WHICH COULD BE ACCUMULATED IN CRITICAL ORGANS BY INHALATION UNDER NOCTURNAL CONDITIONS

Values are based on the Sutton diffusion equation from a point source with no attenuation due to fallout, for a total release of 14 c (200 g). Numbers in parentheses are the powers of ten by which the values are to be multiplied.

Material	$n = 0.5$, $h = 2$, $\bar{u} = 6$, $C = 0.035$		$n = 0.5$, $h = 25$, $\bar{u} = 16$, $C = 0.029$	
	Soluble Pu	Insoluble PuO	Soluble Pu	Insoluble PuO
Critical Organ	Bone	Lungs	Bone	Lungs
Max permissible amount in critical organ, μc	3(-2)	2(-2)	3(-2)	2(-2)
Distance Downwind, m				
560	6.7	4.4		
1000	3.2	2.1	3.2(-10)	2.1(-10)
1800	1.4	9.4(-1)	1.0(-4)	6.7(-5)
3200	6.1(-1)	4.1(-1)	1.6(-2)	1.0(-2)
5600	2.7(-1)	1.8(-1)	6.7(-2)	4.4(-2)
10000	1.1(-1)	7.5(-2)	7.8(-2)	5.2(-2)
18000	4.7(-2)	3.1(-2)	6.6(-2)	4.4(-2)
32000	2.0(-2)	1.3(-2)	2.5(-2)	1.7(-2)
56000	8.5(-3)	5.7(-3)	1.2(-2)	7.8(-3)
100000	3.3(-3)	2.4(-3)	5.1(-3)	3.4(-3)

To compute the time-integrated exposure at the ground point downwind from a burst under these conditions, the Sutton equation takes the form(23)

$$TID = \frac{2Q}{\pi C^2 \bar{u} (\bar{u}t)^{2-n}} \exp \frac{-h^2}{C^2 (\bar{u}t)^{2-n}} .$$

The values of the parameters C, n, and $\bar{u}$ were extrapolated or estimated from meteorological data compiled at the NRTS by DeMarrais and Islitzer.(24) These data show that values of 0.2 and 0.5 for n, the Sutton stability parameter, represent frequently occurring lapse and inversion conditions at the NRTS. The data also indicate that wind velocities, $\bar{u}$, of 6 mph under inversion conditions and of 16 mph under lapse conditions represent some of the more frequent combinations. Although the wind data were obtained at the 20-ft level, these wind velocities are expected to be quite common also at the 2- and 25-m levels where the releases were calculated. The 25-m value of C used here is taken from the value given by DeMarrais and Islitzer, and since no lower values are available, the 2-m value is extrapolated slightly from this. (C is the generalized diffusion coefficient for isotropic turbulence, in meters$^{n/2}$.)

The values of TID have been multiplied by an average breathing rate, 2.32×10^{-4} m^3/sec, and by a retention factor to obtain the values in Tables II and III. The retention factor is taken as 0.18 of the amount inhaled for soluble plutonium in the bones, and as 0.12 for insoluble material in the lungs.

Although the calculations in Tables II and III do not cover every possible situation, they illustrate the magnitude of the dispersal problem. Under the conditions chosen for illustration, personnel in the Central Facilities Area and other installations within 23 miles of the 200-g release might have to be protected during the passage of a low-level cloud under nocturnal conditions. Forty minutes would be available to carry out this protection, with the low windspeed chosen. With higher windspeeds, the shorter time for evacuation would be offset by the increased turbulent dispersion. Further parametric effects of various cloud heights, wind speeds, and temperature profiles are illustrated in the more comprehensive tables compiled for the EBR-II project.(25) The neglect of fallout makes all calculated doses pessimistic by factors of perhaps 10 or more.

The effect of fallout would be to reduce the inhalation doses from the cloud, but to transfer contamination to the ground. Surface contamination levels of 100 and up to 1,000 μg/m^2 of plutonium are considered permissible under various circumstances. Thus, in the worst dispersal pattern, 200 g of plutonium could contaminate 2×10^6 m^2 (500 acres) to the 100-μg/m^2 level, or 50 acres to the 1,000-μg/m^2 level.

Contamination of water supplies has been estimated by computing the maximum activity per square meter which would precipitate out from the cloud due to a sudden shower, dropping essentially all of the cloud at any of various distances. The activity which could be deposited per unit surface area is obtained from an integration of the Sutton equation in the vertical direction, which results in the expression

$$\omega = \frac{Q}{\pi C^2 (\bar{u}t)^{2-n}} ,$$

where ω is the activity per square meter. This activity is then assumed to be diluted by a 3-m depth of water as in an open reservoir to obtain the water concentrations listed in Table IV. The calculation is done for the meteorological conditions of weak lapse, which is a common condition during precipitation periods at the NRTS,[24] and for an average wind speed of 10 mph. The results indicate that contamination of water supplies would be within acceptable tolerances at distances greater than 7 miles. At distances closer than this there are no exposed water supplies, and any water contamination would be subject to considerable dilution by the underlying ground water of the site.

Table IV

GROUND WATER-CONTAMINATION LEVELS

Levels resulting from the release of 200 g (14 c) of plutonium followed by total washout from the cloud at various distances.

$n = 0.25$ $C = 0.141$ $\bar{u} = 10$ $h = 25$	
D, m	Water Concentration, $\mu c/m^3$
560	1140
1000	413
1800	148
3200	54
5600	20
10000	7
18000	2.6
32000	0.9
56000	0.4
100000	0.1
Permissible level	6

APPENDIX A

Explosion Calculations

Program RE-129J is a program in which reactivity is initially inserted at a ramp rate, and the negative effect of fuel expansion is allowed for, as well as the positive Doppler effect, starting with production of sensible heat and continuing until fuel vaporizes. Sufficient pressure from vaporized plutonium must be developed to move the surrounding mass of material and expand the core, reducing the reactivity and terminating the excursion.

Thus, in the first phase of this problem,

$$\frac{dk_{ex}}{dt} = A + B\,n \quad , \quad 0 < t < t_0 \quad ,$$

where

A = ramp rate of reactivity addition,

B = a temperature-dependent coefficient, which may be a function of the integrated power up to any moment during the course of excursion, and

n = neutron density.

Here t_0 is determined so that $\int_0^{t_0} n\,dt$ provides sufficient energy to vaporize the plutonium.

For $t > t_0$, the second phase of the problem, a shutdown coefficient due to expansion of the core by gas pressure is added:

$$\left(\frac{dk}{dt}\right)_{\text{shutdown}} = -\frac{1}{r_0\,M_L}\int_{t_0}^{t} P\,dt \quad ,$$

where

r_0 = initial radius of the core, ft,

M_L = mass loading on expanding gas, and

P = pressure of expanding gas, psi.

The core under consideration contained 23% voids.

To estimate the pressure of vaporized plutonium, the gas, considered ideal, was allowed to fill a void volume equal to 23% of the core volume formerly associated with the now-vaporized plutonium. The subsequent course of the excursion was determined by the addition of energy and the expansion of the core and blanket, as illustrated in the following description.

In ZPR-III, for fast excursions, heat transfer to inert materials would have a time constant of about 20 sec and is neglected. The problems were based on a core in which 4.41% of the core volume was occupied by plutonium, with a density of 15 g/cc, and the core had a reactivity coefficient of -0.5×10^{-6} $\Delta k/k$-°C due to axial thermal expansion of the separate fuel pieces. The coefficient B was broken into two parts, that due to expansion and that due to Doppler effect.

$$B_{expansion} = -\frac{0.239 \times 0.5 \times 10^{-6}}{3 \times 10^{-7} \times 3 \times 10^{10} \times 0.0441 \times 15 \times 0.043}$$

$$= -9 \times 10^{-4} \times 0.5 \times 10^{-6}$$

$$\approx -4.5 \times 10^{-10} \frac{\Delta k}{\text{k-sec-n/cc}} \quad ,$$

where

3.0 = neutrons/fission,

10^{-7} sec = prompt-neutron lifetime,

3×10^{10} = fissions/w-sec,

0.239 = cal/w-sec,

0.0441 = volume fraction plutonium,

15 g/cc = density of plutonium,

0.043 cal/g-°C = specific heat of plutonium.

Thus the factor 9×10^{-4} converts $(°C)^{-1}$ to $(\text{sec-n/cc})^{-1}$. This same factor is used to convert the Doppler coefficient to a value for $B_{Doppler}$. The Doppler coefficient is calculated from the central spectrum of the core. Its variation with temperature is approximated by a series of straight-line segments. Each straight-line segment is expressed in the form

$$B_{Doppler} = B_1 + B_2 \int n \, dt \quad .$$

These substitutions give the equation of reactivity rate the following form:

$$\frac{dk_{ex}}{dt} = A + n\,(B_{expansion} + B_1 + B_2 \int n\,dt) \quad .$$

With a melting point of 650°C and a boiling point of 3500°C, with 40 cal/g to reach melting, 110 cal/g to reach vaporization, and 340 cal/g to vaporize, approximately 500 cal are required to vaporize one gram of fuel.

Fuel melting begins when $\int n\,dt = 10^6$, and after this time $B_{expansion}$ is zero.

We assume a roughly parabolic distribution of power from center to edge of the core. We note that the $\int_0^t n\,dt$ must reach a value of 3.9×10^6 to provide the 150 cal/g necessary to initiate vaporization at the core center. It can then be shown that the radius r of the region in which vaporization of plutonium has begun is given by

$$r^2 = 22\left[1 - \frac{3.9 \times 10^6}{1.4\int_0^t n\,dt}\right] \quad .$$

After vaporization has begun, the method of solving the kinetics changes, so that in this second phase of the problem

$$\frac{dk_{ex}}{dt} = A - \left(\frac{dk}{dt}\right)_{shutdown} \quad ,$$

in which the term on the right is the rate at which reactivity is reduced due to the pressure of the gas forcing the core and blanket apart.

The combined effects are solved by an iterative procedure which permits some vaporization to occur, followed by a slight expansion which reduces dk_{ex}/dt. Iterations of vaporization and expansion at the corresponding values of dk/dt follow the course of the excursion until $\int n\,dt$ no longer increases appreciably. The rate of change of r, due to gas pressure, may be approximated by

$$\frac{dr}{dt} = 12 \int_{t_0}^{t} \frac{P_c}{M_L}\,dt, \text{ in./sec} \quad ,$$

where P_c is the pressure of the vaporized plutonium confined in the region of the core voids within which it vaporized, calculated as an ideal gas, and

then corrected for expansion as the unvaporized parts of the core and blanket are accelerated outward. Without correction, the pressure at the center at time t is

$$P = 8.7 \times 10^{-3} \int_{t_0}^{t} n \, dt \quad ,$$

and

$$P_c = [G] \, P \quad ,$$

where the bracket [G] can be represented in the iterative fashion in which the problem is solved by

$$[G] = \frac{P(t+\Delta t)}{P(t)} = \left[1 - \frac{3 \left[r(t+\Delta t) - r(t) \right]}{\sqrt{22 \left(1 - \frac{3.9 \times 10^{6}}{1.4 \int_{0}^{t} n \, dt} \right)}} \right] .$$

M_L is the mass loading on the gas, that is, the mass of core and blanket divided by the area of the region enclosing the gas. Thus,

$$M_L = \frac{1323}{4\pi r^2} = \frac{105}{r^2} \quad .$$

The shutdown dk/dt is related to the expansion of r by

$$\left(\frac{dk}{dt} \right)_{\text{shutdown}} = \frac{1}{r_0} \frac{dr}{dt} \quad ,$$

where r_0 is the core radius, 15.5 in.

APPENDIX B

Some Properties of Plutonium-Aluminum Alloys

The properties listed below were obtained from communications from R. J. Dunworth, F. L. Yaggee, and F. G. Foote. These values are in substantial agreement with published values in the ANL Metallurgy Division Annual Reports for 1960 (ANL-6330, pp. 55-58) and 1961 (ANL-6516, pp. 69-74). Complete information on these alloys will appear in ANL-6639, Some Mechanical and Physical Properties of Plutonium-rich Plutonium-Aluminum Alloys, by F. L. Yaggee and C. M. Walter (in preparation).

Table B-I

DENSITIES AND EXPANSION COEFFICIENTS FOR Pu-Al ALLOYS

Specimen No.	Composition (w/o)	Density (g/cc) Homogenized	Mean Linear Exp Coeff 10^{-6}/°C (20-400°C) Homogenized
209	0.26	15.71	35(1)
210	0.42	15.62	3.2
207	0.78	15.42	7.3
R-11 TC-2	0.98	15.07	10.6
208	1.22	15.13	12.8
211	1.23	15.08	13.0
ET-1	1.25	15.06	13.7
206	3.06	13.73	18.1

(1) The large mean coefficient was caused by alpha, beta, and gamma phases in the casting.

Table B-II

THERMAL CONDUCTIVITY OF Pu-1 w/o Al ALLOY

Temp (°C)	k (cal/sec-cm-°C)
100	0.022
200	0.027
300	0.033
400	0.038
500	0.044

Hardness

The hardness of a plutonium-1.1 w/o aluminum alloy was measured as 51 R_F (approximately 61 VHN 25 g).

Phases

According to the phase diagram, Fig. B-1, the face-centered cubic delta phase is stable to room temperature for alloys between 0.28 and 1.37 w/o aluminum.

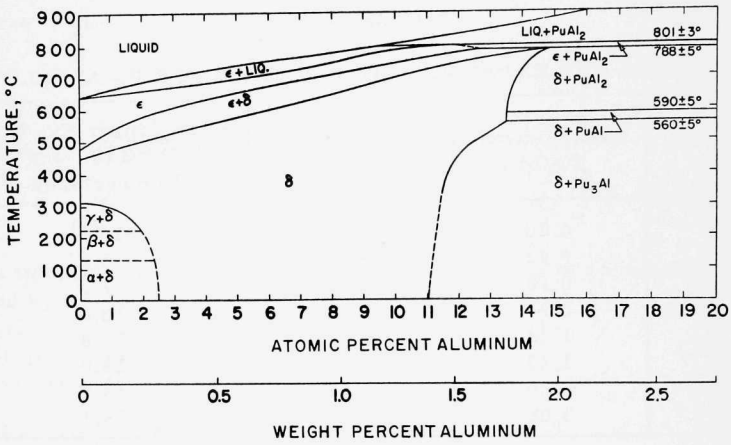

Fig. B-1. Phase Diagram of Plutonium-Aluminum System

1. 1/4 atom percent of Al completely suppresses the delta prime phase (Ref: Ellinger, Los Alamos).
2. At 10% Al the phase $PuAl_3$ cannot be removed (equilibrated) even after heat treatment for extended periods below 350°C (Ref: Ellinger, Los Alamos).
3. Diagram based on information obtained on June 20, 1960 from F. H. Ellinger of Los Alamos Scientific Laboratory.

A plutonium-1.25 w/o aluminum alloy will transform as follows:

from delta to epsilon plus delta at 700°C;
from epsilon plus delta to epsilon at 745°C;
from epsilon to epsilon plus liquid at 785°C;
from epsilon plus liquid to liquid at 805°C.

We might expect $PuAl_2$ and $PuAl_3$ in castings of this alloy (1.25 w/o). Both of these compounds should be removed by annealing at 550°C.

Thermal Cycling

A number of pins were thermally cycled. No growth was found in the alloy after 300 cycles between 50 and 350°C.

Extrusion

The 1.2 w/o aluminum alloy extrudes easily at 325°C.

Rolling

The 1 w/o to 1.2 w/o alloys are cold rolled without difficulty to at least a 90% reduction in area.

Low-melting Plutonium Eutectics

A more restrictive limitation on the high-temperature use of plutonium is its formation of low-melting eutectics with iron, cobalt, nickel, and manganese. The low-temperature iron eutectic contains about 9.5 w/o iron and melts at 412°C.

High-temperature Compressive Strength

The high-temperature strength of the plutonium-1 w/o aluminum alloy was determined in the temperature range 400-600°C. Rod specimens, 0.205 in. in diameter by 1.5 in. long, in the as-cast condition were heated to 400, 500, and 600°C in a consecutive manner and held at each temperature for 20 hr. The decrease in length with time at temperature under several different compressive specimen loads was measured. Approximately 80 percent of the total change in specimen length listed below occurred in the first few hours at temperature.

Table B-III

HIGH-TEMPERATURE STRENGTH
OF A Pu-1 w/o Al ALLOY

Compressive Specimen Load (psi)	Change in Length in 20 hr, $-\Delta L$ (%)		
	400°C	500°C	600°C
7.2	0.75	0.30	2
20.3	0.50	0.70	1
34.6	0.25	1	2
46.8	0.25	1.75	5.25
59.4	0.25	9.25	5.50(1)

(1) Specimen filled out 0.010-in. annulus and was subsequently restrained by

The plutonium-1.25 w/o aluminum alloy exhibits a somewhat higher compressive strength at these temperatures in the as-cast condition. This strength can be significantly improved by a prior vacuum heat treatment of 3 hr at 550°C, as indicated in Table B-IV for a specimen load of 20.3 psi.

Table B-IV

HIGH-TEMPERATURE COMPRESSIVE STRENGTH
OF A Pu-1.25 w/o Al ALLOY

Metal Condition	Test Temp (°C)	Time at Temp (hr)	Change in Length, $-\Delta L$ (%)
As-cast	400	3.7	0.07
	425	1.2	0.08
	450	7.0	0.17
Heat Treated	400	3.75	0.01
	450	2.5	0.01
	500	5.5	0.07
	550	1.8	0.25
	600	1.3	2.7

APPENDIX C

Relation between Reactivity and Core Radius in a Reflected Reactor

by

A. R. Baker*

February 2, 1961

1. Introduction

During the writing of a recent memorandum, ARB-19, there was some discussion about why the relationship between reactivity and core radius should be so different for Assemblies 25 and 29, both large reactors. The following approximate one-group theory was worked out to try to find out why.

2. Theory

For a spherical system, the reactivity ρ is related to the change in core radius, ΔR, from the critical radius R by the following formula:

$$\rho = \frac{3}{f}\left(\frac{\Delta R}{R}\right) \quad .$$

The reactivity ρ is related to the mass increase at the edge by the formula

$$\rho = \frac{1}{f}\left(\frac{\Delta M}{M}\right) \quad .$$

A one-group formula for f will now be derived.

According to one-group theory, the criticality condition is

$$\frac{(\nu - 1)\Sigma_f - \Sigma_c}{D} = \left(\frac{\pi}{R + \delta}\right)^2 \quad ,$$

where

ν is the number of neutrons per fission for core material,

Σ_f is the fission cross section for core material,

Σ_c is the capture cross section for core material,

D is the diffusion constant for core material,

δ is the reflector savings.

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*Attached to ANL from UKAEA, A.E.W. Winfrith, Dorset, England.

Now consider the following hypothetical situation. The core radius is increased by ΔR and ν is increased by $\Delta\nu$ to maintain criticality. Then,

$$\frac{\Sigma_f \Delta\nu}{D} = \frac{2\pi^2 \Delta R}{(R+\delta)^3} \quad .$$

It has been assumed that δ is independent of ν and R which can be seen to be true approximately from Glasstone and Edlund, Section 8.33.(1) In the real situation, the change in radius leads to reactivity given by

$$\rho = -\left(\frac{\Delta\nu}{\nu}\right) = \frac{2\pi^2 D \Delta R}{\nu\Sigma_f (R+\delta)^3} \quad .$$

Hence,

$$f = \frac{3}{\rho}\left(\frac{\Delta R}{R}\right) = \frac{(R+\delta)^3}{2\pi^2 R}\frac{3\nu\Sigma_f}{D} \quad .$$

Similar formulas can be derived for cylindrical systems. Through use of the criticality condition, the following alternative form may be derived:

$$f = \frac{3(R+\delta)}{2R}\frac{\nu\Sigma_f}{(\nu-1)\Sigma_f - \Sigma_c} \quad .$$

This formula is not so reliable as the first form for quantitative calculations because of the difficulty of estimating the contribution of inelastic scattering to Σ_c.

3. Discussion

The first form of the formula has been evaluated for the ZPR-III Assemblies 6F, 9A, 22, 24, and 25 with the parameters calculated by D. Meneghetti and also for calculated reactors A-H.(2) The first five of these are typical of plutonium-fueled power reactors; the last three are similar to ZPR-III Assembly 29, but with three different enrichments.

Since Meneghetti did not calculate the reflector savings δ for Assemblies 6F, 9A, 22, 24, and 25, values were estimated from the calculated values for reactors F, G, and H which have blankets of the same composition. It can be seen from Glasstone and Edlund, Section 8.33,(1) that δ is proportional to D for a given blanket composition. For reactors F, G, and H, the values of δ/D were 10.3, 9.9, and 9.5, respectively. An average value, 9.9, was assumed when estimating the values of δ for Assemblies 6F, 9A, 22, 24, and 25. The results are given in Fig. C-1 and Table C-I. Experimental values and values estimated from pairs of SNG calculations of slightly different radii(2) are also included in Table C-I.

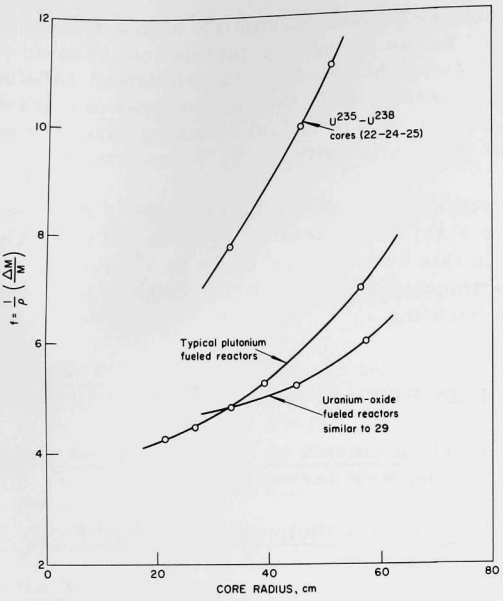

Fig. C-1
f-values

Table C-I

CALCULATED AND EXPERIMENTAL f-VALUES

Reactor		f		
		Expt.	SNG (Meneghetti)	One-group
Plutonium Fuel	A			49.8
	B			7.0
	C			5.2
	D			4.5
	E			4.2
Oxide Fuel	F			6.0
	G			5.2
	H			4.8
	29	5.4		5.2(1)
U^{235}-U^{238} Assemblies	6F	7.5		6.8
	9A	4.8	5.3	6.3
	22	6.1	7.1	7.7
	24	9.3		9.9
	25	8.0	11.4	11.0

(1)Interpolated

It can be seen from Table C-I that Meneghetti's high estimate of f for Assembly 25 is confirmed by the one-group calculation. The accuracy of the experimental values might be expected to be about 10%, but the discrepancies between experimental and calculated values are greater than this. These discrepancies may be due to fuel bunching; they are not due to the small departures of L/D from unity.

It is evident from the second form of the formula for f that the high value, for a reactor of its size, for Assembly 25 is due to the high proportion of absorber, U^{238}, in this core. When there is a larger proportion of absorber, a larger fractional change in ν is required to balance a given fractional change in the buckling.

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