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FISSION PRODUCT RELEASE AND TRANSPORT IN LIQUID METAL FAST BREEDER REACTORS

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Nuclear Safety Information Center

FISSION PRODUCT RELEASE AND TRANSPORT IN LIQUID METAL

FAST BREEDER REACTORS

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FOREWORD

This report was initiated at the request of the Liquid Metal Fast Breeder Reactor Program Office at Argonne National Laboratory to provide a state-of-the-art review on fission product behavior in liquid metal systems.

The quantity of safety data on LMFBFR's accumulated through research and development and operating experience is relatively small when compared to the tremendous body of information on water-cooled reactors. LMFBFR's are now going through a stage of development similar to that experienced by water reactors over a decade ago.

This report should serve as a foundation from which to plan research needed to fully understand fission product behavior in liquid metal systems. The information contained herein has already been incorporated into the "Liquid Metal Fast Breeder Reactor Program Plan, Vol. 10, Safety", USAEC Report WASH-1110, August 1968.

Wm. B. Cottrell, Director
Nuclear Safety Information Center

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PREFACE

The Nuclear Safety Information Center was established in March 1963 at the Oak Ridge National Laboratory under the sponsorship of the U.S. Atomic Energy Commission to serve as a focal point for the collection, storage, evaluation, and dissemination of nuclear safety information. A system of keywords is used to index the information cataloged by the Center. The title, author, installation, abstract, and keywords for each document reviewed is recorded on magnetic tape at the central computer facility in Oak Ridge. The references are cataloged according to the following categories:

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ABSTRACT

Liquid-metal fast breeder reactors will build up large fission-product inventories. Volatile fission products may be vented from the fuel elements to the primary sodium coolant. In operation and after minor meltdowns, the coolant system will retain most of the iodine and other fission products from vented or ruptured fuel elements; some, particularly noble gases, will enter the argon blanket gas. If the coolant system and the nitrogen-filled containment system vessel enclosing it were breached, sodium containing ^{24}Na , fission products, and possibly UO_2 and PuO_2 would enter the air within the secondary containment system. The sodium would burn, and sodium oxide aerosols might sorb or occlude large fractions of most radioactive materials, but not of the noble gases. High-efficiency particulate air filter systems for water-cooled reactors may be readily adaptable for the removal of sodium oxide aerosols by the addition of extensive prefiltering systems. Methyl iodide may not be a problem in these reactors; the major hazards will probably be ^{24}Na , plutonium, fine aerosols, and noble gases.

SUMMARY

Fission product release and transport in a liquid metal fast breeder reactor depends on many parameters, including the physical and chemical nature and half-life of the fission product, the design of the reactor (particularly the fuel elements and the coolant-blanket gas system), and whether a given phase of release or transport occurs during normal reactor operation or as a result of an accident.

During normal reactor operation much of the volatile fission products and some of the solid fission products diffuse through the fuel and are collected in the fuel element, which may have a sealed or vented expansion volume. Fission products may enter the coolant by diffusion through the fuel element cladding or through cracks resulting from cladding failure. Long-lived volatile fission products will enter through vents, if vented. Fission products released into the coolant may deposit on the walls of the vessel, pipes, heat exchangers, pumps, or purification units; some will diffuse into the blanket gas. From the blanket gas, the fission products may be deposited on the walls of the units through which the gas flows, or they may enter the reactor containment system or the reactor off-gas system through leaks. During any fuel reloading process provision must be made for containment of the fission products.

If a fuel element melts down, most of the fission products will go directly to the coolant. Volatile fission products with no affinity for sodium and with relatively long half-lives move from the coolant to the blanket gas (e.g., ^{133}Xe , 5.3d; ^{135}Xe , 9.2 hr; ^{131}Xe , 12 d; ^{85}Kr , 10.3 yr). The short-lived volatile fission products leave decay products in the coolant (e.g., ^{137}Xe -3.9m $\rightarrow$ ^{137}Cs). Fission products that are not volatile and those that have an affinity for sodium are largely retained in the sodium. In case of an accident with the coolant (such as vaporization of the coolant through a leak in the coolant system, and possible consequent sodium fire), fission products may enter the reactor off-gas system. In a design basis accident, the fission products may pass directly to the reactor building or the reactor off-gas system.

1.0 INTRODUCTION

One of the prerequisites to large-scale power production by the operation of liquid-metal fast breeder reactors (LMFBR's) is a high degree of assurance that the containment systems and associated engineered safety features of these reactors can preclude the uncontrolled release of fission products to the environment. In order to minimize the hazard to the public and to reactor personnel, this assurance must hold not only for normal operation and minor accidents, but for severe postulated accident conditions.

The public acceptance and remarkable safety record of water-cooled thermal power reactors were achieved through just such a conservative approach and through exhaustive, continuing research and development work and great care in the design, testing, operation, and regulation of small experimental reactors, prototypes, and finally full-scale commercial reactors. Liquid-metal fast breeders are undergoing the same kind of conservative development and are now in the prototype stage. Therefore the amount of nuclear safety data accumulated through research and development work on LMFBR's and operating experience with them is still quite small in comparison with the tremendous body of corresponding information for water-cooled power reactors.

There is, however, a considerable background of coordinated research, development, and operating experience from which qualitative and semi-quantitative predictions of fission-product transport in liquid-metal fast breeders have been made. Designs of containment systems and engineered safety features have been based largely on these predictions. Also, much of the water-cooled reactor data on and experience with fission product buildup in fuels, high-efficiency air filtration, treatment of noble gases, and other factors is applicable or adaptable to the control of fission products in LMFBR's. On the other hand, fission product transport in LMFBR's is significantly different, primarily because of the effect of a sodium environment on the physical and chemical behavior of fission products.

The current emphasis in LMFBR design is on mixed uranium and plutonium oxide or carbide fuels, sodium coolant with argon blanket gas (both with cleanup systems), a nitrogen-filled containment system surrounding the coolant-blanket gas system to prevent combustion of sodium in case the

coolant system is ruptured, and an outer containment system equipped with an air-cleaning system to control sodium oxide aerosols, fission products, and plutonium in case of postulated accidents. A great amount of design effort is directed toward minimizing or precluding the possibility of super-prompt-critical excursions, which are conceivable in fast breeders and could cause massive releases of fission products and plutonium into the containment system. Much research, experiment, and testing is being done on vented fuel elements, particularly those with vents designed to release only long-lived volatile fission products into the sodium coolant.

The purpose of this report is to present a review of research, development, and experience in the release and transport of fission products in liquid metal fast breeder reactors; the control and retention of these fission products; and recommendations for research and development both in the near future and on a long-term basis. This report discusses the release of fission products from fast reactor fuel elements into the coolant and blanket gas; the ability of sodium coolants to retain certain fission products; and transport in and release from the coolant and the blanket gas. It also treats the various types of accidents, including possible major release of fission products into the reactor building.

The chapters on the release of fission products from fuel identify the fission products of major importance in such releases; two sections analyze the effects of type of fuel and mode of release on the amounts and types of fission products released. Oxide, carbide, and alloy fuel elements are described, as are fuel element venting devices and the effects of venting on the release of fission products into the coolant. A chapter on fission product retention in and release from the coolant describes the physical characteristics of sodium coolants; the fundamental mechanisms of fission-product retention in sodium; and experiments on retention and release.

The chapters on release to the reactor building and off-gas system deal with the combustion of sodium pools and sprays; the production, transport, and control of sodium oxide-fission product aerosols; and the characteristics of sodium-plutonium aerosols. Some of the subtopics in these chapters are natural control, deposition of fission products on surfaces, and coagulation of aerosols. Other subtopics are important

phases of the decontamination of off-gases, such as the sampling and filtration of aerosols.

Another chapter is a brief review of experience with fuel meltdowns and fission product release in a number of fast reactors of various designs; this chapter also reviews experience with two sodium-graphite reactors.

Recommendations on needed research and development work cover the release, transport, and control of plutonium and fission products; the control of sodium oxide-fission product aerosols; engineered safeguards; and inter-facility communication, particularly through information centers.

One of the appendices contains several discussions on fast reactor safety. The other includes several tabulations: thermodynamic data on certain fissile, fertile, and fission product elements; information on yields, hazards, and other characteristics of some 85 fission products; classifications, compositions, and melting points of fuel components and mixtures.

One of the key questions in the acceptability of liquid metal fast breeder reactors is that of safety. Ultimately, the hazards of nuclear reactors to the public involve fission product release and dispersion. Considerable research has been conducted in this area. However, the great majority has been directed to the loss-of-coolant accident in water reactors. The physical and chemical behavior of the fission products in a sodium environment is expected to be significantly different. Further research is required in fission product release under core vaporization conditions, directed to large-scale tests of engineered safeguards, and these tests must be related to realistic accident conditions. Laboratory and pilot plant experiments are required to design these tests and to interpret the results.

Fundamental information is required regarding the failure of a component in the primary system, resulting in the release of coolant and radioactive material. This escaping coolant transfers energy to the surrounding atmosphere and to the containment structure. A second source of energy release is a failure in the sodium-water heat exchanger, resulting in the intermixing of high-pressure water and steam with sodium.

The sodium-water reaction pressure must be accommodated or other failures in the radioactive primary circuit may occur.

Non-reactive gases are the most important elements to be considered in the gaseous release problem, for three reasons: first, they form an important fraction of all fission products; second, they will be affected by free diffusion only; and third, they cannot be easily removed from the gaseous phase. Only by free diffusion and convection are they dispersed into the containment or mixed during flow of the gases. In the category of non-reactive gases are the noble gases, such as krypton and xenon, and possibly nitrous oxide (which while not non-reactive, may behave as though it were). Radioactive decay, together with containment in a closed volume or dilution and discharge, or both, are the only really effective means now available of handling the noble gases.

Reactive gases, on the other hand, may not only change their state in passing into the containment system, but they may also change in concentration markedly while in passage. Reactive gases include hydrogen and tritium, oxygen, oxides of nitrogen, halogen gases or vapors (such as iodine and bromine), and their daughter products. It is necessary to know whether the reactive gases and vapors are in elemental or molecular states, what oxidation states may be available to them, and what chemical components are likely to be formed. The halogen gases, iodine for example, are notorious in this regard inasmuch as they may exist in various states of oxidation as inorganic compounds. Free halogens can also be adsorbed readily on both inert or various reactive surfaces. They react rapidly with most metals and, for instance, sealing plastics and rubber. They can combine with vapors or may be adsorbed on aerosols produced from volatilized metals or their compounds. They also form organic halides which are difficult to remove from the effluent stream by the usual methods. Sodium metal vapor if released will oxidize, and the oxides may become hydrated from moisture within the reactor atmosphere. The resultant oxide or alkali hydroxide will behave differently from metallic sodium in its reaction with halogen fission products, thus further complicating the picture for sodium-cooled reactors.

The state and characteristics of the environment when fission products are dispersed into containment may have a significant influence on their

behavior. In most cases, after a serious accident the fission products escaping from the fuel encounter more than one environment, often successively. A dispersion of a liquid metal coolant may be disseminated rapidly in the atmosphere surrounding the pressure vessel. If this atmosphere is air, oxidation takes place; if it is an inert gas, a vapor system and a condensation aerosol will result. In some cases, because of the severity of the metal-air reactions, it may be necessary to maintain an inert atmosphere in the containment. This will prevent oxidation reactions, but the condensation growth situation will be of importance. In effect, only the kind of condensing vapor will vary, and in some instances the vapors may already have reacted with many fission products.

The activity of the fission products in the blanket gas is generally considerable in relation to the activity of the gas itself. In the coolant, however, during operation and immediately after an accident, the ^{24}Na activity would be as hazardous as the activity due to fission products, or more so. The activity of the fission products in the sodium may become predominant, after decay of the ^{24}Na (15-hr half-life).

The requirements for containment of fast reactor accidents result from three classes of energy release: (1) primary system failure followed by loss of coolant into an inert-gas-filled cell, (2) sodium fire, and (3) prompt critical excursion.

The atmospheric dispersion of large quantities of fission products in a sodium-cooled reactor will probably be associated with large amounts of sodium oxide resulting from the burning of sodium. The chemical and physical interactions of a given fission product with sodium will significantly affect its release from fires and subsequent transport. Engineered safeguards will be required to prevent or control fires, as well as the air-cleaning systems necessary to control aerosols of sodium and fission products.

In the calculation of distance factors for the siting of pressurized-water reactors in the United States, DiNunno *et al.* assumed that a maximum credible accident would release into the reactor building 100% of the noble gases, 50% of the halogens, and 1% of the solids in the fission product inventory. In calculations for gas-cooled power reactors in the United Kingdom, Farmer's model has been used: 60% of the bromine, krypton,

and xenon, 25% of the ^{131}I , 15% of the ^{132}Te and ^{137}Cs , and 0.1% of the ^{89}Sr and ^{90}Sr .

At the present stage of development of liquid metal fast breeder reactors, estimates of release percentages can only be tentative; two of the most important factors that will affect these siting criteria are the radioactivity of the liquid metal coolant and the very high fuel burnup which results in a large fission product inventory.

Major sources of liquid metal fast breeder reactor safety information are the following:

- a. Nuclear Safety Information Center, ORNL
- b. Liquid Metals Information Center, Atomics International
- c. Liquid Metals Information at NASA Headquarters
- d. Information meetings at Argonne National Laboratory
- e. Research and development program at Battelle Memorial Institute
- f. IMFBR Program Office at Argonne National Laboratory.

2.0. FUELS AND FUEL ELEMENTS FOR FAST REACTORS

Uranium-plutonium fuels of three main types are being investigated for potential application to fast power reactors. These include mixed oxides, mixed carbides, and U-Pu-Zr or U-Pu-Ti ternary alloys.

Efforts in development and testing to date have been concentrated on oxide fuels. An initial group of the U-Pu oxide pins has been irradiated to 50,000 Mwd/ton in fast flux in the EBR-II at 14 to 16 kw/ft. One of the twelve pins failed, but it had only 40% of the fuel voidage of current reference design oxide fuel.

Thermal flux irradiations have been extended to about 125,000 Mwd/ton with only minor dimensional changes. Quantitative swelling measurements have been made, as a function of temperature, to burnups of 230,000 Mwd/ton. For fuel operating under typical fast reactor conditions (ceramic fuel surface temperatures of 1000 to 1800°F) the rate of fuel swelling with burnup is substantially lower than that in thermal flux measurements at lower temperatures. The reduced swelling is related to the release of nearly all fission gases at burnups above 4 to 5 at. %.

The effect of stoichiometry on thermal conductivity at low and high burnups has been observed; the effect is smaller at high burnups. The effect of stoichiometry on sodium compatibility has been further investigated and an apparently stable composition found.

Uranium-plutonium carbides are presently being investigated in two principal forms: (1) vibratory-compacted powders of (U,Pu)C or physically mixed powders of UC and PuC, and (2) pressed and sintered pellets of (U,Pu)C. Vibratory compaction is adaptable to remote handling and provides the amount of distributed porosity required to accommodate solid fission product swelling. Pressed and sintered pellets can be made with the desired effective densities either by sintering the pellets to low effective densities or by incorporating axial holes in high-density pellets. Both types of mixed carbide fuel are being evaluated under fast reactor conditions as fuel elements in the EBR-II. Preliminary examinations of a group of six rods recently irradiated to burnups near 3 wt. % with linear heat ratings in the range of 18 to 28 kw/ft show no significant dimensional or other external changes resulting from irradiation.

The most promising metal alloys for fast reactor fuels are U-Pu-Zr alloys. A typical composition is U-15 wt. % Pu-12 wt. % Zr. Alloys of this type have higher melting points than pure uranium, and they are more compatible with stainless steel. The fuel pins are usually injection-cast, then sodium-bonded to the fuel-element cladding. The required void space to accommodate solid fission-product swelling is provided by the sodium-filled annulus between the fuel and the cladding. Elements of this type are being investigated in the EBR-II; in a group of 16 rods irradiated to burnups near 4.5 at. % with linear heat ratings of 10 kw/ft, no significant dimensional or other external changes occurred in the elements as a result of irradiation.

Results of fuel and cladding investigations from fast-flux testing are becoming available at an increasing rate. A continuation of this trend is seen in the seven-fold increase in the number of oxide fuel pins completing irradiation in 1967 over the number in 1966. The introduction of unencapsulated pin irradiations into the EBR-II in 1967 along with the addition of numerous capsules will more than triple the number of pins tested to date, and represents the start of statistical testing of LMFBR fuels.

In studies of potential advanced driver fuels for a fast test reactor, an example of which might be the proposed Fast Flux Test Facility, seven carbide and eight alloy fuel cases were evaluated¹. Fuel-cycle parameters and reactor core designs were investigated with a BNW adaptation of the MIDAS computer code. Analyses of test-reactor core designs demonstrated that both mixed carbides and the newer alloy fuels (e.g., U-Pu-Zr) offer excellent potential advantages as advanced driver fuels for a fast test reactor. Predicted advantages include lower fuel-cycle cost, higher fast flux, attractive fabrication requirements, and safety.

Design analyses of fuel elements for the Fast Flux Test Facility are being performed at Battelle Northwest Laboratories.²

2.1 Oxide Fuel Elements

Fission gas release measurements have indicated that gas release increases with burnup and releases of 80 to 90% are typical at 4% burnup and higher. This gas release is apparently due to a porosity saturation

mechanism in which the open porosity of the fuel increases in a non-linear pattern resulting in increased gas release at higher burnups. The increased gas release is believed to work to advantage in minimizing fuel swelling.¹¹

Basic fuel swelling studies have been done using strong radial restraint. All swelling is forced to occur in the axial direction. Such experiments have provided valuable information on the rate of "inexorable" swelling and effect of peak fuel temperatures ranging from 1400 to 2200°C. The data have been extended beyond the previously reported range³ (to 150,000 Mwd/ton). At burnups to about 15 at. % the swelling rate is maximum at 1800°C central temperature and lower at lower temperatures. It is hypothesized that at lower temperatures (1400°C peak temperature), fission gases are largely retained, but the growth of fission gas bubbles is restricted by the strength of the mixed oxide lattice and the accommodation of fission gas atoms in interstitial lattice positions similar to those occupied by oxygen in slightly hyperstoichiometric oxide. As the temperature is increased, the creep strength of the lattice becomes lower and fission gas bubbles grow more rapidly, leading to a three-fold increase in swelling rate at 1800°C versus 1400°C central temperature. At temperatures above 1800°C, fission gas is released by diffusion so that only a small inventory of gas is retained at steady state. The major portion of further swelling arises from solid fission products. The swelling rates with strong radial restraint are substantially lower than prior data for UO₂. These results indicate that the swelling rate is sharply dependent on temperature. The fuel specimen operating below 1400°C show the least volume increase, while the intermediate temperature specimen at 1800°C indicates the greatest fuel swelling.

The performance of vibratorily compacted mixed oxide fuel rods under fast-reactor conditions was investigated by Brown et al.⁴ at ORNL. The measured gas release was nearly 65% of the theoretical amount generated by fission.

2.1.1 Effect of Stoichiometry in Mixed Oxide Fuels at High Burnup

A capsule which contained two fuel pins within a sodium-filled annulus was irradiated in the GETR to a burnup of 127,000 Mwd/ton (28.4×10^{20} fissions per cm³ of fuel) at the average pellet position. The top pin

contained 90.1%-theoretical-density solid pellets of UO_2 -25 wt. % PuO_2 with an oxygen/metal ratio of 2.00; values for pellets in the lower pin were 91.6% and 1.96. The test was designed to compare the behavior of stoichiometric and hypostoichiometric fuels at high burnup and high temperatures. The surface temperature of the cladding was 1050°F .⁵

No external dimensional change took place in the cladding. The maximum diametral increase was 0.5 mil (0.3 % ΔD). The original 2.4- to 3-mil diametral gap (annulus) no longer existed and fuel had swelled into this region that represented 2-1/2% of the original pin volume. Each pin had developed a longitudinal center hole that measured about 40 mils in diameter, about 6-1/4% of the fuel void. None of the large porosity typical of the original 90%-theoretical-density structure remained.

A dense region with grain-boundary porosity outlining columnar grains was observed in the fuel microstructure. The outer boundary of this structure, corresponding to the 1700 to 1900°C temperature of columnar grain formation, was not clearly defined but seemed to end at a high concentration rim of solid fission products.

Gas releases of 76 and 82% for the stoichiometric and hypostoichiometric fuels, respectively, were measured. These high values are typical for mixed-oxide fuels at high burnup and seem related to a lattice saturation mechanism.⁵

Operating under similar conditions, the two fuels showed gas releases that were approximately the same, and equivalent center hole diameter and location of columnar grain formation. Although previous testing⁶ at low burnup had indicated that hypostoichiometric fuel had a higher thermal conductivity, no significant differences could be determined in these specimens.

2.1.2 Fission Product Swelling of Oxide Fuels

Fuel swelling is presently the major design consideration for satisfactory fuel performance in fast reactors. To establish a better understanding of oxide fuel behavior at high burnups, an analysis of the swelling due to nonvolatile fission products in both UO_2 and $(\text{Pu}_{0.2}\text{U}_{0.8})\text{O}_2$ fuel was made.⁷

The procedure used to calculate the contribution of fission products in the swelling process of fuels was as follows: (1) determine the net amount of oxygen available from the fission process as a function of burnup; (2) define the concentration distribution and most probable oxidation state of fission products. Assign volumes occupied by these products; (3) compare the volume of fission products (and their compounds) to the volume of fuel present at a given burnup. By summation, compute the differences in volume with and without the fission products.

Theoretical considerations and experimental data show the most probable state of the solid fission products within the fuel matrix to be as follows⁷:

Soluble Elements - Zr, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd

Nonmetallic Inclusions - Sr and Ba Zirconates

Metallic Inclusions - Mo, Tc, Ru, Rh, Pd major components; no uranium and plutonium

Volatile Elements - Rb, Cs, I

Ionic Compounds - Br, I, Sr, Te.

Assuming a complete utilization of the vacancies created, the maximum solid fission-product swelling rate will be $0.13\% \Delta V/V$ per 10^{20} fissions/cm³ for UO_2 or $(Pu_{0.2}U_{0.8})O_{2.0}$. If none of the vacancies is consumed, the swelling rate will be as high as $0.54\% \Delta V/V$ per 10^{20} fissions/cm³.

The main conclusions from this work are as follows: the probable average volumetric contribution of the fission products in either UO_2 or $(U_{0.80}Pu_{0.20})O_2$ was calculated to be $\Delta V/V = 0.35\%$ per 10^{20} fissions/cm² of fuel. Depending upon operational thermal history of a given fuel element, the initial advantage of using a hypostoichiometric fuel of better thermal conductivity may diminish or disappear with increasing burnup. Because of available oxygen from the fission process, and its reaction with the interior of the clad, out-of-reactor compatibility tests of oxide fuels with cladding may not be relevant.

2.2 Carbide Fuel Elements

Stoichiometric and hyperstoichiometric carbide increases in volume at a rate of approximately 1.7 to 2.0% per atomic percent burnup.¹¹ If cladding strain is to be avoided, the fuel element must have enough void space to accommodate all fuel swelling expected during its lifetime. An

element designed to achieve 10 at. % burnup, for example, must contain void space within the fuel or radially adjacent to it that is equivalent to 20% of the fuel volume; this is provided if the effective fuel density does not exceed 83.5%.

The required void space in gas-bonded fuel can be incorporated by use of low-density pellets, high-density cored pellets, or compacted powders. All three approaches are under active development. In the case of sodium-bonded fuel, the required voidage is most readily supplied by the sodium-filled annulus between high-density pellets and the fuel cladding.

A sodium bond has unquestioned advantages over a gas bond in potentially increasing the thermal performance of carbide fuel rods. If, for example, the specific power of mixed carbide is to be 200 w/g with a maximum allowable center fuel temperature of 1500°C, a sodium-bonded design permits a fuel diameter of 0.39 in.; under the same conditions the maximum diameter of a helium-bonded carbide element is 0.26 in.⁸ On the other hand, sodium bonding accelerates carbon transfer from hyperstoichiometric carbide fuel to many common cladding alloys¹¹, and hyperstoichiometric (high carbon content) mixed carbide fuel is usually used because stoichiometric mixed carbide fuel is difficult to manufacture on a large scale.

Addition of powdered metals or compounds to nonstoichiometric (U,Pu)C powders for the purpose of improving compatibility with stainless steel has been proposed⁹. The additions or stabilizers react with the secondary phases to form more refractory and less reactive secondary-phase products and can be justified provided neutron economy and irradiation stability are not adversely affected.

Compatibility couples of unstabilized and Fe-stabilized fuel pellets helium-contact-bonded to Type 304 stainless steel were tested for 1000 hr at 650 and 800°C. No reaction was observed at the fuel-cladding interfaces. Also, compatibility couples of unstabilized, Fe-stabilized, and Cr₂₃C₆-stabilized hypostoichiometric (U,Pu)C pellets sodium bonded to Type 304 stainless steel pellets were tested at 800°C for 1440 hr. Metallographic examination of the fuel and cladding pellets and cladding tubing showed no evidence of reaction in any of the combinations¹⁰.

2.3 Alloy Fuel Elements

The favored most promising alloy fuel element design is based on a cylindrical fuel pin sodium-bonded to a tubular jacket¹¹. Sufficient annular fuel swelling is permitted to accommodate solid fission products and to enable the release of a significant fraction of the fission gases, which can be collected in a plenum in the upper end of the fuel element.

Recent observations on fission gas release¹² support the theoretical prediction¹³ that metal fuels will release substantial amounts of fission gas after fuel volume increases of approximately 30%. Metallographic examinations of irradiated metal fuel which had increased in volume to this extent indicate that fission gas bubbles have become interconnected so that the fission gas can escape to the surface of the fuel.

Fuel volume increases of 30% can be readily allowed without cladding strain by using a fuel element with ~ 75% effective fuel density. The necessary voidage is supplied by the sodium-filled annulus between the fuel pin and the jacket. Prototype uranium-plutonium alloy fuel elements of this type have been irradiated without jacket failure or distortion to burnups as high as 12.5 at. %. The prototype elements included a plenum large enough to accommodate both sodium displaced from the annulus and fission gas released from the fuel.¹¹

The successful outcome of these experiments indicates that a firm basis now exists for the design of high-burnup alloy fuel elements. Both released fission gas and fuel swelling from solid fission products can be accommodated without jacket failure or plastic strain¹¹.

U-Pu-Zr alloys are the most promising fast-reactor fuel alloys developed to date¹⁴. They are of interest not only as fuel for liquid-metal-cooled fast-breeder reactors, but also as advanced driver fuel for the EBR-II and the Fast Flux Test Facility.

Sixteen 34-in.-long U-Pu-Zr alloy fuel elements irradiated to burnups of about 4.3 to 5.1 at. % were recently removed from the EBR-II reactor. Neutron radiography of the 16 encapsulated irradiated fuel elements indicated that all elements were intact. Fourteen of the capsules were punctured and no fission gas was detected, which verified that the elements were intact. Dimensional measurements of the elements showed no significant changes as a result of irradiation. Transverse as well as

longitudinal metallographic sections of the fuel elements failed to show penetration of the fuel into the cladding materials. The sections also show that the fuel had swelled to the internal diameter of the cladding and the bond sodium had been displaced into the gas plenum¹⁴.

2.4 Safety Aspects of Vented Fuel Elements

Realization of the potential of fast ceramic reactors for achieving low fuel-cycle cost requires fuels of high burnup and high power density in order to minimize fuel fabrication, recovery, and inventory costs. These fuels, however, release considerable quantities of gaseous fission products, which must be either contained within the fuel cladding or released into the coolant and/or blanket gas. The fission gases can be retained in the core by thick cladding or by low density fuel, but both methods result in poor neutron economy.

Extension of the cladding to provide a plenum (reservoir) above or below the core reduces the pressure buildup in the fuel element by increasing the volume within its cladding. This allows a decrease in cladding thickness, but necessitates an increase in fuel element length, which is limited in practice by economic considerations. Furthermore, the buildup of fission gas pressure can still become quite high, because although the plenum is outside the core it is still an integral part of the cladding.

Buildup of fission-gas pressure within the fuel element cladding can be avoided by venting the fuel element into the coolant or into the coolant blanket gas. The objective is to relieve the pressure buildup due to fission gas without permitting release of daughter activity or ingress of sodium into the fuel-blanket region.

2.4.1 Venting Devices

Methods considered for venting include venting through a capillary tube to the cover gas (bypassing the coolant) and venting into the coolant through one of several devices designed to prevent coolant from entering the fuel element. In Fig. 2.1, one type of venting device is compared schematically with a conventionally clad fuel element and a vented-to-plenum element.

The General Electric diving-bell design is simple, does not extend to cover gas, and does not rely on moving parts or special material

Fig. 2.1 Schematic Comparison of Vented and Nonvented Fuel Elements
(from ref. 15)

properties¹⁵. The vent is shown schematically in Fig. 2.2. The volume within the bell, and thus the length of the vent, is determined by the criterion that sodium cannot be forced into the fuel region on shutdown.

A somewhat similar design considered at Atomics International¹⁶ includes a gas trap and fuel expansion space. The gas trap is designed to accommodate external pressure and temperature changes by balancing the changing hydrostatic head of sodium with an internally variable gas pressure. Variable gas pressure is achieved by permitting the gas in the trap to have a changing volume through the use of the internal tube within which the sodium stands at different heights. Liquid sodium is thus prevented from contacting fuel, and fission gases are vented through the trap.

Investigators at the Institute for Reactor Development in Karlsruhe¹⁷, while recognizing that the GE diving-bell design promises very reliable operation, began a study of porous plugs of sintered metal, with the goal of developing a venting device that would be much shorter than the diving bell, thus reducing coolant pressure drop. Smidt and Sommer¹⁷ stated that two problems are of importance: the ability to retain sodium from the fuel and the ability to stay porous for the fission gases. In the case of a reactor shutdown and lowering of the fuel temperature, the outside overpressure of the sodium amounts to 1-2 atm. In a simple apparatus the following plug materials were tested: nickel, iron, 18% Cr-8% Ni steel, and 18% Cr-10% Ni steel, with pore sizes between 1 and 5 microns. Sodium leakage started at a pressure difference of about 1.5 atm for sodium temperatures of 500°C, and at about 0.5 atm at 600°C.

The first plugging experiments by artificial evaporation of iodine did not lead to a pressure buildup of the gases. Smidt and Sommer considered more tests necessary for evaluation of the porous plug.

Computer calculations by Cherry and Rutland¹⁸ showed that, for the design parameters used, a pressurized sodium-bonded pin can withstand a greater degree of local flow blockage before fuel melting occurs than an unpressurized sodium-bonded pin (64% flow reduction as opposed to 48% for the cases studied). A pressurized sodium-bonded pin is less likely to lead to propagation of failure because the point of incipient fuel melting occurs at the top of the fuel rod rather than at the hot spot as in an unpressurized sodium-bonded pin.

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Fig. 2.2 "Diving Bell" Vent (from ref. 15)

Pressurization to 2.5 atm occurs in an unvented fuel pin when a pin filled at 300°F and atmospheric pressure is brought to normal operating temperatures. The increased boiling point is sufficiently high to maintain bond integrity and prevent fuel melting to the point of bulk coolant boiling, as is the case with helium-bonded pins. In vented fuel the bond and sodium coolant are at essentially the same pressure (atmospheric for cases analyzed); therefore, according to the calculations by Cherry and Rutland, loss of bond occurs before bulk coolant boiling. The significance of this effect on the venting of sodium-bonded fuel elements should be evaluated further by experiment and additional design studies.

2.4.2 Effect of Venting on Release of Fission Products to Coolant

The volatile fission products that escape from the fuel will be released into the coolant from every fuel element that is vented to the coolant, greatly increasing the volatile fission product burden of the coolant. On the other hand, the amount of nonvolatile fission products introduced into the coolant by leaching of the fuel is reduced to the extent that venting reduces cladding ruptures. Leitz¹⁹ emphasized the importance and desirability of having the first line of defense function properly, that is, to have the fission products insofar as possible retained within the fuel itself. In regard to this he remarked that the objective of venting a ceramic fuel to coolant is specifically to release selectively the long-lived fission gases so that the pressure stress on the fuel pin can be reduced; hopefully, this will lead to a less frequent failure of fuel pins, thus reducing contamination of the coolant through leaching. Leitz considered it altogether credible that the venting to coolant may, in fact, reduce the burden of fission product contamination in the coolant rather than increasing it. He stated further that he understood the leaching of ¹³⁷Cs from the fuel to the coolant to have led finally to the shutting down of the BR-5 reactor.

A preliminary demonstration test of vented-to-coolant fuel by O'Neill et al. at General Electric^{15,20} was designed to identify the fission products released, to measure their release fraction, and to determine the delay time experienced by the gases prior to their escape.

The test fuel rod was comprised of 95% dense (90% overall average including the fuel-cladding gap), 20 wt. % PuO₂-80 wt. % UO₂ pelleted

fuel, 0.25 in. in diameter by 5 in. long with a 2-in. long UO_2 blanket. The fuel operated at linear powers of 20 to 24 kw/ft in a thermal flux which approached conditions for central melting. Total exposure was 16,500 Mwd/ton. Results of this experiment are given in Table 2.1. Release of the long-lived and stable fission gases agreed closely with a calculated release of 40-50% for these operating conditions. Table 2.1 shows the dependence of gaseous release fraction on half-life. When these data were plotted as in Fig. 2.3, the slope gave the holdup time, which in this test was near five days. This explained the lack of daughter activity that escaped into the simulated coolant and cover gas.

The only fission-gas daughter released in significant quantity was ^{133}Cs , the daughter of 5.3-day ^{133}Xe . It turned out that neutron activation of ^{133}Cs to ^{134}Cs produced the majority of activity present outside the fuel, excluding that due to ^{24}Na . The release fractions of all other nuclides (including fissile) (^{89}Sr , ^{90}Sr , ^{91}Y , ^{95}Zr , ^{103}Ru , ^{131}I , ^{137}Cs , ^{140}Ba , and ^{144}Ce) were at or near the detection limit in the range from 10^{-6} to 10^{-10} . No deleterious effects to the fuel were observed due to possible exposure to sodium vapor. Traces of sodium were observed at the top of the "blanket", which may have been responsible for retention of radioiodine in this test.

O'Neill et al. concluded that the fissile elements and the nongaseous fission products and daughter elements did not distill out of the fuel (and blanket), and the gaseous fission products were sufficiently delayed in their release so that ^{134}Cs seemed to be the major concern to vented-to-coolant system design.

Table 2.1. Fission Products Released to Coolant or Cover Gas¹⁵

Nuclide	Half-life	Release Fraction
<u>Gases</u>		
⁸⁵ Kr	10.6 y	0.44
^{131m} Xe	12 d	0.30
¹³³ Xe	5.3 d	0.27
^{133m} Xe	2.3 d	0.05
¹³⁵ Xe	9.2 h	0.0003
<u>Solids</u>		
¹⁰³ Ru	40 d	4×10^{-10}
¹³¹ I	8 d	1×10^{-8}
¹³⁷ Cs	29 y	1×10^{-6}
Other (Sr, Y, Zr, Ba, Ce)		10^{-6} to 10^{-8}

Fig. 2.3 Fuel-Pin Holdup Time (from ref. 15).

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3.0 FISSION PRODUCT RELEASE FROM REACTOR FUEL

In an accident that leads to extensive failures of fuel element cladding, fission products will be released from the fuel. These fission products may be partially or totally trapped in the primary coolant and blanket gas, may be released to the secondary containment, or may in part escape to the atmosphere.

The quantities of fission products in a given reactor core at a given time can be accurately estimated from the history of that core.¹ However, the fraction that will be released under a given set of circumstances cannot be accurately estimated in any easy way.

The ability to evaluate the release of fission products is dependent upon knowledge of their chemical and physical forms. Processes favoring release of fission products to the environment and those favoring retention in the fuel were discussed by Parker et al.² The chemical forms of the fission products have a strong effect on their release from fuel during reactor accidents and on their subsequent behavior. Although methods of determining directly the chemical form of released fission products remain to be devised, their probable chemical forms can be inferred from observations of the effects of various environments on the extent of fission product release.

The two physical properties most relevant to fission product release from overheated reactor fuels are the vapor pressures of the elements and compounds that can form under accident conditions and the free energies of formation of the compounds; the vapor pressures are reflected in volatilities, and the free energies of formation indicate the stability of the compounds at elevated temperature (Appendix B, Table B1). Thermodynamic data apply, strictly speaking, only to equilibrium conditions, which seldom, if ever, exist in reactor accidents. Consequently, it is necessary to be cautious in using such data to predict the behavior of fission products under accident conditions. However, at elevated temperatures resulting from loss-of-coolant accidents in reactors with high melting fuels, equilibrium will probably be at least approached, so that conclusions based on thermodynamic considerations are of some value. Kingery and Wygant³ have discussed other limitations on thermodynamics, with special reference to ceramic materials.

3.1 Fission Products of Major Importance in Releases from Fuel

The first step in analyzing the release of fission products from core to containment is to determine the amount of fission products present at any given time in core life and at any time after shutdown. The following information is required to permit the calculation of a given isotope formed during fission: composition of fuel, time of irradiation, flux density of the reactor or facility, relative fission yields of each isotope along the decay chain of the mass number of interest and any branching ratios involved, half-lives (or decay constants) of the isotopes along the decay chain, and cross sections for neutron capture by the nuclides, if these are significant, to the degree of accuracy desired. Several computer codes are available to make these calculations (e.g., ISOGEN at BMI).

Over 200 different isotopes are formed in the fission process. The most important of these are listed in Appendix B, Table B2, approximately in order of the volatility of the elements but with some changes dictated by observed behavior in fission-product release experiments.

Since there are so many elements and radioactive species in the fission spectrum, it is desirable to simplify the analysis, by reducing the number of species under consideration. One such scheme is shown in Table 3.1. A study of Table B2 shows that krypton, xenon, and iodine are very important, because they are volatile and are present in large quantities; in addition, iodine concentrates in the thyroid and, consequently, is biologically potent. Radioactive cesium, even though its yield is low, is moderately volatile and, when ingested, constitutes a hazard to the whole body. Tellurium has very short-lived high-yield isotopes. It also is moderately volatile and contributes a health hazard through the ^{132}I daughter product of ^{132}Te . Strontium has a relatively high radioactivity and is a long-lived hazard to bone and lung, even though it is relatively non-volatile. Barium is of high yield and slight volatility and hazardous to bone and lung tissue. Ruthenium is highly volatile under strongly oxidizing conditions. Other isotopes in Table B2 are excluded from Table 3.1 because of low volatility, low fission yield, relatively low toxicity, or very short half-life.

Table 3.1 The Most Hazardous Fission Product Activities in Order of Decreasing Volatility

Element	Activity [Kilocuries/Mw(thermal)] After 1 yr of Irradiation		Weight (g)
	At Shutdown	1 Day After Shutdown	
Xe, Kr	385	62	61
Iodine	289	53	2.6
Te, Cs	220	37	40
Ru	27	27	19
Sr, Ba	144	98	31

The time in core and the power level of a reactor are major factors in determining the inventory of fission products. The short-lived isotopes reach their saturation concentration soon after reactor startup, while the longer-lived activities, those with half-lives in the range months to years, continue to accumulate during an average reactor core life. After extended irradiation, about 50% of the total activity is always associated with the refractory elements (the rare earths, zirconium, and niobium). The halogens, rare gases, alkaline-earth metals (barium and strontium), and tellurium are each present initially to the extent of 10 to 15% of the total activity. Tellurium falls below 8% after 24 hr or less, and iodine drops below 6% in not more than 10 days. The barium-strontium group is the only one of the four mentioned above that gains in abundance after 10 days. The refractory group continues to increase to 70 to 80% of the total after this short period. For periods longer than 50 days, ^{106}Ru , ^{137}Cs , and ^{90}Sr become the most significant radioactivities in addition to the refractory group.

Special mention of ^{239}Np and ^{239}Pu is required in regard to reactor safety. In low-enrichment reactors, neutron capture by ^{238}U continuously produces 2.3-day ^{239}Np , which contributes a large fraction of the total gamma activity of the core at shutdown. Decay of ^{239}Np produces ^{239}Pu at a high rate in fast breeder reactors, in addition to the ^{239}Pu in the fuel at startup. Fortunately, neptunium and plutonium both form stable oxides (NpO_2 and PuO_2) which are comparable in volatility to UO_2 , and a large fraction of these biologically potent species would be expected to remain in the reactor core or the primary coolant under most accident conditions.

3.2 Effect of Type of Fuel on Fission Product Release

In considering the release of fission products, it is convenient to divide the fuels into six classes (as shown in Appendix B, Table B3): metals and alloys, oxides dispersed in a metallic matrix, oxide fuel material, carbides, miscellaneous compounds (sulfides and nitrides), and liquid fuels.¹

The metals and alloys are generally characterized by normal metallic behavior and by melting points below 2000°C; there are some minor variations in the behavior of these metals and alloys on melting, due to differences in their chemical activity. In the case of the oxides dispersed in a metallic matrix, the melting point of the fuel is still determined by that of the matrix, but there are some differences in the behavior of the dispersed oxide. The oxide fuel materials are generally characterized by melting points above 2000°C and by a relatively high degree of chemical inertness. The carbides, in general, have high melting points but are characterized by vigorous chemical reaction with a number of materials which could conceivably be part of the reactor system, such as air and water. The miscellaneous compounds (two uranium sulfides and uranium and thorium nitrides) are grouped together for convenience. The liquid fuels are salt and alloy fuel systems that are liquid at reactor operating temperatures.

Fuel melting temperatures have been included in Table B3 because this is one of the most important physical properties affecting the extent of fission product release. Melting ranges estimated from published phase diagrams are given for some materials that do not have unique melting points. The amount of a fission product released on heating irradiated fuel is largely a function of the fuel melting temperature and the vapor pressure of the fission product; however, since identical experimental conditions have rarely been maintained in fission-product release experiments with different fuel materials, it has been difficult to establish the nature of the relationship. Furthermore, the release of fission products is also affected by chemical reactions, solubility in the fuel or its environs, ratio of fuel surface area to fuel volume, duration of molten condition, composition and pressure of cover gas, and other parameters.

The degree of melting is always difficult to postulate, since an uncertainty exists concerning the rate of heat loss from the reactor core. The melting of fuels with reasonable burnup levels invariably leads to high release rates for the volatile elements (Xe, I, Te, Cs). The lowest-melting fuels may also release some Ru and Cs, in addition to Xe, I, and Te; the higher-melting ones release all of these and may also release some Sr and Ba.

Data on release of fission products from various types of fuel were collected by Parker and Barton¹ and arranged according to the fuel classification scheme given in Table B3. Some information on oxidation of fuels is included for materials that oxidize readily under possible accident conditions. An understanding of factors affecting oxidation rates is essential for an adequate assessment of the hazard of the release of fission products that may result from destruction of fuel elements by oxidation. Where experimental information on fission product release from a fuel is lacking, a similar fuel system for which there is information is indicated as the best available source of information.

Information on the release of several specific types of fuels is contained in Table B3. These fuels include (U,Pu)O₂, (U,Pu)C, UN, US, US₂, plutonium alloys, and PuO₂.

3.3 Release of Iodine from Uranium and Uranium Monocarbide

In studies on the chemical states of iodine released from uranium and UC into helium between 1200 and 2500°C, Castleman and Tang⁴ found that in an inert atmosphere, two iodides and atomic iodine are released from metallic uranium and UC fuels (Table 3.2). The relative quantities of each species are primarily a function of fuel temperature. The two iodides are tenaciously held on surfaces at temperatures of ~ 275 and ~ 200°C in an inert atmosphere, but readily evolve elemental iodine into an oxidizing environment.

Castleman and Tang also found that where iodine is released during a uranium metal-water reaction, conversion to HI can be validly estimated from thermodynamic considerations. In a sodium environment, however, essentially all of the iodine will be converted to NaI when the fission product concentrations are of the order of parts per million.

Table 3.2. Summary of Metallic U and UC Studies in Inert Atmosphere⁴

Fuel Sample	Probable Iodine States	Conditions and Remarks
U	UI	~ 1200° - 2000°C Plateout temperature 275°C Synthesized deposit composition UI _{0.9}
U	UI + I	~ 2100° - 2500°C
U	(U _x I) + I	> 2500°C (U _x I) plates out at 200°C
UC	Species generally similar to metallic U	Primarily atomic iodine released at temperatures in great excess of the melting point of UC

A study by Castleman et al.⁵ of the diffusion of iodine in hypo-stoichiometric uranium monocarbide was carried out to measure the rate of iodine release from irradiated arc-cast UC spheres ranging in diameter from 0.3 to 0.4 cm. Diffusivities were obtained from these data by employing the appropriate solution to Fick's law for diffusion in a sphere. The primary objective of the experiments was to ascertain whether diffusion within individual grains or within the entire spherical specimen was the rate-controlling step in the diffusion process. To accomplish this, several UC specimens (average carbon content 4.60 wt. %) were annealed to 1760° and 1850°C prior to irradiation. The diffusion from these specimens was studied in the usual manner. Diffusion coefficients, calculated by assuming the sphere radius to be the controlling diffusion boundary, were plotted vs reciprocal absolute temperatures together with diffusion coefficients evaluated from unannealed specimens. This plot showed wide scatter and no general correlation between the specimens.

Subsequently, the experimental data were recalculated using grain size data obtained from UC annealing studies carried out with the same material. These experiments showed that the grain structure of the carbide specimens is important in determining the release rate. Diffusivities were calculated from the appropriate solution to Fick's law,

using average grain size as the diffusion geometry. The logarithms of the diffusivities, calculated on this basis for unannealed specimens as well as those annealed at 1760° and 1850°C, were found to vary linearly with the reciprocal of the absolute temperature, giving an activation energy for iodine diffusion in UC of 110 kcal/mole.

Transport studies are being conducted by Castleman et al.⁵ to develop an understanding of the factors controlling fission product vaporization from molten materials. It is expected that the results will eventually lead to a model for predicting fission product release from molten fuels and claddings as a function of time, temperature, volume-to-surface area ratio, and fission product solubility in the condensed phase.

A number of analytical models were developed for predicting fission product release rates from condensed phases. All of these indicate that the volume-to-surface area ratio (liquid depth) of the condensed phase is important in influencing release rates.

Recent analytical and experimental studies⁵ proved that fission product release from relatively nonvolatile condensed phases is diffusion controlled. An experimental investigation of iodine release from 0.3, 0.8, and 1.6-cm deep liquid uranium samples showed the percentage release vs time to be strongly liquid-depth dependent. The important parameter is the dimensionless diffusion parameter, which is equal to the diffusivity multiplied by time and divided by the liquid depth squared. This result was expected for mechanisms following Fick's law.

3.4 The Light Bulb Model of Fission Product Release

Experiments in various laboratories and in reactors have provided much data on release of fission products under conditions that simulate reactor accidents. The effects of important variables on fission-product behavior have generally been recognized, but interpretations of the data have been largely empirical. The "light bulb" model of fission product release proposed by Miller⁶ seems to explain most, if not all, of the available release data.

Miller comments that Fonda⁷, in studies of the incandescent lamp, observed that tungsten filaments lost weight (at a given temperature) more rapidly in a vacuum than in the presence of a nonreactive gas and related this to Langmuir's⁸ theory that heat loss from incandescent wires

in gases is controlled by conduction of heat through a stationary gas film around the wire. Fonda proposed that evaporation of material from the filament in a nonreactive gas is controlled by diffusion through a similar stationary gas layer. Miller points out if it is assumed that a heated specimen of reactor fuel has such a non-reactive gaseous boundary layer and that diffusion (by Fick's law) through this layer controls the rate of release of fission products, then the theories of Langmuir and of Fonda may be applied.

The model has been tested with data from several sources and satisfactorily explains the observed behavior of fission products and fuel. The model describes the dependence of the fraction released on the composition and pressure of the cover gas, temperature, surface area, and amount of the molten fuel (solvent), the time at temperature, the chemical form of the fission products. The success of the model in predicting correctly the dependence of fission product release rate on identity of cover gas is especially significant, since other theoretical release models do not predict such a dependence.

Application of the model to the emission of fission products below the melting point of UO_2 indicated that in post-irradiation annealing experiments above 1600°C , the rate-limiting step in the release of fission products and the vaporization of UO_2 is boundary layer diffusion. The model shows that the evaporation of fuel contributes substantially to the release of the fission products and must be considered in an explanation of fission product behavior.

The model indicates that maximum possible release of fission products and fuel would occur in a vacuum and that the release could then be expressed by a modified Langmuir equation for vaporization. It is most likely, however, that at large scale the gas film will be present and that a concentration gradient will exist. It is further likely that the small-scale experimental data may indicate greater release than would actually be experienced at large scale, since the actual concentration gradient may be less than that postulated for the light bulb model, in which the concentration outside the gas boundary was assumed to be zero. This possibility points out the need for larger-scale fission product release experiments to investigate the effects of size on the release mechanism.

The subject of scale-up as it pertains to the U. S. Nuclear Safety Program has been discussed further elsewhere.^{9,10}

This process of modeling simulated reactor accident events will continue. As the program of scale-up in experiments on fission product behavior develops, models such as the light bulb would be refined and developed, so that the consequences of a maximum credible accident can realistically be evaluated in relation to the siting of reactors near populated areas.

3.5 Effect of Mode of Release on Amounts and Types of Fission Products Released

The release of fission products from the fuel, through the sodium bond (if sodium-bonded), through the cladding, and into the primary coolant can be of any degree of severity and can also differ in regard to what fission products are released to the coolant. These differences in degree and type of release are dependent not only on fuel, burnup, flux, decay and capture processes, and operating temperatures but also on the extent of plateout or sorption on the inside walls of the fuel element, the extent of containment by filters or traps within fuel elements of certain designs, the size and location of the orifice through which they escape the cladding, and the fuel temperature at the time of release.

Fission product releases by diffusion through intact cladding or by leakage through pinholes or very small cracks in the cladding are usually an operational problem only. Releases from larger cracks become a safety problem when the cumulative product of crack sizes and number of defective fuel elements contaminates the primary coolant to a level that significantly increases the hazard of a rupture in the primary coolant system.

If the fuel elements are vented (see Chapter 2) to the coolant, the volatile fission products that escape from the fuel will be released into the coolant from every vented element, greatly increasing the volatile fission product burden of the primary coolant. On the other hand, to the extent that venting of fuel elements reduces cladding ruptures, the amount of nonvolatile fission products introduced into the coolant by leaching of the fuel will be reduced. This is discussed in detail in Chapter 2.

In the case of fuel meltdown, the quantity and type of fission products introduced into the coolant depends again on a number of factors, particularly the degree of meltdown, the duration of the molten condition and the accessibility of coolant to the melted fuel. In the extreme case for a single fuel element, when meltdown is complete and coolant sodium is in intimate contact with the fuel, essentially all the fission products from that element may circulate freely in the primary coolant system if the fuel is fragmented sufficiently on solidification. Melted fuel elements can, of course, plug the coolant system, causing meltdown of other elements, and under certain conditions can increase the flux density in the lower part of the core enough to produce a fast transient, but these two problems are beyond the scope of this chapter.

3.6 Tritium Generation and Hazards

An investigation of tritium generation and release in PM (portable medium) nuclear power plants was conducted by Ray, Wooton, and Barnes¹¹. The investigation revealed that tritons generated in the ternary fission of ^{235}U are by far the major source of tritium. Other sources contribute approximately 10% to the total production. In the ternary fission process, the triton fission fragments are generated at the rate of approximately 8.7×10^{-5} triton per fission, with an apparent Gaussian energy distribution about a most probable energy of 7.5 to 9 Mev. The resultant tritium migrates to the reactor primary coolant by diffusion from the fuel elements.

Tritium decays by emission of a soft beta particle (18 kev end-point energy, 5.7 kev average energy) with no attendant gamma radiation. Consequently, it presents little radiological health hazard so long as it is prevented from entering the body. Furthermore, exposure to an atmosphere containing gaseous T_2 or HT is a relatively minor hazard, because less than 0.1% of the assimilated gas is retained in the body.¹² This is reflected by the relatively high concentration limit for submersion in an infinite cloud containing tritium gas as given in the Code of Federal Regulations, 10 CFR 20 (2×10^{-3} $\mu\text{C}/\text{ml}$). The hazards in such an exposure appear to result from the formation of tritiated water, HTO, by oxidation of some of the tritium gas in the body¹³.

The primary hazard of tritium is exposure to HTO vapor or liquid, because of the rapid uptake of this material. Over 98% of the tritium activity inhaled as HTO vapor is absorbed through the respiratory system, and absorption of HTO through the total skin area occurs at a rate comparable to that through the lungs.¹³ Liquid HTO is hazardous only when ingested into the system. The exposure limit for a 40-hr week is given in 10 CFR 20 as 5×10^{-6} $\mu\text{C}/\text{ml}$ for HTO vapor in air, and as 1×10^{-1} $\mu\text{C}/\text{ml}$ for HTO in liquid water.

3.7 Meltdown of Fuels in Sodium

3.7.1 Analyses of Meltdown Experiments in the TREAT Facility

Two series of experiments, each containing three sample clusters in flowing sodium, were run in transient loop experiments in the Transient Reactor Test Facility (TREAT). Each cluster contained a single EBR-II Mark-I-type pin surrounded by a ring of six dummy pins, all on an EBR-II pitch. Results of both series have been reported.^{14,15} Analyses of these cluster experiments, together with analyses of earlier TREAT experiments on single EBR-II pins in stagnant sodium capsules¹⁶ have also been reported¹⁷, as a summary of the knowledge gained on the behavior of single EBR-II type fuel pins when subjected to severe transient heating in a sodium environment.

On the basis of the earlier stagnant sodium experiments and the detailed analyses of the single-pin loop tests, the following conclusions were made:

1. Maximum fuel-cladding interface temperatures $> 950^{\circ}\text{C}$ produce extensive alloying of fuel and steel cladding.
2. For the thermal conditions typical of stagnant sodium, an interface temperature of 1000°C marks the approximate failure threshold, with cladding penetration by eutectic formation presumably playing the dominant role; and cladding rupture due to internal pressure can be predicted.
3. In the case of flowing sodium, an interface temperature of 1000°C can be greatly exceeded without failure if failure is predicted neither by cladding penetration nor internal pressures. If bond sodium pressures > 10 atm are calculated, molten fuel

will be driven upwards into the expansion space inside the cladding. Upward movement of fuel within the cladding can relieve pressure so that a pressure rupture does not occur. Failure did not occur when the interface temperature was rising at approximately $4000^{\circ}\text{C}/\text{sec}$ at the time of calculated pressure rupture, but it did occur when the calculated rate was approximately $7000^{\circ}\text{C}/\text{sec}$.

3.7.2 A Large High-Temperature Flowing Sodium Loop for Fast-Reactor Fuel Element Meltdown Experiments

The Transient Reactor Test Facility at the National Reactor Testing Station is a pulsed reactor available for transient experiments on fuel elements¹⁸. A sodium loop has been constructed at this site.¹⁹ The large TREAT loop is a unique facility for fuel meltdown studies under flowing sodium. A facility of this type has not been available previously to experimenters working in the area of fast-reactor safety.

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4.0 FISSION PRODUCT RETENTION IN AND RELEASE FROM LIQUID SODIUM SYSTEMS

In the analysis of reactor accidents, there are several factors unique to sodium-cooled, fast reactors. These include: (1) the potential release (defected fuel elements or accident) or intentional (vented fuel elements) release of fission products into sodium, (2) gettering properties of sodium for many of the fission products, (3) radioactivity of the primary sodium coolant, (4) sodium combustibility, and (5) the presence of sizeable quantities of Pu. Item (2) is of primary concern in this chapter.

Fission products and plutonium can be released from the reactor fuel under various conditions, ranging from leakage from vented fuel elements, or leakage and washout by the coolant of defected elements, to a major release during melting or a massive release during a violent nuclear excursion. A large and difficult research program will be required to identify fission-product species and fractions released for each set of circumstances and for degrees of violence to be anticipated in a nuclear excursion.

The retention of fission products in the coolant; rate and extent of release to cover gas; plateout in the sodium or gas system; and chemical form as a function of temperature, temperature gradient, coolant-flow rate, and fission-product species should be determined for normal reactor operating conditions.

The major physical characteristics of a sodium system to be considered in the evaluation of a program to study fission product transport are¹ the following:

1. Sodium as a reactor coolant has excellent heat transfer characteristics which may serve to modify the physical form of fission products and fuel fragments released into it.
2. Characteristically, the sodium coolant operates at temperatures of 400 to 500°F below its boiling point at essentially atmospheric pressure.
3. It is chemically reactive with many of the fission products released from fuel.
4. The system is highly pyrophoric and requires the presence of an inert cover gas at all times when it is in a molten condition.

The initial conditions of fission product release in various reactor accidents are a function of burnup, environment, etc. Also, throughout the transport path to environmental release, there are frequent modifying conditions which may serve to reduce the potential radioactivity, and to change the initial physical or chemical form in which the fission products may have been released. For example, chemical reaction, solubility, precipitation, plating and deposition on reactor structures may occur during either the sodium transport phase or during the transport of sodium vapor carrying or in combination with released fission products. In either case, the amount of mobile activity present is reduced.

The rate and extent of vaporization of fission products from sodium, the chemical states of the vaporized fission products, and the thermodynamic properties of fission product-sodium solutions are being studied both experimentally and theoretically at Brookhaven National Laboratory. Experimental and analytical studies on the behavior of fission products released to the sodium coolant during normal operation, including the extent of retention in the sodium, the rate and extent of release to the cover gas, and the rate and extent of plateout on surfaces, is being conducted at Atomics International. The transport, plateout, and trapping of long-lived fission products in a sodium loop are being investigated at Los Alamos Scientific Laboratory. Experiments in an intermediate-scale system are being conducted at the French CEA's Centre d'Etudes Nucleaires, in Cadarache. These investigations are described briefly in this chapter.

4.1 BNL: Vaporization of Fission Products from Sodium and Thermodynamic Properties of Fission Product-Sodium Solutions

Experimental and theoretical studies on the behavior of fission products in sodium are being conducted at Brookhaven National Laboratory to provide three types of fundamental data: the thermodynamic properties of fission product-sodium solutions, the extent and rate of release of fission products during vaporization of the sodium, and the chemical states of fission products vaporized from sodium. Studies reported by Castleman, Tang, and Mackay² showed that at concentrations expected in reactors, iodine vaporized as NaI from sodium while cesium was transported

as the element. The vaporization of cesium and NaI from sodium solutions was studied experimentally. The vaporization behaviors of strontium, cesium, barium, rubidium, and tellurium were predicted from theoretical considerations.

From these studies (described in more detail in the subsections below), Castleman, Tang, and Mackay drew the following conclusions: At concentrations equivalent to those expected in a reactor, cesium iodide is not stable in a sodium environment. Free cesium and sodium iodide are the important species vaporized from sodium solutions. The excess free energy of mixing of sodium iodide in sodium at infinite dilution was measured experimentally, yielding the value 9100 ± 400 cal/mole. Excess free energies of mixing at infinite dilution, which are generally difficult to obtain experimentally, are readily obtained from equilibrium vaporization techniques. A method (proposed by Tang, Castleman, and Mackay) for extrapolating activity coefficients to infinite dilution gave satisfactory excess free energy of mixing values for the systems Cs-Na, Rb-Na, Sr-Na, and Ba-Na. The excess free energy of mixing values for the systems Sb-Na and Te-Na are somewhat less certain. The vaporization of cesium from sodium is in general agreement with calculated vaporization curves. These studies show that fission product vaporization from sodium can be satisfactorily calculated knowing values of excess free energies of mixing and the appropriate vapor pressures.

4.1.1 Chemical States of Iodine and Cesium Vaporized from Sodium

A comparison of the estimated standard free energies of formation of the uranium iodides with that of sodium iodide showed that if the uranium iodides entered sodium they would undoubtedly react to form NaI. In considering reactions of iodine with other fission products, compound formation with cesium was found to be among the most probable, based on relative concentrations and thermodynamic values. An attempt to calculate the CsI/NaI distribution in sodium as a function of composition and temperature² revealed that, although sufficient information was available for calculating the activity coefficients for cesium and sodium iodide in sodium, no such data were available for cesium iodide. Therefore, no accurate prediction of the relative distribution could be made. In view of the uncertainties in the theoretical calculations, an experimental investigation of the Cs-I-Na system was conducted.

A study of the species vaporized from solution proved to be useful for determining the relative importance of NaI and CsI in sodium systems. Since interactions between cesium and iodine might occur, it was desirable to assure that the quantity of CsI formed would not be hindered by the reaction kinetics. Therefore, it was decided to start with CsI, dissolve it in sodium, and study the vapor species above the solution.

A thermochromatographic method³ for studying the states of semi-volatile fission product vapor species was used. Briefly, the method consists of separating the fission products vaporized from a solution by condensing them or their compounds in a tube having a linearly descending temperature profile along the wall. The deposition temperature of the species is determined by the radioactivity distribution in the tube at the completion of an experiment. The behavior of these species is compared with that of standards in much the same manner as done in conventional chromatography.

In these experiments², CsI was tagged with ^{134}Cs and ^{131}I , dissolved in liquid sodium contained in a stainless steel boat, and then placed in the thermochromatographic apparatus. A thin-walled stainless steel liner was placed inside the usual quartz deposition tube to eliminate reactions between sodium and quartz. After the apparatus was heated to a predetermined temperature, the boat containing the sample was rapidly introduced into the sample heating zone. The species were released into a helium carrier gas at a fixed temperature and their respective deposition temperatures were measured. The deposition profiles of these species were compared with those of sodium iodide and cesium iodide.

The results of the experiments, carried out over the range 485 to 748°C and with cesium and iodine concentrations as high as 7.7×10^{-3} mole fraction, are shown in the bottom graph of Fig. 4.1. Without exception, these results show that cesium and iodine are independently transported in the vapor phase. The only difference between the experiments at low and high temperatures was the length of run needed to transport a given quantity of material away from the sample boat; the distribution of the product was not affected.

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Fig. 4.1 Thermochromatograms for the System Na-I-Cs (from ref. 2).

Other experiments were carried out to prove that CsI is stable under these experimental conditions in the absence of sodium. The results, shown in the central graph of Fig. 4.1, clearly show that CsI is transported and deposited as the compound. Note that these ordinates show the relative counting rates of the two species and do not represent the total quantities of the atoms.

A third set of experiments was performed employing NaI in place of the CsI. Sodium iodide was also observed to deposit as the compound (top graph of Fig. 4.1). The deposition temperature of pure NaI (465°C) is in good agreement with that of the iodide transported from the CsI-Na solution (475°C). These experiments show that CsI converts to NaI and cesium in a sodium environment.

Subsequent to these free vaporization experiments, a similar investigation, using a Knudsen cell in place of the open sample boat, showed essentially the same results: within the limits of detection all the iodine is transported as sodium iodide and the cesium is transported as the pure element.

4.1.2 Vaporization of Fission Products from Sodium

The vaporization of sodium iodide and cesium from sodium solutions was measured experimentally, employing equilibrium vaporization methods.² Castleman, Tang, and Mackay mentioned that equilibrium vaporization follows the Rayleigh equation

$$f_i = 1 - (1 - f_j)^A,$$

where f_i and f_j are the fractions of species i and j vaporized. Here A is given by

$$A = P_i^{\circ} \gamma_i / P_j^{\circ} \gamma_j,$$

where P_i° and P_j° are the vapor pressures of the species i and j , and γ_i and γ_j are their respective activity coefficients. If sodium is considered to be component j , the Rayleigh equation above relates the fraction of fission product i vaporized to the fraction of sodium vaporized. Fortunately, in the systems of interest here $\gamma_j \approx 1$. Therefore γ_i is the appropriate activity coefficient at infinite dilution and a function of temperature only.

An equilibrium distillation apparatus was constructed to measure the relative vaporization of cesium and sodium at constant temperature. The

apparatus consists of an isothermal vaporization cell and a condenser, connected by a heated transport line. A cesium-sodium alloy, tagged with ^{22}Na and ^{137}Cs , was introduced into the apparatus under an inert atmosphere. The apparatus was heated to a specified temperature and evacuated. The vaporization of the alloy was continuously monitored with suitable scintillation counting equipment. The experimentally determined vaporization curves are in general agreement with the calculated curves shown in Fig. 4.2. The vaporization curves were calculated using the Rayleigh equation given above, together with the required activity coefficients evaluated from excess free energy of mixing values, which were calculated from the cesium-sodium phase diagram (see Section 4.1.3).

A Knudsen cell was employed to study the vaporization of sodium-sodium iodide solutions. Sodium and sodium iodide, tagged with ^{22}Na and ^{131}I , were released into a flowing helium stream maintained at a pressure equivalent to that of the sodium at the experimental temperature. The vaporization of sodium and sodium iodide was continuously followed using a scintillation counting method. Although the equations of molecular flow do not apply in the pressure range employed, this cell did enable equilibrium vaporization experiments for a system with components of widely varying volatility. Experiments at 650, 710, and 830°C gave the excess free energies 9700, 8950, and 8600 cal/mole, respectively. The experimental error is approximately $\pm 4\%$. Even though these values show the same trend with temperature as those estimated by Pitzer⁴, within the precision of the experiments, the use of the average value 9100 ± 400 cal/mole was recommended.

With the activity coefficients calculated from this excess free energy, the vaporization curves for the sodium iodide-sodium system were calculated for 700, 900, and 1100°K. These are also shown in Fig. 4.2.

Curves for the equilibrium vaporization of the fission products barium, strontium, rubidium, and tellurium from sodium are shown in Fig. 4.2 as well. Here, the activity coefficients were calculated from the appropriate free energy of mixing values (see Section 4.1.3). The vaporization of antimony is negligible ($< 10^{-3}\%$ for $\sim 100\%$ sodium vaporized at 1100°K) and is not plotted.

Fig. 4.2 Fission Product-Sodium Equilibrium Vaporization (from ref. 2).

4.1.3 Thermodynamic Properties of Fission Product-Sodium Solutions

Castleman, Tang, and Mackay² stated that the equilibrium properties of variable-composition liquid systems can be accurately calculated employing the methods provided by the classical thermodynamics of solutions, and that one of the most useful concepts is that of the chemical potential, which is the free energy of mixing of component i in a solution.

The excess free energy of mixing, ΔF_i^E , was given by

$$\Delta F_i^E = \Delta F_i^M - RT \ln X_i = RT \ln \gamma_i,$$

where ΔF_i^M is the free energy of mixing, X_i the mole fraction, and γ_i the activity coefficient of component i .

Excess free energies are very difficult to measure at low concentrations, and very few methods are available. The authors² found that a variation of the Rayleigh distillation technique provides a useful method for measuring free energies at infinite dilution and have measured values for the NaI-Na system as discussed in Section 4.1.2.

The authors noted that many metallic solutions often satisfy regular solution restrictions and that, in theory, the equilibrium data on phase diagrams can be used for calculating activity coefficients for regular solutions and have been employed extensively. However, they pointed out that great practical difficulties are experienced in extrapolating to infinite dilution with the usual methods, which either assume some particular analytical equation for the variation of activity coefficient with composition, or involve a graphical integration technique employing the Gibbs-Duhem relation. Often these techniques involve a rather uncertain extrapolation to infinite dilution which can introduce large errors.

A combination of the use of the phase diagram for estimating the activity coefficients of the solutes, together with a method proposed by Tang, Castleman, and Mackay⁵ for extrapolating activity coefficients to infinite dilution, was found to be most satisfactory for many systems. A more complete description of the use of this method, particularly for the cesium-sodium systems, is given in ref. 2.

The excess free energy of mixing of cesium in sodium at infinite dilution was calculated from the cesium-sodium phase diagram by two different methods. The authors' method gave a ΔF_{Cs}^E at infinite dilution of

2050 cal/mole. This compares well with the experimental value of 1900 cal/mole for the heat of solution.⁶ Using the van Laar equation⁷ to correlate the activities obtained from the phase diagram led to a ΔF_{Cs}^E of 1920 cal/mole.

The excess free energy of rubidium in sodium at infinite dilution was calculated using the method discussed above. The calculated ΔF_{Rb}^E (1715 cal/mole) compares well with the experimental heat of solution (1750 cal/mole).⁶ A value of 1790 cal/mole was obtained for the excess free energy of mixing from the van Laar correlation method.

For the strontium-sodium system, the van Laar method was unsatisfactory, since the activity coefficients could not be represented by a two-parameter equation. However, the authors' method proved to be very satisfactory. The excess free energy calculated for this system was 2885 cal/mole.

For the barium-sodium system, the method of the authors was again quite satisfactory for obtaining the excess free energy of mixing. A value of 2450 cal/mole was obtained.

For the sodium iodide-sodium system, both the van Laar correlation method and the authors' method were unsuitable, because of a large immiscibility gap throughout nearly the entire range of composition. Using Wagner's method for immiscible systems, a ΔF_{NaI}^E of 2060 cal/mole was obtained.⁸ If Henry's law is assumed to apply for the metal-rich phase, the ΔF_{NaI}^E becomes 7820 cal/mole at 750°C. Using another method, Pitzer⁴ calculated the expression $\Delta F_{NaI}^E = 14,200 - 6T$ for this system. This gives a value of 8060 cal/mole for the excess free energy of mixing at the same temperature. Experimentally (Section 4.1.2), the authors found the value to be 9100 ± 400 cal/mole for the range 650 to 830°C. These measurements showed the excess free energy of mixing to be substantially temperature independent within the precision of the experiments. This result is to be expected, since the solution approaches regular behavior at infinite dilution.

None of the above methods was applicable to the antimony-sodium system. Therefore, the authors employed a method used by Pitzer⁴, assuming the solutions to be regular throughout the entire range of composition. The calculated ΔF_{Sb}^E was -6740 cal/mole.

For the tellurium-sodium system, none of the methods used for the other systems was applicable for calculating ΔF_{Te}^E . For this system a plot of $(RT \ln \gamma_{\text{Te}})/X_{\text{Na}}^2$ versus $X_{\text{Te}}/X_{\text{Na}}$ gave a straight line which was easily extrapolated to zero mole fraction tellurium. This amounted to correlating the data with a power series in composition. The excess free energy at infinite dilution was -4050 cal/mole.

4.2 AI: Retention of Fission Products in Sodium, Release to Blanket Gas, and Plateout

Early studies in fission product retention included an evaluation of fission product activity resulting from the SRE channel blocking¹⁰ in 1959. Following the failure of 14 elements of the 43 in the reactor, several thousand curies of fission products were released into the sodium coolant. Activity concentrations ranged from 0.74 $\mu\text{C/g}$ for ^{131}I to 13.9 $\mu\text{C/g}$ for ^{95}Zr - ^{95}Nb . Only xenon and krypton were measured in the gas space. However, considerable decay time, before gas sampling, may have prevented observation of short-lived material such as ^{131}I .

Severe cases in which up to 70% of the iodine inventory is released to the sodium have been tabulated. This compilation suggests that under these temperature and experimental conditions approximately 2×10^{-4} of the iodine trapped in sodium may be available for further release to the environment.

Experiments have also been performed at Atomics International to evaluate the extent of retention, by the sodium, of fission products released into high-temperature sodium.⁹⁻¹⁷ Sodium temperatures were 500 and 1000°F for in-pile studies and ranged from 900 to 1400°F for out-of-pile studies.¹⁰ In the in-pile studies, small disks of preirradiated uranium metal were melted and a portion of the fission product inventory was dispersed in molten sodium by a transient neutron burst. In the out-of-pile study, fission products were injected by recoil during the preirradiation and by diffusion during the heating. Data were obtained in both cases by sampling the helium cover gas space above the sodium interface and relating the fission product activity observed to that measured in the sodium and to the estimated inventory of the preirradiated fuel.

When expressed as distribution coefficients, experimental results at Atomics International for the release of iodine from an equilibrium sodium system showed some agreement with a theoretical distribution coefficient.¹³

Experimental results for cesium and strontium showed fairly logical distribution coefficients, although theoretical correlation for them was not yet available.¹³ Extensive plating and deposition on surfaces were observed. Therefore, it was suggested that fission-product retention studies should be related to the effects of configuration and plateout. From SRE studies and other work at Atomics International, carbon (and perhaps other impurities in the sodium) may be important in the depletion of fission products circulating in the sodium coolant after release from the fuel.

The results of these experiments at Atomics International disclose several facts:¹⁰

1. No one experiment has provided sufficient data to conclusively demonstrate fission product retention under all conditions. Although accomplishment of a single experiment to achieve this objective seems improbable, the high-temperature experiments have provided considerable data. The results have been consistent in indicating the extent to which fission products, especially ^{131}I , can be retained in the sodium.
2. The extent of ^{131}I retention in high temperature sodium appears to be of such magnitude in the sub-boiling case that isotopes which are normally less hazardous acquire a dominant role in determining the consequences of reactor malfunction and fission product release.
3. Although the retention mechanism may be largely chemical reaction with sodium, other fission products, uranium, or other capsule constituents, which in turn might modify volatility or other characteristics and thus decrease the tendency to release, other factors which could contribute to retention are absorption, migration, or diffusion into capsule surfaces. These factors would also prevent observation of fission products in the gas space. This same condition would prevail in a typical reactor environment with the large surface areas of reactor vessel, control rods, etc. to provide deposition areas.
4. The unexpected ^{131}I activity on the gas tubing maintained at 1000°F indicates that deposition may take place in high-temperature regions. Release as measured above the surface of the sodium plenum

may therefore be different in magnitude from the release measured through the colder portions of the sampling tubing. This condition would also prevail in a typical reactor configuration; the gas would probably be released through the minute cracks and penetrations of a leaking reactor top shield. It has been recommended¹⁰ that the iodine measured outside of the sodium volume be differentiated into "mobile" and "fixed". Mobile is defined as that portion of the isotopic activity (regardless of the chemical or physical form) retained on the gas-stream sampling devices; this portion is considered available for release to the containment. The fixed portion is that activity observed on the tubing walls at high temperature.

Studies and experiments are being conducted at Atomics International to describe the distribution of fission products and other contamination sources released to the coolant of a liquid metal fast breeder reactor (LMFBR).¹¹ These studies include the formulation of radiological hazard and radiation indices to aid in determining the most significant fission products, the evaluation of the importance of activated core materials which are subsequently corroded into the coolant stream, the classification of fission products on the basis of physical and chemical properties into analog groupings to reduce the number of isotopes which must be studied in sodium test loops (Table 4.1), and a preliminary study of the residual radiation levels which could result from fission product plating on heat exchanger surfaces, thereby limiting access for maintenance or repair.

The principal activities planned by Atomics International for the future are the initial operation of the Fission Products Behavior Loop (FPBL), which is expected to provide much valuable data on fission product behavior in stainless steel systems; the programming of the digital computer code for predicting radioisotope distributions in an LMFBR system; the use of this code to perform preliminary systems studies; and the continued surveillance and application of fission and corrosion product behavior patterns in operating reactor systems such as EBR-II, Fermi, and the SNAP 8 developmental reactor.

Table 4.1. Analogue Grouping and Expected Fate of Various Fission Products

<u>Analogue Grouping</u>	<u>Expected Fate in Stainless Steel System</u>
Cs	Volatilization or deposition as complex oxide
Ba, Sr	Deposition as oxide
Mo, Ru, Sb, Ag*	Diffusion into stainless steel
Ce, La, Nd, Y, Zr	Deposition as oxide or diffusion into stainless steel
Nb	Deposition as oxide or diffusion into stainless steel
H ³ , I, Te	Vaporization or diffusion into stainless steel

*May not be a successful analogue grouping.

4.3 LASL: Transport, Plateout, and Trapping of Long-Lived Fission Products in a Sodium Loop

Fission products released to sodium coolant from failed or deliberately vented reactor fuel elements may limit access to the primary coolant system and affect the consequences of a loss-of-coolant incident. Depending on the fission product inventory anticipated in the coolant, it may be desirable to concentrate uranium, plutonium, long-lived energetic gamma-emitting isotopes, and short-lived biologically hazardous isotopes at specific locations within the primary system.

To this end, the interaction of plutonium-based fuels with sodium, the release and distribution of fission products from irradiated fuel to sodium, and methods by which this distribution may be altered are being investigated at Los Alamos Scientific Laboratory.¹⁸⁻¹⁹ Long-lived fission products are added to a small, forced-convection sodium loop by immersing irradiated plutonium-base fuel in the sodium. Five-gram quantities of a Pu-Co-Ce alloy, irradiated to a level of 10^{19} fissions and cooled for several years, are the activity sources. During the experiments, fuel specimens are maintained in the liquid state between 435 and 600°C. Migration of activity from the fuel to removable specimens and traps within the sodium system is detected by in situ gamma-ray spectrometry and by radiochemical analyses of the adsorbers and traps after test.

In the studies at LASL, the behavior of ^{137}Cs released from a metallic fuel to hot-trapped sodium suggested that cesium is a mobile, volatile species which is weakly adsorbed on stainless-steel surfaces operating in the 300 to 600°C range. Cesium initially deposited on metal surfaces in the sodium circuit migrates, over a thousand hours or so, to surfaces in the cover gas regions. The migration appears to consist of vaporization from high-temperature sodium surfaces, diffusion through inert gas, and condensation on lower temperature stainless steel surfaces. In one instance, approximately 60% of the available ^{137}Cs had moved into the cover gas regions before the experiment was terminated. This activity can be partially removed from the gas regions by immersing graphite beds in the sodium or by temporarily flooding the gas regions with sodium. Graphite beds also scavenge cesium activity from the sodium circuit, reducing the activity on stainless steel surfaces by factors as high as 70. By flooding cover gas regions with sodium, as much as 90% of the cesium activity present in these regions can be removed.

Plutonium, ^{90}Sr , ^{155}Eu , and ^{106}Ru underwent temperature-dependent fractionation on stainless-steel and zirconium surfaces in hot-trapped sodium. Differences in species activity by factors of 20 to 50 were observed between metal specimens that had been immersed in regions of different sodium temperatures. Acid leaching is required to remove Pu, ^{90}Sr , ^{155}Eu , and ^{106}Ru from these specimens.

The ability of sodium-oxide cold traps to collect and hold long-lived fission products is also being examined at LASL.¹⁸ A full-flow cold trap operating between 155 and 200°C has been used to scavenge residual ^{137}Cs from a sodium system and to collect ^{137}Cs newly released from irradiated fuel. At an operating temperature of 155°C, the trap collected an estimated 57% of the ^{137}Cs originally contained in the fuel. However, this activity is not fixed in place. Small changes (20°C) in trap operating temperature have been followed by 10 to 20% changes in total cesium content.

The small-scale experiments at LASL correlate rather well with observations in actual reactor operation, which are treated briefly in the next section and in Chapter 8.

4.4 Retention by Sodium Coolant of Fission Products from Fuel Elements Failed During Actual Reactor Operation

In several cases, there was an opportunity to analyze the distribution of fission products in the primary system of a sodium-cooled reactor that

had been operated with failed fuel elements. The BR-5 fast reactor was intentionally operated with failed fuel elements for just such a study. In the SRE, a sodium-graphite reactor, a massive release of fission products from 13 of its 43 fuel assemblies was undetected for several days because of the complete containment provided by its shielding system. These cases are discussed more fully in Chapter 8, Sections 8.1.1 and 8.2.6.

In the BR-5 reactor, predominant fission products in the coolant were ^{137}Cs , zirconium, niobium, ^{131}I , barium, and lanthanum. At low power, 20 to 30% of the activity was from ^{137}Cs ; after shutdown, the major activity was from ^{131}I . Cold traps for sodium oxide were effective in trapping solid fuel fission fragments. The only activities released into the gas blanket during operation were isotopes of the inert gases xenon and krypton.²⁰

Similarly, in the SRE, xenon and krypton were the only activities released into the blanket gas. No ^{131}I was detected except that combined with sodium. The sodium apparently traps the bulk of the fission products released from the fuel.²¹

Measurements of the fission-product activity present in the coolant and cover gas systems of the Fermi reactor were begun immediately after the October 5, 1966 incident in an attempt to determine the amount of fuel melting that had occurred.²² This information was needed before fuel unloading operations could be begun to ensure that no safety hazard existed from possible fuel reassembly. Empirical data needed for this type of analysis were found to be woefully lacking. Therefore, the question of relating the radioactivity level to probable fuel failure was a most perplexing one and numerous assumptions were necessary. The measurements made, analysis technique, and conclusions reached are described in Chapter 8, Section 8.2.4.

In the EBR-II, the cladding of a single experimental capsule failed in 1967. The failed capsule was located and identified by blanket-gas monitoring²³, radiochemical analyses²⁴, and logical analysis of the results of each. Isotopes of cesium found in the sodium coolant indicated that the fuel in the failed capsule had been operating at or near its melting point. Isotopes of xenon were found in the blanket gas.

4.5 CEA, Cadarache: Transport and Plateout of Fission Products in an Intermediate-Scale Sodium-Argon System

Experimental studies of the transport, plateout, and filtration of fission products released, by melting irradiated uranium, into liquid sodium with an argon sweep gas are being conducted by Barthoux and Saroul²⁵⁻²⁶ at the French CEA's Centre d'Etudes Nucleaires in Cadarache. To be as realistic as possible, an intermediate-scale system was selected. The dimensions and the mechanical and thermal characteristics of the system are as close as possible to those in the Rapsodie reactor. The distributions of ^{131}I , $^{140}\text{Ba-La}$, and ^{133}Xe in liquid sodium, in wall deposits, in argon sweep gas, and in the sweep gas filtration-sampling system have been measured.

In each experiment an irradiated fuel specimen is melted by fusion in liquid sodium in a titanium crucible, which is connected to the bottom of a stainless steel tank by a vertical injection tube. The tube (which approximately simulated the path from the upper blanket of the Rapsodie core) is 40 cm long, with an inside diameter of 2.16 cm (somewhat smaller in test 4), and contains 150 cm³ of sodium (105 cm³ in test 4). The tank (the dimensions of which correspond to those of the reactor tank above the core) is 250 cm high, with an inside diameter of 40 cm, and has a volume of 240 liters. The tank contains three sampling probes (one at the inner wall, one near the center, and one between the other two) at each of six levels, and a single probe near the bottom. The sweep-gas filtration and sampling system above the tank consists of a coarse trap to condense sodium vapors, two asbestos filters and four activated charcoal filters each 1 cm thick, and either a 5-cm bed of activated charcoal at liquid-nitrogen temperature to trap noble gases or an ionization chamber.

Each test reported to date (the first series) was conducted with 120 liters of static sodium, maintained at 550°C in both the tank and the injection tube, and with the remaining volume in the tank filled and swept with argon. (The second series will be conducted with flowing sodium.) In each test, a 17-g specimen of unenriched metallic uranium (clad with aluminum), irradiated for about 40 hr in a flux of 2 to 5 x 10¹² neutrons cm⁻²sec⁻¹ and allowed to decay for 14 days, was melted and kept molten for 3 min, after which the volume above the sodium level in the tank was swept

with argon at about 12 liters/min for 30 min. At the time of each test, the approximate activities of the principal fission products in the fuel specimen (in millicuries) were: ^{131}I , 27; $^{140}\text{Ba-La}$, 60; ^{133}Xe , 55; and ^{137}Cs , 0.2.

After several preliminary tests, the distribution of fission products was measured in three main tests (tests 4, 6, and 7); other tests were also conducted. In the preliminary tests and test 4, the activities of the uranium before and after fusion indicated that about 50% of the ^{131}I and about 30% of the $^{140}\text{Ba-La}$ were released from the fuel; these release values were used in calculating the distributions in tests 4 and 6. In test 7, activity measurements indicated that 22% of the ^{131}I and 0.2% of the $^{140}\text{Ba-La}$ were released. Release of ^{133}Xe was assumed to be near 100%. It was noted that these out-of-pile experiments were not designed to, and could not, determine rates of fission product release from fuel; the measurements of release fractions were used in calculating distribution percentages throughout the system.

In test 4, 40% of the ^{131}I released from the fuel specimen was found in the liquid sodium in the injection system; 33% was found in wall deposits, which were assumed to be entirely on the walls of the injection system; and only 27% was found in the liquid sodium in the tank. Of the $^{140}\text{Ba-La}$ released, 13% was in liquid sodium in the injection system, 84.5% was in wall deposits, and only 2.5% was in liquid sodium in the tank.

After test 4, the diameter of the injection tube was increased, to increase diffusion of fission products into the tank. In the next test (test 5), however, no activity was detected in samples of liquid sodium from the tank; it was assumed that all activity was trapped in the injection system, possibly because of faster condensation of sodium vapor in the enlarged tube. Therefore, in tests 6 and 7, argon was injected (2 liters/min during fusion, then 12 liters/min for about 20 sec) into the bottom of the injection tube, just above the crucible, to create turbulence in the tube.

In test 6, 58% of the ^{131}I released was found in liquid sodium in the injection system, 35.3% in wall deposits, and only 6.7% in liquid sodium in the tank; of the $^{140}\text{Ba-La}$, 20.6% was in liquid sodium in the injection system, 76.1% was in wall deposits, and only 3.3% was in liquid sodium in the tank.

In test 7, measured retention in liquid sodium in the injection system was lower; only 20.6% of the ^{131}I was in liquid sodium in the injection system, 49.5% was in wall deposits, and 29.9% was in liquid sodium in the tank. Of the $^{140}\text{Ba-La}$, only 8.3% was in liquid sodium in the injection system, 82.9% was in wall deposits, and 8.8% was in liquid sodium in the tank. Furthermore, the wall deposits, which in the preceding tests had been assumed to be on the walls of the injection system, were found in test 7 to be on the walls of both the injection system and the tank and on stainless steel coupons submerged in the sodium in the tank during test 7.

In tests 6 and 7, in which argon was injected into the bottom of the injection tube, the distributions of ^{131}I and $^{140}\text{Ba-La}$ within the tank itself were practically homogeneous. In test 4, with no argon injection at the bottom, ^{131}I concentration decreased with increase in sodium level, and $^{140}\text{Ba-La}$ at an appreciable concentration was found only in the bottom of the tank. Even in test 7, where the concentrations in the tank were homogeneous, ^{131}I concentrations in the crucible and in the injection tube were respectively 7700 and 330 times as high as that in the tank, and $^{140}\text{Ba-La}$ concentrations were respectively 9000 and 490 times as high as that in the tank.

After each test, the tank was drained (with the sodium at 250°C in test 7) and then decontaminated with alcohol, 5% HNO_3 , and rinse water, successively. In test 6, alcohol removed 530 μCi of ^{131}I and 12.5 μCi of $^{140}\text{Ba-La}$; 5% HNO_3 removed 2×10^{-4} μCi of ^{131}I and 5.2 μCi of $^{140}\text{Ba-La}$; water removed 2×10^{-4} μCi of ^{131}I and 10^{-4} μCi of $^{140}\text{Ba-La}$.

In test 7, 99% of the ^{131}I in the wall deposits was removed by alcohol, indicating that about 99% of the ^{131}I plated out was in the sodium deposit itself. The quantity of ^{131}I plated out was independent of the thickness of the sodium deposit, which led the investigators to believe that the ^{131}I was essentially in a thin sodium film. The concentration of ^{131}I in a sodium deposit averaging 0.6 mm in thickness was about 300 times the concentration in the liquid sodium in the tank.

In test 6, 25 g of sodium vapor condensed in the vapor trap; this sodium contained 8×10^{-3} μCi of ^{131}I per gram of sodium. The ratio of this concentration to the ^{131}I concentration in the main body of liquid sodium (5.5×10^{-3} $\mu\text{Ci/g}$) was about 0.16, showing significant entrainment

of ^{131}I by vapor. Dividing the total amount of ^{131}I in the vapor (2×10^{-2} μCi) by the total volume of sweep gas (about 360 liters) gave a concentration of 5.5×10^{-5} $\mu\text{Ci/liter}$ in the argon; the ratio of this concentration to that in the main body of sodium (5.5 $\mu\text{Ci/liter}$) was 10^{-5} , which was taken as the ^{131}I transmission coefficient. In test 7, this coefficient was 2×10^{-5} (^{131}I concentrations were 7.5×10^{-4} $\mu\text{Ci/liter}$ in gas and 35 $\mu\text{Ci/liter}$ in the main body of sodium). In a test with sodium at 560°C , the ^{131}I transmission coefficient was 10^{-4} .

In test 6, about 2.6×10^{-3} μCi of ^{131}I was found on the first asbestos filter, just behind the vapor trap; none was detected on the other asbestos filter or the charcoal filters.

In test 4, about 15% of the total amount of ^{133}Xe originally in the uranium specimen was found in the cold trap in the argon sampling system. In test 7, about 10% of the ^{133}Xe originally in the specimen was detected in the argon by an ionization chamber. In a test in which ^{85}Kr was injected directly into the sodium, 20% was detected in the argon by an ionization chamber. Note that these noble gas releases were from more or less static sodium.

The authors remarked that only ^{131}I and noble gases were found in the argon sweep gas, and that the ^{131}I was all in the sodium vapor, undoubtedly as NaI . From this they concluded that small leaks in the blanket gas system of a reactor would release only noble gases; the iodine would be trapped in condensates of sodium vapor.

The lack of sodium circulation was considered largely responsible for the high retention of fission products in the injection system and for the high retention of noble gases in the sodium. (A second series of tests will be conducted with flowing sodium.) On the other hand, the high retention in the injection system and the very high degree of plateout on metallic surfaces indicated that fission product retention in sodium is strongly influenced by system configuration. In the case of the noble gases, the roles of solubility and compound formation in liquid sodium were considered to need much additional study. With regard to the iodine distribution, it was noted that transmission of iodine into the argon by sodium vapor is related to the iodine concentration in the liquid sodium at the sodium-argon interface.

The low fission product concentrations in these experiments (0.035 μCi of ^{131}I per gram of liquid sodium in the tank in test 7, for example) were considered important; Saroul²⁶ stated that, from the work of Castleman, Tang, and Mackay², fission product concentration strongly affects the transmission, by sodium vapor, of fission products from solution in liquid sodium.

Barthoux and Saroul²⁵ also commented that only a large-scale experiment could realistically simulate the fuel assembly, the sodium flow rates, and the amount of sodium vapor produced in a real reactor accident, and that the results of these intermediate-scale experiments could not be extrapolated to a real reactor accident. On the other hand, these experiments were considered well suited to studies of parameters such as sodium temperature, wall temperature, and the concentrations of fission products and impurities, which would be very difficult to study in a large vessel.

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5.0 SODIUM FIRES IN REACTOR CONTAINMENT ATMOSPHERES*

Since the introduction of sodium-cooled reactors with the resultant large-scale use of sodium, there has been a continuing interest in the problem of sodium fires in reactor installations and attendant particulate matter release. Sodium fires can be divided into combustion of a sodium pool and combustion of a sodium spray. A special case of the spray fire is one in which sodium is introduced explosively into an oxidizing atmosphere.

Of major concern in pool fires involving a reactor coolant are the transport of sodium activity in the smoke from the fire and, if conditions responsible for the fire have also caused failure of the fuel cladding, fission product transport.

Early in the history of sodium-cooled reactors, a continuing series of research programs investigating effects of sodium fires was begun. In association with the Naval Reactors Program, Mine Safety Appliances Company¹ and Knolls Atomic Power Laboratory² conducted tests to determine techniques for suppressing fires in enclosed spaces following sodium spray leakage. These tests generally examined the effects of reducing oxygen concentration as a means of controlling fire intensity. To provide design data for the Fermi reactor system, Atomic Power Development Associates³ and the Detroit Edison Company^{4,5} ran a series of tests in a 20-ft³ chamber to evaluate the effects of sodium injection on containment vessel pressure. Measurements were made of chamber pressure and the rate of pressure rise as a function of initial oxygen concentration. The effect of spray dispersion was also investigated. In connection with EBR-II hazards evaluation, Argonne National Laboratory carried out explosive injection tests in a 70-ft³ chamber.⁶ These tests produced the most severe pressure surges that have been noted experimentally. Argonne National Laboratory recently conducted a foreshortened program⁷ using very small-scale explosive injections. This work was started in support of the FARET reactor and was not carried beyond a few preliminary tests. British work has generally been of a fundamental nature, with no engineering-scale tests having been reported in support of their liquid-metal-cooled reactor program.

*A more comprehensive treatment of this subject is contained in "Sodium Burning and Aerosol Release - An Evaluation of the State of the Art," by R. E. MacPherson (ORNL-TM-1937, May 1968).

By far the major work on sodium fires is being carried out by Atomics International. This work was started in support of the SRE, continued throughout the Hallam program, and is supported on a continuing basis as part of their program on sodium-cooled reactor technology. Work has been done on both small- and large-scale sodium fires to develop information on burning rates⁸⁻¹⁰, sodium oxide and fission product release rates¹⁰⁻¹³, and the characterization of aerosols released from fires^{11-12, 14}.

5.1 Spray Fires

Spray fire problems are compounded by the high reaction rate that results from intimate contact between the two reactants. The rate of energy release in a spray fire is a function of reactant temperatures, spray energy or degree of dispersion, humidity, oxygen concentration, and shape of the gas volume. Spray fires that involve the explosive introduction of sodium into a surrounding atmosphere are of particular concern because the extremes of temperature and pressure would be expected. There is experimental evidence^{6, 15-16} that pressures approaching the theoretical maximum are possible under these conditions. Recently, interest has developed in the possibility of shock wave formation due to the rapid combustion of sodium vapor.¹⁷ Magnification of pressure pulses imposed on containment vessels by this mechanism must be considered. Droher and Gracie have presented an excellent summary⁸ of spray fire research up to 1960.

5.1.1 Ignition of Sodium Sprays

In spray fires, the primary variables affecting ignition are oxygen content of the atmosphere and droplet size distribution. Cowen and Vickers¹⁸ conducted experiments in which small jets of sodium were forced from holes 8 to 20 mils in diameter into an air atmosphere. The moisture content of the air atmosphere was varied, and it was generally found that ignition occurred even as low as 250°F irrespective of moisture content in the test environment. Other tests using moist and dry nitrogen resulted in no ignition.

Droher and Gracie reported⁸ on tests in which 1000°F sodium was sprayed into oxygen-depleted air. Most of the tests were conducted at a moisture content of about 0.04 wt. %. Some smoke was produced at 0.5 vol. %

oxygen, and incandescence of sodium was noted at 5 vol. % oxygen. When the test was repeated at a moisture content of 3 wt. %, 210 grains per lb of air, no incandescence was noted in an oxygen concentration of 4 vol. %. Droher and Gracie also indicated that very fine particles of sodium (from condensed vapor) had been observed to ignite at room temperature in the air, while drop-size particles had been seen to ignite just above the melting point.

In tests conducted by Nelson and Nelson², sodium was injected into a controlled-atmosphere chamber. No ignition was noted when the oxygen level was 10 vol. % with the sodium at 828°F. However, toward the end of the injection sparking was noted, probably due to a decrease in the droplet size as the flow accelerated and/or injection gas breakthrough occurred. At a very low injection pressure (approximately 1 psi), 1000°F sodium was injected in 7.5 vol. % oxygen, and no ignition occurred throughout the test. At higher injection pressures, sparking was noted at this oxygen concentration. At an oxygen level of 5 vol. %, no sparks were noted even at high injection pressures.

These tests tend to confirm the generally accepted design criteria that sodium plumbing in a reactor system can be maintained in vaults having an oxygen concentration equal to or less than 5 vol. % as a means of avoiding serious consequences from system leakage. As an example, the SEFOR hazards evaluation¹⁹ uses this basic premise in considering sodium fires. The equipment cells are open to the atmosphere only when the sodium in the primary system has been cooled below 350°F. The containment vessel is designed to accommodate the sodium burning that would result from a failure of the seal between the equipment cells and the main containment vessel. Such a failure would result in a burning rate estimated at 5 lb/hr/ft². It would produce a peak pressure of 30 psig in the containment vessel, 6 min after a pool filling the entire basement area of the equipment cells began to burn. No case of a spray fire was considered because of the protective atmosphere enclosing the coolant system.

As indicated above, it is conventional practice to install sodium circuits in cells with controlled atmospheres. Rupture of a sodium-containing system can, in the worst case, produce a sodium pool fire having less severe and more predictable time-dependent pressure and temperature

characteristics. Tests of spray fires into air and controlled atmospheres have generally defined this requirement, but no systematic study of the spray fire has been made to date.

5.1.2 Effect of Type of Release on Severity of Spray Fires

Available information on spray fires can be summarized briefly. A leak in a sodium system that produces a jet or spray of sodium into an air atmosphere will undoubtedly produce ignition followed by a sodium fire and fume problems, regardless of the air humidity or the sodium temperature. The type and intensity of the fire will be a function of the size of the leak, the surface area of the burning sodium, and the availability of oxygen to sustain combustion.

If the sodium system is enclosed and the atmosphere surrounding the system is controlled at an oxygen concentration of 4 to 5%, ignition of a sodium spray is suppressed, and the possibility of fire must be assigned to secondary effects such as a resulting breach of the containment and inleakage of air. This containment provision has been used commonly in sodium-cooled reactors. It should be realized that even with inert atmospheres or controlled atmospheres, a pressure increase in the containment space can result from spray leaks of sodium due to heat transferred to the gas and/or reaction with the amount of oxygen that may be present. In the latter case, sodium oxide fumes have also been noted below the ignition point.⁸

Sodium spraying into enclosed spaces containing oxygen at concentrations above the level known to suppress ignition will produce fires accompanied by pressure and temperature transients that are a function of the size of the leak, the oxygen supply, the burning rate, and the heat transfer characteristics of the system. Experiments indicate that essentially theoretical adiabatic yields of pressure and temperature can be attained in some instances where sodium is explosively injected into an entire enclosure. Actual cases of interest probably are less serious. It is difficult to visualize a real case in which an explosive injection efficiently fills an entire containment volume. Compartmentalization and less efficient mixing cause the pressure and temperature transients to be less severe, because larger volume is available to absorb the transient generated in the reactive volume. The possibility exists that shock waves generated by the expansion of the reactive volume

into the unaffected gas space in the containment vessel could produce wall loadings on the vessel in excess of those which would be created by simple temperature and pressure equilibration of the reactive zone with the remainder of the containment complex.

In the case of the more likely spray fire, one in which sodium is issuing at a discrete rate into air or an atmosphere capable of supporting combustion, pressure measurements have ranged from very low values up to the maximum reported, 38 psig.⁵ Such results are expected, because the basic mechanism is a time-dependent equilibration between a combustion process supplying heat and various heat transfer processes distributing it. Only one attempt to describe the situation analytically has been reported, and reasonable agreement with experimental results is shown in that case.⁵ However, the model is quite basic, and further refinement appears justified.

5.2 Pool Fires

The burning of sodium from a molten pool and the accompanying release of energy, sodium oxide fumes, and fission products are of primary concern to safety studies of sodium-cooled reactor systems.

5.2.1 Effects of Reactor Design and Type of Release on Pool Fires

A recent consideration²⁰ of design concepts for sodium-cooled reactors indicated three general system arrangements to be of interest:

1. The primary sodium system and reactor core are in one large sodium-filled vessel that is penetrated by the secondary coolant lines. The large vessel is installed in a shielded cell that normally contains inert gas. An inert gas space occupies the top of the vessel, and a shield plug separates the primary sodium from the containment shell enclosing the entire system.
2. The primary system components are interconnected in a conventional piping arrangement and installed in the containment structure with necessary shielding.
3. The individual primary system components are installed in separate sealed cells, which serve as shielding and primary containment. Interconnecting piping runs in sealed vaults. A secondary containment covers the entire system. The primary containment can be filled with inert gas.

Based on these reactor containment arrangements, a series of possible accident types may be formulated:¹⁸

(a) Leakage of Sodium into a Cell Containing an Inert Atmosphere. Due to the lack of oxygen in the cell, the major considerations are temperature and pressure transients caused by heating of the gas in the cell. The rate of introduction of the sodium, the degree of dispersion, and the uniformity of mixing with the cell atmosphere will determine the severity of the transient. If the leak were accompanied by a breach in the cell containment, air could enter and a sodium pool fire could result.

(b) Leakage of Sodium into a Cell Containing an Oxygen-Bearing Atmosphere. A more severe pressure and temperature transient would be possible in the case where an air atmosphere is present in the cell. The primary variables determining the course of the transient would be the same as in (a) above, augmented by chemical reaction effects. Exhaustion of oxygen would limit the extent of the accident, but again violation of the containment could lead to a sustained sodium pool fire due to air entry.

(c) Direct Release of Sodium into the Containment Building Air Atmosphere. A reactor using the second type of containment discussed above would have the capability of introducing sodium directly into the air atmosphere of the main containment enclosure. Such accidents were also considered in the safety evaluations for the EBR-II²¹ and FARET⁷ reactors, although the former uses the first arrangement and the latter proposed to use the third. Again, both initial injection conditions and resultant accumulation and burning of sodium pools must be considered in evaluating the consequences of the accident.

5.2.2 Sodium Oxide and Fission Product Release

A major consideration in evaluating the hazards of pool fires is the release of activated sodium and fission products by sodium fires. In most of the sodium burning tests conducted to date by Atomics International, an attempt has been made to determine sodium oxide release fractions. Many of the tests have also used a radioactive tracer in the sodium to get a feel for the fission product release problem.

Nelson, Baurmash, and Koontz¹¹ reported sodium burning experiments in a 40-ft³ chamber, using small sodium batches. In three experiments, release fractions of oxidized sodium ranged from 0.25 to 0.54. Burning of the sodium charge was incomplete in at least two tests.

Lauben¹⁰ has reported a series of 11 small-scale tests in which sodium and iodine release fractions were measured. The initial sodium mass varied from 0.5 to 6 g, and initial sodium temperatures were from 800 to 1100°F. Material balances on the sodium were good, ranging from 91 to 110%, with seven runs in the range 95 to 105%. Iodine material balances ranged from 66 to 116%, with six runs in the 90 to 110% range. The maximum figure for airborne sodium was 19% of the original, with seven runs showing 7% or less. The maximum figure for airborne iodine was 10% of the original, with seven runs showing 3.4% or less.

In one of the 11 runs, the iodine content of the metallic sodium was determined before and after the fire. A decrease from 360 to 12 ppm was noted, indicating the iodine had been selectively removed from the sodium, probably by sorption on sodium oxide. In a supporting experiment, a sodium-sodium iodide mixture was passed through a sodium oxide bed supported on a stainless steel filter. The iodide content of the sodium was reduced essentially to zero.

Lauben¹⁰ also reported on oxide and iodine release measurements from a 615-lb fire (LF-1). These figures were higher than was generally true in the small fires, being 27% of the total for sodium and 8% of the total for iodine.

Koontz, Nelson, and Baurmash conducted some preliminary iodine release experiments in a small laboratory device designed for the generation of aerosols with controlled and predictable size characteristics. Sodium containing sodium iodide was evaporated at 1000°F into a nitrogen stream that was mixed with a second, larger stream of nitrogen. It was determined that the I/Na ratios in the liquid and vapor phase were equal.¹²

Lauben and Larson¹³, in a continuing series of tests in the modified large fires apparatus (LF-2), have reported iodine release results showing a temperature dependence. With a burning pool surface temperature of 1300°F, the released sodium contained iodine in one-half the concentration of the iodine in the sodium pool. With a surface temperature of 1000°F, the released-phase concentration was about one-fifth the concentration in the pool.

Although measurements of release fractions of sodium oxide from pool fires have been inconsistent, there are strong indications that the majority

of the oxide formed remains behind in the fire residue. Recent measurements of release fractions have ranged from very low values up to 30 or 40%. Iodine release fraction measurements are generally inconclusive at present.

An examination of the pool fires program at Atomics International reveals a growing sophistication in equipment and experimental techniques for the investigation of sodium fires. Koontz et al.²⁰ have itemized their theoretical and experimental program in this area, and this work is well under way. Bench-scale equipment is available for measurement of release rates of sodium, sodium oxide, and fission products and for determination of released-particle size distribution. A laboratory test chamber is in use for the characterization of aerosols released from fires and for the determination of activity, mass, and particle size distribution as a function of time after a controlled sodium fire. Suitable instrumentation for accurate determination of test results is being developed as needed.

Recent work at Atomics International is discussed in several progress reports.²²⁻²⁴

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6.0 FISSION PRODUCT TRANSPORT AND SODIUM OXIDE AEROSOLS

The gaseous fission products released into the containment system may be divided into two components, reactive and nonreactive. Reactive gases include hydrogen and tritium, oxygen, oxides of nitrogen, halogen gases or vapors (such as iodine and bromine), and their daughter products. Non-reactive gases are the noble gases, such as krypton, xenon, and possibly nitrous oxide (which while not non-reactive, in fact, may behave as though it were).

The most important elements to be considered in the gaseous release problem are the nonreactive gases, for three reasons: first, they form an important fraction of all fission products; second, they will be affected by free diffusion only; and third, they cannot be easily removed from the gaseous phase. Only by free diffusion and convection are they dispersed into the containment or mixed during flow of the gases. Radioactive decay, together with containment in a closed volume or dilution and discharge, or both, is the only really effective means now available for handling the noble gases.

Reactive gases, on the other hand, may not only change their state in passing into the confinement or containment system, but they may also change in concentration markedly while in passage. Consequently, it is necessary to know whether the reactive gases and vapors are in elemental or molecular states, what oxidation states may be available to them, and what chemical compounds are likely to be formed. The halogen gases, iodine for example, are notorious in this regard inasmuch as they exist in various states of oxidation as inorganic compounds. Free halogens can also be sorbed readily by both inert and various reactive surfaces. They react rapidly with most metals and, for instance, sealing plastics and rubber. They can combine with vapors or may be sorbed by aerosols produced from volatilized metals or their compounds. They also form organic halides which are difficult to remove from the effluent stream by the usual methods. Sodium metal vapor if released will oxidize, and the oxides may become hydrated from moisture within the reactor atmosphere. The resultant oxide or alkali hydroxide will behave differently from the original element in its reaction with halogen fission products, thus further complicating the picture for sodium-cooled reactors.

In most cases, after a serious accident the fission products escaping from the fuel encounter more than one environment, often successively. For liquid metal systems, a dispersion of a liquid metal may be disseminated rapidly in the air or inert atmosphere surrounding the pressure vessel. If this environment is air, oxidation takes place, whereas if it is an inert gas, a vapor system and a condensation aerosol will result. Thus it is apparent that the state and characteristics of the environment when the fission products are dispersed into containment may have a significant influence on the behavior of fission products.

In the case of liquid metal coolants, the reactions expected will depend on the kind of atmosphere into which the coolant is released. In some cases, because of the severity of the metal-air reactions, it may be necessary to maintain an inert atmosphere in the containment. This will prevent oxidation reactions, but the condensation growth situation will be of importance. In effect, only the kind of condensing vapor will vary, and in some instances the vapors may already have reacted with many fission products.

6.1 Aerosol Production and Transport

The fission products are likely to be present in the sodium as dissolved metals; exceptions are the rare gases, which escape unreacted, and iodine, which would be predominantly in the form of sodium iodide and to a smaller extent cesium iodide and iodides of the other highly electropositive metals. The concentrations of iodides other than sodium iodide depend on the temperature and the concentrations of the reactive metals in the sodium. Upon exposure to air, fission products at the surface will be oxidized if the surface temperatures are sufficiently high.

With rapidly burning sodium, oxygen concentrations at the surface will probably be low, diffusion of fission products to the surface of the sodium will be slow (this will be a strong function of the convective currents in the sodium), and further oxidation could cease. Important questions arise regarding the details of the fire and the behavior of the reaction products, since the sinking of oxide particles in the sodium could result in formation of fission product oxides which tend to have very low volatilities. On the other hand, failure of fission products to oxidize at the surface because the gas is depleted in oxygen could result in

substantial volatilization of fission products as metals. This is very dependent on diffusion of fission products to the surface (the extended time-scale of the burning process and likelihood of liquid convection are favorable to good diffusion). These metals may form oxides as they arise through the air-burning region (assuming that the burning process occurs predominantly in a gas-phase reaction zone). If such volatilization and oxidation occurs, then the fission product oxides may not behave differently from the sodium oxide which is formed, and may subsequently agglomerate, fall out or plate out. To the extent that oxidation of fission products in the liquid sodium occurs, the retention of these fission products by the sodium can increase.

Iodine is an exception to this. Its release could occur by vaporization of the iodide, by dissociation of NaI, CsI, or other metallic salt, or by entrainment. The entrainment mechanism must also be considered for the release of sodium and dissolved fission products which adhere to particles carried up by convective air currents. Sodium and fission products could also become trapped in the oxide formations which adhere to the walls as a result of the burning process, with the possible preferential absorption of fission products.

Release of a given fission product, regardless of the form or the mechanism for its release, will be different only if its method of transport is due to its separation from the sodium aerosols. For those whose activity is accounted for by the sodium aerosols, no matter what the burning temperature or release mechanism, it can be inferred that their transport is the same as that of sodium.

Since there may be mechanisms which reverse the process of agglomeration after settling or plating, some series of tests must be designed to establish whether such reversals take place.

6.2 Sodium Aerosol Research

In a program at Atomics International to characterize the release and transport of effluents and energy generated during sodium fires¹ one major phase is concerned with the production of aerosols from sodium fires and with the transport and control of this particulate matter inside reactor containment enclosures. The results of this program to date have been summarized in the four papers which are briefly described here.

Lauben and Larson² discuss the theoretical aspects of the sodium-oxygen reaction region heat and mass transfer. Pertinent experimental results presented have been mentioned previously (see Chapter 5, Section 5.2.2).

Koontz, Nelson, and Baurmash³ discuss a laboratory device being used to generate reproducible sodium aerosols in gas media of known composition for standardization of aerosol sizing devices. The device permits vaporization of sodium into a hot nitrogen stream and subsequent reaction and/or condensation by mixing with a second nitrogen stream having a controlled oxygen concentration. Experiments were carried out to determine system operating conditions that would produce useful particle concentrations in a size distribution typical of steady-state sodium fires in low-oxygen environments. Size distribution was monitored with an electrostatic precipitator and with modified impactors. The sodium aerosol generating device will be extremely useful in subsequent fundamental investigations of aerosols.

Nelson, Baurmash, and Koontz⁴ discuss the laboratory equipment being used in their aerosol studies and some of the early results. A 40-ft³ chamber is available in which 2- to 6-in. burn pots can be installed. Instrumentation is available for monitoring temperatures throughout the volume, the oxygen content of the contained gas, and the aerosol plate-out at various heights and radial positions within the chamber. Fallout on the floor is similarly measured, and visual observation of the experiment is possible. Gas samples can be taken at various points to monitor particle size.

Test results presented show good agreement in the relative rates of fallout on the floor and plate-out on the walls for three sodium batches, one of which was considerably larger than the other two. Measurements at three different heights of particle concentration and amounts of deposition were the same, indicating that stirred settling was in progress. Size distribution measurements taken on samples from the chamber have not produced good agreements between the electrostatic precipitator and the modified impactors, unlike the case in work with the standard aerosol generator described two paragraphs above. This problem must be eliminated before work on fission product aerosol characterization can proceed.

Spiegler et. al.⁵ have developed an analytical treatment (discussed further in Chapter 7) of the processes involved in aerosol generation, coagulation, and settling. The results from computer solutions of the pertinent equations are being compared with experimental results from the tests discussed above.

The course of the program on aerosol production, transport, and control is closely allied to the test program on sodium pool fires. The end objective is to develop an analytical model that completely characterizes sodium fires and aerosols produced by these fires. As indicated in Chapter 5 (Section 5.2.2), rapid progress is being made at Atomic International in developing definitive experimental techniques for the controlled investigation of sodium fires. As the techniques, both analytical and experimental, being developed in the aerosol program are refined and proved, they can be applied to the large fires apparatus and eventually to the large-scale modeling facility.

6.3 Aerosols of Plutonium, Sodium, and Alloys

Safety analysis of sodium-cooled, plutonium-fueled, fast reactor plants must be concerned with the possibility of fires involving plutonium as well as sodium. Design of an air cleaning system for such a facility requires basic data defining the aerosol characteristics of sodium and plutonium released during a fire.

6.3.1 Fractional Release and Size of Aerosols from Oxidation of Large Pieces of Plutonium Metal

A study was conducted by Mishima⁶ of Battelle-Northwest to obtain information on the amount and nature of the particulate oxide released during the oxidation of large pieces of metallic plutonium at relatively high air velocities. Four large metallic plutonium specimens, ranging in weight from about 450 to 1770 g, were ignited. Ambient air was drawn over the ignited metal at ~ 525 cm/sec, the metal was allowed to oxidize completely, and the particulate oxide entrained in the air stream was collected. Recorded temperatures from thermocouples embedded in the metal ranged from 500 to 1000°C during the oxidation. From 2.9×10^{-4} to 0.049 wt. % of the plutonium was released during the sampling periods (from 22 to 90 min) giving release rates ranging from 0.032 to 0.0045%/hr for the bare metal and a release rate of 0.00029 wt. %/hr for a specimen

covered with MgO sand soon after ignition. The amount of particulate oxide released in these runs appears to depend on the phase of the starting metal and the physical conditions during the oxidation.

The sizes of the particles airborne under these conditions is surprisingly small. The air velocity used (525 cm/sec) is theoretically capable of supporting a particle over 200 microns in diameter of the density of PuO_2 . The largest particle found on the electron microscope was approximately 8 microns in diameter. Undoubtedly some particles between 8 and 200 microns in diameter were formed; both Mishima⁷ and Stewart⁸ have reported their presence in the residue from the air oxidation of metallic plutonium. Either the particles of this size range are present in the mass of oxide and trapped by the surface or the particles reported previously are formed during some subsequent period.

As mentioned above, the amount of fine particulate oxide airborne during the oxidation of metallic plutonium above ignition temperatures in air at a velocity of 525 cm/sec ranged from 0.032 to 0.0045 wt. %/hr. In these experiments, the amount released by delta stabilized metal was lower than from the alpha-phase metal. The mass median diameter of the particles airborne was 4.2 microns, with no particles greater than 8 microns detected. Immersing the ignited metal in MgO sand can reduce the amount released by an order of magnitude.

It is of interest to note that all four runs showed temperature "spikes" of the type reported by Schnizlein and Fisher⁹. The samples for this study were obtained by Mishima in cooperation with the Plutonium Chemistry Section of Isochem, Inc., who performed the ignitions.¹⁰ An earlier review¹¹ by Mishima had noted the need for further study.

6.3.2 Characteristics of Aerosols from Sodium, Plutonium, and Plutonium Alloys

Size characteristics of the aerosols produced during sodium and plutonium fires were determined by Ettinger, Moss, and Busey¹² for atmospheres ranging from 20.8% oxygen, 79.2% nitrogen to 100% nitrogen. Gram quantities of metallic sodium, plutonium-cobalt-cerium alloy, and alpha- and delta-phase plutonium metal were burned separately. Samples were collected on electron microscope grids; electron micrographs were made and were sized on the basis of equivalent area (projected diameter).

Count median diameter (c.m.d.) and geometric standard deviation (σ_g) were determined graphically from log-probability plots of the data. Mass median diameter (m.m.d.) was calculated using the Hatch and Choate equation¹³:

$$\log \text{m.m.d.} = \log \text{c.m.d.} + 6.9 \log^2 \sigma_g.$$

The aerosol produced by burning gram quantities of sodium was compared with that produced by a fire involving 600 lb of sodium. Data were obtained regarding the fraction of plutonium alloy airborne during a plutonium fire. Fires simulating a reactor accident involving both fuel and coolant defined the relative airborne concentrations of plutonium and sodium.

The aerosol produced by burning gram quantities of sodium in atmospheres of various oxygen concentrations and humidities had the following size characteristics: c.m.d., 0.07 to 0.91 μ ; σ_g , 1.68 to 3.25; m.m.d., 0.90 to 14.3 μ . Under small-scale burning conditions sodium aerosol size was not related to oxygen content, but showed some dependence on humidity. Sodium aerosol characteristics under large-scale (600 lb) burning conditions at various low oxygen concentrations were: c.m.d., 0.20 to 1.09 μ ; σ_g , 1.53 to 2.82; m.m.d., 0.53 to 16.17 μ . Under large-scale burning conditions, sodium aerosol size was not related to oxygen concentration. Comparison of these data showed that sodium aerosols produced from small- and large-scale fires have comparable size characteristics, and that aerosol parameters can be determined for any special conditions without building a large burning system.

The aerosol collected directly above burning chips of plutonium-cobalt-cerium alloy (57.7% plutonium) showed the following size characteristics: c.m.d., 0.04 to 0.09 μ ; σ_g , 1.24 to 1.54; m.m.d., 0.05 to 0.14 μ . The aerosol collected directly above burning metallic plutonium turnings (alpha and delta phase) showed the following size characteristics: c.m.d., 0.02 to 0.06 μ ; σ_g , 1.24 to 1.76; m.m.d., 0.03 to 0.13 μ . Oxygen concentration of the burning atmosphere did not affect the particle size of the aerosol produced from burning plutonium metal or alloy.

When plutonium-cobalt-cerium alloy was burned, the fraction airborne ranged from 1.8×10^{-7} to 38.8×10^{-7} , depending on burn temperature and oxygen concentration.

Fires involving both plutonium alloy and sodium produced airborne particulates with a plutonium-sodium ratio ranging from 0.34 to less than 0.008%. The high initial ratio was rapidly reduced as the molten sodium blanketed the release of plutonium.

6.4 Effects of Plateout, Type of Containment, and Containment Leak Rate on Postaccident Dose Rates

Hafele, Heller, and Schikarski¹⁴ investigated the interrelationship between assumptions of release of radioactive materials from fuel, decontamination of the containment atmosphere by plateout or artificial means, type of containment system, leakages of inner and outer containments, and radiation burden released to the environment. Instantaneous release of radioactive materials from the fuel in a 1000-Mwe sodium-cooled fast breeder was assumed to result from fuel melting and vaporization caused by a fast nuclear excursion. ICRP recommendations were followed in dose calculations. Plutonium doses to bone and iodine doses to thyroid at 500 meters downwind were calculated. For simplification, noble gases were not considered.

In these investigations, a pessimistic set of release fractions (Set A), 50% of the halogens, 50% of the volatile solids, and 10% of the solids was considered because, as remarked by the authors¹⁴, such a set was being discussed and considered in various groups related to the USAEC's Division of Reactor Licensing. A conventional set (Set B), 10% of the halogens, 50% of the volatile solids, and 1% of the solids, was based on the values of DiNunno *et al.*¹⁵ with a reduction in halogen release by a factor of 5 because of the trapping capability of sodium; this set referred to the then current state of the licensing procedure for the SEFOR reactor and was applied in the safety analysis of the Karlsruhe Na-2 prototype reference design.

Two optimistic sets of "release fractions" were also considered by Hafele, Heller, and Schikarski. These sets, called "aerosol models", were based on estimates of the amounts of solid radioactive material that could remain airborne within the containment atmosphere after an accident,

rather than estimates of the total amounts released from the fuel. Set C, 10% of the halogens, 50% of the volatile solids, and 0.01% of the solids, was mentioned to have been used in the first SEFOR safety analysis, but later refused by the USAEC because of insufficient knowledge of the behavior of aerosols. The authors¹⁴ commented that the verification of this set is the goal of an aerosol program at Karlsruhe. Set D, 1% of the halogens, 10% of the volatile solids, and 0.01% of the solids, was a modification of set C that was believed possible when more experimental data became available.

Under the parameters used in the calculations, doses based on Set A or Set B and a single containment with a leak rate of 0.5% of the contained volume per day were far in excess of acceptable doses. Based on Set A, double containment was inadequate; for a double containment with an outer containment leak rate of 0.5%/day, Set B gave satisfactory results provided the inner containment leak rate was not over about 10%/day. For a double containment with the outer containment vented to the outside atmosphere through a 75-meter stack (with no filter system), Set B gave reasonable dose rates for an inner containment leak rate of 0.5%/day provided the off-gas flow rate from the outer containment did not exceed about 100%/day.

The somewhat optimistic Set C, based on low release fractions for plutonium aerosols, gave acceptable dose rates for a double containment with an outer containment leak rate of 0.5%/day even when the inner containment leak rate was 50%/day. Only the optimistic Set D, making full use of assumed data for aerosols of plutonium and other isotopes, gave dose rates for the single containment with 0.5%/day leak rate that were close to permissible values.

Hafele, Heller, and Schikarski concluded that the aerosol models (Sets C and D) are believed to provide much more realistic data for solid materials and to describe much better the activity transport after large accidents in sodium-cooled fast breeder reactors. They further concluded that, in view of the forthcoming era of 1000-Mwe fast breeder reactors, there is a strong, well founded incentive to put great emphasis on the investigation of aerosol behavior in the context of fast reactor accidents.

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7.0 SAMPLING, CHARACTERIZATION, AND FILTRATION OF SODIUM-FISSION PRODUCT AEROSOLS

In a sodium-cooled fast breeder reactor, sodium-24 (from neutron activation), plutonium-bearing fuel materials, and associated fission products which may be present during accidents involving release of primary coolant are a potential radiological safety problem. Noble gas fission products are not hazardous in the sodium spill accident if they have been removed from the coolant during normal operation. In a core using vented fuel elements or having some failed elements, volatile fission products such as iodine may enter the coolant and be available for release from a sodium spill.

Several AEC-sponsored projects have investigated the release of fission products into sodium coolant^{1,2} (see also Sections 4.1 and 6.2 of this report). Investigations¹ have shown that iodine combines chemically with the coolant to form sodium iodide. The distribution of volatile fission products such as iodine and cesium between liquid sodium and an inert cover gas is under study.³ Detailed characterization of the sodium aerosols and iodine released from a sodium fire is being investigated⁴ under the overall sodium fires program at Atomics International.

Some of the major phases of research and development in the sampling, characterization, and filtration of aerosols of sodium oxide (and sodium metal under certain conditions) and associated fission products are discussed in this chapter.

7.1 Sampling Sodium and Fission-Product Aerosols

Techniques for sampling aerosols which contain foreign contaminants of low allowable concentrations, such as fission products and fissile material, make sizing particle distributions by electron microscopy of samples collected by electrostatic precipitation an inadequate tool. Additionally, techniques which characterize an aerosol at only one time and place will not describe it completely, unless the particle concentration and sizes are so small that no further change in the distribution will occur.⁵ A number of empirical approaches have been used at Atomics International to characterize the matter released from sodium in an environment of nitrogen with a low oxygen and humidity content.

Sampling techniques described by Nelson, Baurmash, and Koontz⁶ enable the characterization of the aerosols produced from sodium fires in a controlled-environment test chamber. By using plating and fallout deposition samplers combined with cascade impactors, electrostatic precipitators, and millipore air samplers, studies were made of the spatial and temporal behavior during the production, dispersion growth, and removal of the suspended particulates. The role of each sampling technique in verifying analytical models was discussed.

Samplers used in these experiments can be divided into two types: deposition samplers and aerosol samplers. Deposition samplers have been built to automatically collect samples of wall plating, ceiling plating, and floor deposition. High concentrations $> > 1 \mu\text{g}/\text{cm}^3$ are easily measured. Aerosol samplers are used manually to collect spot samples for particle size and concentration measurements.

Aerosol samplers which are used only to obtain concentration data are prewashed membrane filters, with $0.2\text{-}\mu$ pore size, and three midget impingers (in series), using water as the collecting medium.

Cascade impactors are used to collect samples for particle size measurements and to obtain concentration data. A 7-stage, 400-jet Anderson⁷ impactor (modified), 5-jet and 10-jet Andersen impactors, and a single-jet, 7-stage impactor based on the design of Mercer⁸ were used. [Nelson, Koontz, and Baurmash later reported¹⁷ that round jet impactors tend to break up agglomerates and are not recommended for use in obtaining distribution data.]

The authors⁶ presented a summary of four experiments performed in the laboratory test chamber.

Automatic deposition samplers were used to measure the mass of material deposited in fixed time intervals during a sodium oxide aerosol experiment in the laboratory test chamber. Figure 7.1 shows the relative mass accumulated on the floor and walls as a function of time.

In Tests 2 and 3, the air concentration vs time and the total deposition were measured at three different heights. The concentrations and amounts deposited were the same at these heights, thus verifying that stirred settling was occurring.

It was also noted that about 34% of the oxidized sodium left the pot and escaped into the air: this was therefore the effective fraction released.

The experiment designated as Test 3 in Fig. 7.1 was the largest (pounds of sodium consumed and area of pot) that has been attempted in the laboratory test chamber. As such, it allowed observation of a large change in the initial oxygen concentration over a long time period.

Fair agreement between size distributions obtained with impactors and with an electrostatic precipitator have been obtained with a particle generator.⁴ However, a typical experiment in the laboratory test chamber leaves much to be desired as far as agreement between the two techniques is concerned. The importance of agreement has to do with the ultimate objective of characterizing the sizes of the associated fission products which are released during sodium fires; electrostatic precipitators were considered unsuitable, and round jet impactors broke up agglomerates. Figure 7.2 shows the median aerodynamic diameters obtained from two different detectors as a function of time for Test 2. The other experiments give similar results. Computed median aerodynamic diameters from count median diameters give a similar trend, i.e., an increase in size followed by a decrease as time proceeds.

By using a variety of techniques for sampling, information can be obtained to characterize the aerosols released from sodium fires. The results of these experiments will be used to develop and validate analytical models which predict the following relationships:

1. The dependence of distribution, particle size, and concentration on the source term and time.
2. The effects of concentration on settling, wall plating, and particle growth rate.
3. The importance of wall surface area and floor area on the suspended particle concentration.

7.2 Characterization of Source Terms for Aerosol Transport

The production of radioactive aerosols as a result of an accidental spill of sodium coolant from a nuclear reactor has been studied by Lauben and Larson.⁹ The vaporization and combustion of sodium were discussed, and results of a test program designed to measure burning rate and particulate

Fig. 7.1 Plating and Fallout Data - Laboratory Test Chamber (from ref. 6).

Fig. 7.2 Median Aerodynamic Diameter Variation with Time for Test 2 (from ref. 6).

release rate were presented. The buoyant forces in the gas provide a mechanism for transport of particulate from the scene of the spill, and an analysis of these forces was given.

Vaporization from the liquid sodium pool, which would result from a postulated primary coolant spill in a sodium-cooled nuclear power plant, provides a source term for aerosol transport. The vapor may react in the gas phase or simply condense forming aerosols of sodium and sodium oxides, and possibly fission products in combination with or separate from the sodium product aerosols. The energy for vaporization and gas-aerosol transport is supplied by the gas-phase sodium combustion reaction and condensation, by possible heterogeneous surface oxidation, and from the initial heat content of the liquid sodium.

The low flame of the sodium pool fire tends to create a region of high-temperature gas plus combustion products just above the sodium. The presence of cooler, heavier gas well above the flame establishes buoyancy forces which remove the hot gases from the flame by updrafts while cooler gases flow down toward the flame.

The concept of buoyancy forces creating significant gas velocities near the flame can be extended to a hypothetical sodium spill in an open reactor primary sodium cell. Typical geometry for the system under consideration is shown in Fig. 7.3. In such a fire the cell floor would be covered with sodium, which would burn with the air in V_1 . After an initial rapid transient in which all gas flow was away from the fire, buoyancy forces would create a "counter-flow" of gases in the cell access opening. Thus, building gases would replenish oxygen in the cell and promote the burning. A digital computer code, SOFIRE, has been developed at Atomics International for analyzing this problem. The code takes into account heat transfer rates to the cell floor and walls and to building walls. It allows for building gas inleakage and exhaust, and can be used for constant pressure or for containment pressurization fires.

In a SOFIRE calculation of fire conditions in a fast breeder reactor plant, the gas velocities in the cell opening were of the order of 15 ft/sec upward and 12 ft/sec downward. For the geometry considered, the cell floor area was 288 ft², while the access opening was 72 ft². These velocities were high enough that the O₂:N₂ ratios in V_1 and V_2 were within 3% throughout the fire.

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Fig. 7.3 Model for a Sodium Fire in a Primary System Cell (V_1) Open to a Reactor Building (from ref. 9).

The influence of the above gas velocities on aerosol transport during a sodium fire has not been analyzed. It is clear, however, that gas movement will be rapid enough to allow an appreciable fraction of airborne sodium oxide to be carried from a fire in an open cell to the room above the cell.

7.3 Formation and Agglomeration of ZrO_2 , PuO_2 , and UO_2 Aerosols

Studies dealing with the formation and agglomeration of aerosols produced upon the high temperature vaporization of nuclear fuels were initiated by Castleman et al.¹⁰ As a prelude to studies with plutonium fuels, an investigation of the vaporization of ZrO_2 and UO_2 into a flowing helium stream and its subsequent condensation to an aerosol in an aging chamber was undertaken.

ZrO_2 samples, heated to a temperature slightly in excess of 3000°C in about 45 sec, were vaporized into a flowing helium stream and transported to an aging chamber. Within 10 min, at a calculated initial concentration of 10^6 particles per cubic centimeter of gas, the particles showed significant agglomeration. The Aitken particles within the aging chamber were sampled with a modified Watson-type thermal precipitator. Several electron micrographs obtained from the grids showed small spherical particles, measuring 50 to 250 Å. The particles were agglomerates into long twisting chains ranging from 0.05 to 0.3 micron in length.

Particles of similar size were also noted in electron photomicrographs of the UO_2 aerosol deposit. No significant change in the 200-Å spheres and their agglomeration into long twisting chains, which approach 1 micron in size, was noted upon varying the vaporization and coagulation times. Additional tests by Castleman et al. are in progress to determine the important parameters which affect particle size.

The coagulation of aerosols produced by the vaporization of fast reactor oxide fuel materials and sodium is being studied at Atomics International¹⁷ and Brookhaven National Laboratory.¹⁸ Both PuO_2 and UO_2 agglomerated rapidly into chains, many of which were 1 to 2 microns long. Sodium oxide intertwined chains of UO_2 ; in one experiment a combination of UO_2 and Na_2O coagulated so rapidly that no UO_2 was found in the gas phase less than 1 hr after vaporization. Coagulation constants were determined experimentally in the work at BNL.

7.4 Coagulation of an Aerosol

An IBM 7094 digital computer was used by Spiegler et al.⁵ in solving the equation that describes in time the behavior of an aerosol. The equation was derived by Muller^{11, 12}, based on earlier work by Smolukowski¹³, and was later refined by Whytlaw-Gray and Patterson¹⁴, Goldman¹⁵, and others.

The work by Spiegler et al. resulted from the desire to formulate better the dispersal of radioactivity from a large sodium fire such as could result from accidents in fast nuclear reactors. Containment buildings for these plants will need carefully designed filtering systems, the specifications for which will in part be determined by the behavior of sodium oxide aerosols. The topics investigated include coagulation of a heterogeneous aerosol, coagulation coupled with stirred settling, and an external source that continuously generates new particles. In some instances, approximations to the code results may be obtained, provided simplifying assumptions are made.

The results indicated that the aerosol concentration is a more important parameter than the initial particle spectrum. One of the striking facts to emerge from even the limited number of problems worked by the code is the rapid disappearance of small particles, providing the initial concentration is high enough. Another observation of the cases studied is that the mechanism of a source term (i.e., time interval during which the source exists) is unimportant, provided the times of interest in a study are much longer than the duration of the source term.

Spiegler et al. found indications that the initial value of σ , the width of the log-normal distribution, is important in determining such matters as the time for 50% of the weight to settle, and the change in mean radius of the suspended component as a function of time. Since these parameters may have an important bearing on the performance of filters, future experiments will attempt to make accurate measurements of σ or analogous quantities if a log-normal distribution does not exist. Another parameter of importance will be the total mass density present; this quantity will be proportional to the radioactivity present when sodium is in the form of ^{24}Na .

Code calculations are now being performed which include wall plating as well as settling and agglomeration. When obtained, these results will be compared with experimental findings for a relatively small closed chamber (volume $\sim 40 \text{ ft}^3$).

7.5 Sodium Aerosol Filtration

In the event of a coolant loop rupture in a sodium-cooled reactor, sodium may be vaporized and burned, forming a dense fume of metal oxide in the containment vessel atmosphere. To prevent vessel rupture and leakage to the atmosphere, release of overpressure must occur with retention of particulate matter and radioiodine.

In a study by Himot et al.¹⁶ various configurations of graded deep-bed filters were tested to evaluate their application for controlling such fumes and to determine their maximum filter loading capacity. Minimum filter requirements are 99% removal of particles and iodine from an exhaust gas stream containing up to 30 lb of sodium without exceeding a resistance of 20 in. H_2O across the entire filter at a maximum exhaust flow rate up to $1000 \text{ ft}^3/\text{min}$ and a maximum temperature of 250°F . Oxygen concentration may reach 2%. Under these conditions, the fume may be liquid sodium (m.p. 203°F) or solid; when solid, it may be condensed sodium metal or sodium oxide. Sodium metal retained on the filter may change physical state as filtration proceeds and may oxidize if oxygen enters the filter container.

Tests were performed with the equipment shown in Fig. 7.4; the major components were a sodium melting and vaporization vessel, a heating system, a blower to circulate the carrier gas, and a filter holder. Controls were installed to regulate carrier gas flow rate and temperature of the molten sodium. Ports were placed at numerous locations for temperature and differential pressure measurements, aerosol sampling, and the addition and measurement of oxygen in the nitrogen carrier gas.

Table 7.1 shows the physical properties of filter media used in this study. Fiber diameter was determined microscopically with a calibrated Porton graticule. Porosity was computed from the weight of a known filter volume.

A deep bed filter pack constructed of 2-in.-deep layers of stainless steel fibers 1000, 500, 250, 125 and 60 microns in diameter packed $4\text{-}5 \text{ lb}/\text{ft}^3$,

plus two 2-in. layers of 30- and 15-micron glass fibers packed 15 lb/ft³, plus a pleated absolute filter, was shown to be able to collect and store 0.75 lb of sodium and sodium oxide fume per square foot of filter face area. Filter resistance reached 20 in. H₂O when operated at 10 fpm face velocity. Reductions in filter face velocity extend filter life and provide low resistance with high fume storage capacity. The efficiency of these filter packs is > 99% for all particles. No iodine, as elemental iodine or sodium iodide, penetrated the filter pack when the sodium melt contained 3.5 ppm iodine as sodium iodide.

Table 7.1. Filter Media

Medium	Average Fiber Diameter (μ)	Fiber Diameter Range (μ)	Bulk Density (lb/ft ³)	Porosity (% voids)	Supplier
Stainless mesh	1000	115-1000	15-30	94-98	a
No. 3 Steel wool	95	40-200	2-8	98.4-99.7	b
Rollmat, coarse fiber	85	20-250	0.5-2.0	98.7-99.6	c
Stainless wool, coarse	76	30-130	2-8	98.4-99.7	b
Stainless, wool, medium	62	20-90	2-8	98.4-99.7	b
Dynel (24 denier)	49		0.9	98.8	d
No. 2 Steel wool	47	23-91	2-8	98.4-99.7	b
U-Trim-It	40		2.2	98.6	e
Amerglass	32		0.25-7.0	98.5-99.8	e
Rollmat, fine fiber	28	16-65	0.4-2.0	98.5-99.7	c
K115, curled glass	27	23-32	1.5-6.0	96-99	e
No. 1 Steel wool	26	16-65	2-8	98.4-99.7	b
PF334	7	2-14	0.5-2.0	98.5-99.6	e
PFO18	7		0.46	99.7	e
FMO11	2.5		0.43	99.7	e
MSA, type B	2.5	1.2-10	10	93.5	f
AAF 50	1.5		0.45	99.7	c
FM 004	1.5		0.47	99.7	e
PF105	1.2		0.31	99.8	e
Absolute filter paper 1106D	< 1				f
Pleated absolute filter	< 1				g

- a. Volks Limited.
- b. American Steel Wool Manufacturing Company, Inc.
- c. American Air Filter Company, Inc.
- d. Union Carbide Corporation.
- e. Owens-Corning Fiberglas Corporation.
- f. Mine Safety Appliances Company.
- g. Flanders Filters, Inc.

Fig. 7.4 Sodium Fume Test Apparatus (from ref. 16).

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8.0 EXPERIENCE WITH REACTOR DESIGNS, FUEL MELTDOWNS, AND FISSION PRODUCT RELEASE

One of the promising approaches to economical nuclear power is the liquid-metal-cooled, high-temperature, non-pressurized, reactor. A high coolant temperature (500 to 650°C) is possible without pressurization of the reactor heat extraction system. This permits good steam conditions and high thermal efficiency in the power conversion equipment.

The use of a liquid metal such as sodium as a heat transfer fluid has many advantages. Sodium melts at 97.8°C and is therefore easy to keep liquid in a power plant system; it boils at 882°C and thus permits adequately high coolant temperatures with the system at atmospheric pressure. Because liquid metals, sodium in particular, have extremely high thermal conductivity, they are excellent heat transfer media. The fluid dynamics of liquid sodium is similar to that of water. A further advantage of sodium is the general absence of chemical corrosion in metal systems; this enables the use of a wide variety of steels and other alloys as construction materials.

Sodium has certain disadvantages. It reacts chemically with oxygen and water and must therefore be isolated from them. Further, exposure of sodium to neutrons, as in a reactor, produces ^{24}Na , a strong gamma emitter with a half-life of about 15 hr, and this creates a serious problem in nuclear power plant applications. For these reasons (in the present conceptions) the steam boiler has been separated from the primary sodium coolant, which extracts the reactor heat, by a secondary sodium coolant system, which does not become radioactive - and carries the heat from the primary coolant system to the boiler. The vault necessary to contain the radioactive primary sodium provides the shielding required and also acts as a container for the inert gas atmosphere which surrounds the primary system.

The emphasis in this chapter is on sodium-cooled fast reactors with either ^{235}U or ^{239}Pu as the primary fissionable isotope and with stainless steel as the major material for all primary components and coolant systems. Examples of sodium-cooled graphite-moderated reactors and of NaK-cooled reactors are included because their design and operating history is pertinent to the operational safety of fast reactors.

No definite design pattern was established by the first group of fast reactors (EBR-I and -II, Dounreay, and Fermi), nor was there any fixed pattern

of maximum credible accident. The chance of complete coolant loss varied from very unlikely to essentially impossible. A loss of most of the coolant flow upon a loss of primary electric power was a possibility in two designs, but each had a small battery-operated backup motor or pump and each provided upflow that aided natural convection. Provisions for removal of afterheat varied appreciably.

According to the EBR-II and Enrico Fermi hazard reports, no "credible" accident leads to large-scale core meltdown; nevertheless, the two reactor-design groups postulated such accidents, assumed that reassembly occurred, calculated the reactivity rate and energy yield pessimistically, and provided primary blast-containment structures and missile protection within the outer containment shell.

The high-power high-burnup fast power reactors contemplated for the future present some new safety considerations. The very much larger radio-activity inventory is an obvious hazard. If a credible accident were capable of releasing the bulk of these fission products into the reactor building, the previously used single containment shell might not prove adequate for many sites. Most methods developed for reducing the building pressure and fission-product concentration in the containment building following a bad accident in water-cooled reactors are probably not directly applicable, and a new approach to engineered safeguards may be required.

Removal of the large fission-product after-heat could be difficult if the primary and secondary systems were disrupted in the accident. Special provisions might be required to prevent or limit an accident (e.g., a sodium-water reaction) that could disrupt the entire secondary system unless an alternate means of afterheat removal were available.

A large amount of fissionable material representing several critical masses if reassembled at full density would be present in the core. The guided dispersal of such fuel into a subcritical configuration in the event of a slow meltdown accident and the maintenance of an appropriate amount of coolant on this material may prove to be a difficult problem if such recourse is necessary.

A basic understanding of the phenomena involved and a detailed application of this knowledge to the course of physically realistic accidents are required to properly assess the real state of affairs concerning the possibility

of explosive damage from postulated nuclear accidents in large fast reactors. One small fast reactor core did melt during an unusual experiment with no untoward safety consequences.¹ With appropriate design it may be possible to ensure continuation of such good fortune in the unlikely event of serious malfunction.

8.1 Sodium-Graphite Reactors

8.1.1 Sodium Reactor Experiment (SRE)

The Sodium Reactor Experiment (SRE) is a sodium-cooled graphite-moderated reactor system² that operates with slightly enriched fuel at a thermal power level of 20 Mw. It was originally designed solely as a reactor experiment, but a small turbine-generator set and a steam generator were added.

The reactor structure is unique in that the required containment is immediately adjacent to the core, and no external sphere or pressure-containment building is provided. This design is based primarily on the following three considerations:

1. The coolant is essentially at atmospheric pressure, and thus it represents negligible stored mechanical energy.
2. The fuel, moderator, and coolant are chemically compatible, and thus there is no potential for release of chemical energy.
3. The system has a large thermal capacity that is available for absorbing any conceivable unplanned release of nuclear energy.

The stainless-steel core tank, which contains the moderator and fuel elements, is surrounded by a second containment vessel, which has dimensions such that, in the event of a leak in the reactor vessel, the sodium coolant will be retained and the level of the sodium in the reactor pool will not drop below the level of the outlet nozzles. An inert-gas atmosphere is maintained in the second containment vessel, and therefore sodium circulation can be continued even in the event of failure of the core tank. Should both these vessels fail, tertiary containment is provided by a carbon steel "cavity liner", which serves to support thermal insulation on its inner surface. The space between the secondary container and the cavity liner is such that a rupture of both the two inner vessels will not drop the sodium pool level below the top of the core. Thus adequate decay heat cooling of the fuel elements would be available in the unlikely event that both inner containers ruptured. Leak detectors are provided to indicate failures of the two inner tanks.

No activity transport could be measured during the first two years of reactor operation, and during that time sodium was circulated in the system for approximately 16,000 hr, 4500 hr of which was at a high temperature and a large temperature differential. During this two-year period, two fuel elements were found to have pinhole leaks, and there was some release of gaseous fission products, detected by the presence of ^{133}Xe in the reactor helium atmosphere.

A massive release of fission products throughout the primary system occurred in July 1959^{3,4}. Thirteen of the 43 fuel assemblies sustained damage sufficient to cause failure of the cladding on all seven rods of the assembly and to form at least some iron-uranium eutectic that was molten for a short period of time in the reactor. Measurements of cover-gas activity extrapolated to the time of the fuel-element damage indicated that approximately 10,000 curies of mixed fission-product activity remained undetected for some days because of the complete containment afforded by the SRE shielding system. This occurrence, which caused some fuel-element temperatures to rise abnormally high, at least to 1430°F, came about during a period of reactor operation in the 2- to 3-Mw (thermal) range.

Analysis of the helium cover gas shortly after discovery of the activity release showed that only the noble gases were released. Xenon and krypton were identified as the sole contributors to cover-gas activity. No ^{131}I has been detected except that combined with sodium. Removal of the irradiated fuel from the reactor reduced the cover-gas radioactivity to near normal background, despite the presence of fission products in the sodium. Disposal of piping and components replaced during the recovery period has not proved to be a severe problem. The normal methods used for cleaning components containing uncontaminated sodium were applied, with the additional precaution of retaining the sodium-steam reaction products which apparently contained all the fission products not deposited on pipe walls. Fission products remaining on pipe walls created radiation levels of only a few milliroentgens per hour, and these components could be disposed of as low-level wastes by conventional methods. The reaction products were disposed of as low-level liquid waste by diluting and mixing with concrete.

Operation of the SRE for three and one-half years, including a period of recovery from extensive fuel damage, has demonstrated the following:

1. The reactor system is both stable and responsive to power changes.
2. Maintenance and repair of sodium systems uncontaminated by fission products is a routine procedure.
3. Release of fission products to the primary system of a sodium-graphite reactor does not result in hazards to operating personnel or to the general public when the SRE containment philosophy is employed; the sodium apparently traps the bulk of fission products released from the fuel.
4. Detection of fission-product release by monitoring the cover gas is effective.
5. Repair of contaminated sodium reactor systems is feasible.
6. Prediction of sodium-graphite reactor behavior, both in normal and abnormal situations, has been confirmed.
7. Special containment measures, other than those currently employed, are unnecessary.
8. The sodium-graphite reactor concept has been demonstrated, by operation of the SRE, to be technically feasible.
9. The SRE is conservatively designed with respect to power capability.

8.1.2 Hallam Nuclear Power Facility (HNPF)

The operating experience of the Hallam Nuclear Power Facility (HNPF) demonstrated the safe, simple operation of a sodium-cooled graphite-moderated reactor.⁵ Full-power operation of this system was first achieved in July 1963, with steam supplied to a 100-Mw turbine generator. Failures of equipment, including intermediate heat exchangers and sodium pumps, resulted in interruptions in power generation for limited durations; almost all equipment failures encountered were of the type common in the startup of any large, complex industrial plant. Eventual failures of a moderator-element cladding resulted in the current facility shutdown. Notwithstanding these operational difficulties, operation of the HNPF has proven the inherent safety features of the low-pressure liquid-metal coolant with its high retention of fission products, the high thermal efficiency attainable with high-pressure high-temperature superheated steam, and the ease of control and simplicity of operation and maintenance.

8.2 Fast Reactors

8.2.1 Dounreay Fast Reactor (DFR)

The Dounreay Fast Reactor⁶⁻¹⁰ was the first liquid-metal-cooled reactor in the British program. This discussion is in terms of the first core and does not cover later fuel element designs or modifications made for its use as a test reactor. The primary system was designed to have a large number of independent circuits using components of the modest capability then available. Powering each pair of the 24 primary pumps with diesels rendered any sudden loss of coolant flow essentially incredible unless there was a complete loss of coolant through a major leak. The primary NaK coolant system was designed to operate at low pressure and to be essentially double-walled so that such a major leak was unlikely. However, the high power density and low heat capacity of the core made the probability of fuel-element meltdown very large should the core become uncovered. Hence a special annular fuel element was designed, one leading to the sequential failure of the cladding upon loss of coolant so that the uranium in each fuel element would run down the central flow passage, now devoid of coolant. A catch pot was placed below the lower blanket to divert such molten fuel into a subcritical configuration. Downflow of the coolant through the core was chosen partly to allow the coolant to assist the removal of molten fuel from the core in the event that such failure occurred while NaK was still flowing.

A large steel containment shell surrounds the reactor and primary system. The shell was designed to contain any fission products, to withstand a NaK fire, and to dissipate after-heat should the primary and secondary systems be disrupted.

8.2.2 Experimental Breeder Reactor I (EBR-I)

The EBR-I consisted essentially of an unmoderated reactor coupled to a heat sink.¹¹ The heat sink consisted of (1) a primary NaK coolant circuit that removes heat from the reactor, (2) a secondary NaK circuit that removed heat from the primary NaK, (3) a superheater, boiler, steam turbine, condenser, and economizer (water) circuit that removed heat from the secondary NaK circuit, and (4) an electric generator driven by the turbine. The primary NaK flow was from an elevated constant-level tank into the reactor, through the heat exchanger, and down to a receiving tank. From the

receiving tank it was continuously pumped up to the constant-level tank, which contained an overflow to the receiving tank. Further details of the heat-sink system are described by Zinn¹² and Trocki et al.¹³

8.2.3 Experimental Breeder Reactor II (EBR-II)

In the EBR-II^{6, 14-17}, the entire low-pressure primary system is submerged in sodium, which, in turn, is contained in a large double-walled tank. The tank sits in a close-fitting concrete cavity so that even failure of both steel walls of the tank would not lead to uncovering of the core. Hence loss of coolant from the core by leakage is deemed incredible by the EBR-II designers.

The two externally powered primary-sodium-system pumps are backed up by a small battery-driven electromagnetic pump that is always available in the primary system for an emergency. With flow normally in the upward direction, natural circulation would quickly set in upon loss of primary flow, and only minor temperature rises would be expected if a reactor scram occurred properly. Should the secondary loop be inactivated, shutdown heat could be removed from the primary system with natural circulation and conduction to the atmosphere. The large body of sodium acts like a huge heat sink and helps to minimize shutdown-cooling problems. The submerged design and the coolant-inlet features also make impossible any sudden change in coolant temperature or sudden introduction of large quantities of moderator with their attendant reactivity effects.

The EBR-II was provided with a double containment system. The primary system includes the biological shield and the contents of a large tank in which the entire primary system is submerged. Its function is to contain the effects of a nuclear energy release equivalent to the detonation of 300 lb of TNT without breaching. Missile shielding is included above the reactor. The steel secondary containment building is designed to withstand a major sodium fire and to contain any fission gases that might escape from the primary system.

Results of studies of the pressures that might be generated by sodium fires are reported in the EBR-II hazards report.¹⁵ Oxidation from a stagnant pool should be slow, and many hours would be needed for all the oxygen in the building to be reacted. Sodium release as a pressurized spray would lead to much faster reaction rates. A maximum pressure of 38 psig was measured

in some tests involving the spraying of sodium into a 532-liter vessel.¹⁸ The EBR-II containment system is designed for this type of fire.

The third and potentially the most severe fire would occur if sodium were explosively ejected into the building atmosphere, as might conceivably occur from an uncontained nuclear excursion. A series of experiments¹⁹ involving explosive ejection gave a maximum instantaneous pressure of 80 psig. It is difficult, however, to extrapolate the experimental conditions directly to EBR-II since (1) not all the oxygen is directly available, and (2) the surface available for heat loss is relatively less. Similar pressures are conceivable though; so protection against such an eventuality is provided by the primary containment system, which is designed to prevent any such expulsion in the unlikely event of a serious nuclear accident.

Experiments on fuel-element failure mechanisms in TREAT have shown that the sodium bond in the EBR-II element can rapidly expel molten metallic fuel at the point or points of cladding failure.²⁰ If large numbers of fuel elements failed simultaneously at the same axial position, this expulsion could provide a mechanism for partial concentration of fissile material and hence the rapid addition of reactivity. This mechanism was not considered in the preliminary hazards report. Subsequent detailed analysis of the coherence of failure, i.e., the relative times at which fuel pins in different parts of the reactor reach the failure temperature, has indicated spreading in the time of fuel release sufficient to keep this mechanism no worse than those previously hypothesized.²¹

8.2.4 Enrico Fermi Reactor

The entire sodium system of the Enrico Fermi Reactor^{6, 22-24} is surrounded by a secondary containment system to prevent loss of coolant. Around the reactor vessel and part of the primary piping, this containment is provided by the primary shield tank. The balance of the piping and the intermediate heat exchangers and pumps have essentially double-walled construction. In case of a failure within the primary shield tank, however, the sodium level within the reactor could fall to a point below the critical level needed to ensure natural circulation were it not for a special reserve supply of sodium that was provided to maintain a proper sodium level in this event.

As a backup to the main motors driving the three primary pumps, pony motors that are powered by an emergency bus fed by either a storage battery or an emergency diesel generator are supplied. Simulator studies indicate that these pony motors are sufficient to prevent core melting in the event of primary motor failure, even if no scram rods are inserted into the reactor.

The Enrico Fermi hazard report²⁴ states that the only credible accidents that could lead to fuel-element melting would have to be caused by subassembly plugging or fuel swelling and that only one subassembly should be involved at a time. However, "a hypothetical meltdown accident" involving the entire core was analyzed. The postulated causes were (1) flow failure without scram and without backup-pony-motor operation or (2) loss of sodium from the primary system.

In either case it was assumed that the sodium would leave the core and that the fuel would melt and collapse under the force of gravity. As the fuel collected into a dense configuration, reactivity would be gained, leading to a burst in power, the vaporization of some of the fuel, and violent disassembly. A total energy release in the vicinity of 7×10^8 cal was calculated with pessimistic assumptions. Only part of this energy is available to do work and produce damage; thus the accident was estimated to be equivalent in its explosive effects to about 500 lb of TNT, although the energy release rate may be different. The primary containment structure, which includes the primary system, the primary shield, and the secondary shield, was designed to absorb blast effects from an energy release of 1000 lb of TNT, except for the possible missile action of the rotating shield plug. The provision of a machinery dome to restrain the rotating plug completed the blast protection. It also would prevent large-scale expulsion of sodium from below the floor, where a nitrogen atmosphere is maintained, to above the floor, where large amounts of sodium in highly dispersed form could cause a substantial fire and significant pressure generation within the outer steel containment structure. The latter actually is designed to withstand a major sodium fire, as was the case for EBR-II, but the designers deem it incredible. After-heat removal would be required in the event of the hypothetical accident, and this would be achieved through the non-insulated steel walls of the outer containment structure.

8.2.4.1 Anomalous Reactivity Effects in the Fermi Accident

The available records were analyzed²⁵ to determine the time history of the anomalous reactivity effects which occurred during the incident at the Fermi reactor on October 5, 1966. Three prime records were used: the power history; the shim and regulating rod history; and the history of the sodium inlet temperature on the secondary side of the intermediate heat exchanger. The power history was integrated through the neutron kinetics equations to obtain the net reactivity as a function of time. It was also integrated with the feedback transfer function through the convolution integral to obtain the feedback reactivity. The shim and regulating rod histories were translated into reactivity using the proper calibration curves. Finally the reactivity effects due to coolant inlet temperature changes caused by changes in the steam plant were taken into account. The difference between the net reactivity and the sum of the three describable effects was designated as the anomalous reactivity. The first appearance of anomalous reactivity occurred about 21 min before fission products were released in sizeable amounts, and probably can be attributed to overheating of the core.

8.2.4.2 Results of Fission-Product Activity Analysis to Determine Extent of Fuel Failure in Fermi Accident

Measurements of the fission-product activity present in the coolant and cover gas systems of the Fermi reactor were begun immediately after the October 5, 1966 incident in an attempt to determine the amount of fuel melting that had occurred. This information was needed before fuel unloading operations could be begun to ensure that no safety hazard existed from possible fuel resassembly. Empirical data needed for this type of analysis were found to be woefully lacking. Therefore, the question of relating the radioactivity level to probable fuel failure was a most perplexing one and assumptions were necessary. The measurements made, analysis technique, and conclusions reached have been described by Mueller, Beaudry, and Feldes.²⁶

To estimate the extent of fuel failure from the argon cover gas and sodium coolant activities, samples of each were taken at various locations in the reactor and measured for gross beta activity (Fig. 8.1) and activity of the principal isotopes present (^{137}Cs , ^{89}Sr , ^{131}I in sodium; ^{85}Kr , ^{133}Xe in argon). From these data the total activity present was determined and this was adjusted for estimated losses due to solubility, plate-out and fractional fission-product release. These adjusted activities were then

compared to the calculated activity buildup in the core prior to the incident to determine the amount of fuel meltdown.

The consensus of all analyses²⁶ was that a fuel failure equivalent to the complete meltdown of approximately two core subassemblies (2% of the core) had occurred. This result is in good agreement with the reactivity loss observed at the time of the incident. The following observations were also made:

1. Noble gas release appeared to be larger than obtained from SRE meltdown analyses²⁷ and various experiments²⁸ (50% vs 10% or less).
2. Particulate release minus plateout losses for the first sodium sample taken also appeared to be larger than expected (~ 10%).
3. Considerable plateout in the primary system occurred (Fig. 8.1). Less than 1% of the original fission products remained in the sodium at the time of reporting.
4. Contrary to expectations, measurements indicated very little plateout in the heat exchangers. Cold trapping had not removed much of the activity at the time of reporting.
5. Although solubility data for noble gases in sodium vary greatly^{29,30} conservative calculations show their retention in sodium under accident conditions is small (< 20%).
6. Measurements show nonuniform distribution of the released fission product activity in both the argon and sodium systems (Fig. 8.1).
7. Sodium analysis appears less reliable than cover-gas analysis. For best accuracy, sodium samples should be taken as soon as possible after melting; gas analysis should not be attempted until short-lived activities have died out.

8.2.5 Los Alamos Molten Plutonium Reactor Experiment I (LAMPRE-I)

LAMPRE I³¹ is a small fast reactor that operates at powers up to 1 Mw (thermal). The reactor is used primarily for testing materials involved in the development of molten plutonium fuel elements. In present LAMPRE I cores, 23 kg of molten Pu-Fe fuel is contained in about 140 sodium-cooled tantalum capsules, each approximately 0.4 in. O.D. The entire active core region is approximately 6 in. in diameter by 6 in. high. Space is provided at the top of the fuel elements for the accumulation of fission product gases.

Fig. 8.1 Total Sodium and Cover Gas Activity After Incident at Fermi Reactor (from ref. 26).

Power operation with the first (Mark I) core began in April 1961. The results of operation of this core have been reported elsewhere.³² Operations with the second (Mark II) core were completed in June 1963. The Mark II core was operated at powers up to 1 Mw (thermal) for a total of 105 Mwd ($\sim 1/2\%$ depletion), during which two capsule failures occurred. Out-of-pile experiments indicate that failures of the Mark II fuel elements can be expected when the tantalum temperature exceeds 650°C ($\pm 25^{\circ}\text{C}$). This temperature is consistent with the observed incidence of failures in the Mark II core. Even though the two failures resulted in the escape of ~ 75 and 30g of plutonium into the sodium coolant, no operational difficulties were experienced.

Reactivity measurements during operation indicate that almost all of the fission gas accumulates in the 2-in. gas space above the fuel in the capsule. Post-irradiation measurements of the amount of gas in the gas space above the fuel also confirm the release of fission gas from the body of the fuel.

The Mark III core includes a test of a new fuel alloy, Pu-Co-Ce, and of its compatibility with tantalum. In the Pu-Co-Ce fuel, the plutonium content may be varied from 0 to 13g of plutonium per cubic centimeter without significantly altering the melting point of $\sim 450^{\circ}\text{C}$. Preliminary tests indicate that this alloy (at least for plutonium contents below 8 g/cm^3) may be significantly better with respect to compatibility with Ta containment than is the Pu-Fe alloy.³³

From a safety point of view, it may first be noted that there is no meltdown problem, in the usual sense. The principal accident potential lies in a major loss of capsule integrity. LAMPRE I experience has indicated that minor capsule failures are not a hazard. Obvious precautions are indicated to prevent nuclear accidents in case of a major loss of capsule integrity by avoiding large undiluted regions in which the plutonium alloy could assemble. All accident analyses to date indicate that the thermal expansion of the molten plutonium is adequate to prevent a major nuclear excursion.

8.2.6 BR-5 Fast Reactor (USSR)

The fast reactor BR-5³⁴ was designed for 2% burnup; some of the mixed oxide (PuO_2 and UO_2) fuel had reached 5.8% burnup at the time of reporting.

This reactor³⁵ was deliberately operated with failed fuel elements to study the release of solid and gaseous fission products into the coolant and gas blanket of the primary circuit. Study of the sodium coolant radioactivity showed predominant effects to be due to ^{137}Cs , zirconium, niobium, ^{131}I , barium, lanthanum. At low power levels, 20 to 30% of the contamination activity was from ^{137}Cs . After shutdown, the activity observed was mainly from ^{131}I ; most of the ^{137}Cs had deposited on the walls of the coolant system (and in cold traps) when electric heating of the coolant system was shut off. Cold traps for sodium oxide were also effective in catching solid fuel fission fragments.

The experiments proved that only inert gases (xenon and krypton isotopes) were released into the gas blanket during reactor operation. Iodine was not detected.

The BR-5 fuel element cladding showed considerable evidence of embrittlement following burnups to 5.8%. The cladding had reached an nvt of 3.5×10^{22} at elevated temperature (approximately 500 to 600°C). The fuel had increased in volume approximately 1 to 1.5% per percent burnup.

Leakage from fuel elements was shown not to have resulted in any appreciable difficulties with the equipment.

In view of the future use of uranium monocarbide fuel elements in BR-5, some of the new fuel material has been loaded into the reactor for study.

8.2.7 Rapsodie and Phenix Reactors (France)

The Phenix reactor will be a 250-Mwe sodium-cooled prototype reactor loaded with $\text{UO}_2\text{-PuO}_2$, the primary cooling systems being inside the reactor vessel. After Rapsodie, Phenix will be the second fast reactor built in France.

The main features of Rapsodie were defined in 1958 as well as its maximal power (20 Mw). The reactor reached criticality on January 28, 1967.

As an experimental reactor, Rapsodie should bring a valuable contribution in two fields: technological experience with a sodium-cooled fast reactor, and fuel element testing. The latter is undoubtedly the most important one, and therefore Rapsodie will be essentially a facility for fuel element testing and development. No power production is provided. Phenix, on the contrary, will be a power reactor, and is intended to be the prototype of the future large fast breeder power plants.

The design studies of Phenix started at the beginning of 1965, following investigations aimed at the definition of the general characteristics of future large fast reactors, in order to outline the goals of the program. The studies are still in progress. Construction is scheduled to begin in 1969.

A great part of Phenix philosophy can be attributed to the experience of building and operating Rapsodie³⁶⁻³⁸. The idea that sodium is easily mastered by the cubic yard, that it is less pleasant by the cubic foot, and that a single inch of small diameter sodium pipe is always a problem and an expense has been confirmed at Rapsodie. The Phenix "integrated" layout reflects this experience.

Many components of Phenix will be purely developments of Rapsodie components. Chapelot et al.³⁶ believe that sodium-cooled fast reactors can be very much simplified. This also is to be found as a basis of the Phenix philosophy. There is almost no field in which simplification cannot pay, but control systems, fuel handling systems, and auxiliary cooling systems are most promising for future cost reductions. Finally, a major conclusion is that sodium-cooled fast reactors are just reactors, differences have been over-estimated, and almost all troubles and difficulties could have been met in building and starting up any other type of reactor.

Power variations resulting from reactivity problems are not considered as likely sources of trouble, due to limitations imposed on reactivity insertions and on their rate (speed of control rods).

Considering the primary cooling reliability the chosen design gives the best overall safety, and the permanence of the primary sodium level is warranted by the vessel designs. In case of a primary pump failure, the other two pumps will ensure the continuity of cooling. Auxiliary motors deliver a minimum pumping flow rate (in case of a general power failure); they will also be used after a reactor shutdown, until the decay power is sufficiently low to be dissipated by natural convection. Finally in case of complete secondary-circuit failure (for instance following a nuclear accident damaging the intermediate heat exchangers), emergency cooling will be provided around the main tank to dissipate the decay power.

The most serious type of accident presently investigated³⁶ concerns a localized fuel melting caused by a cooling failure (partial or total plugging of a sub-assembly, either by a foreign substance or by cladding

deformation on one or several pins). The structure of the subassembly (percentage of sodium) has been chosen to reduce this risk to a minimum. It is felt moreover that such an accident is hardly likely to spread beyond a few subassemblies. This fact should be confirmed by experimental studies in the Cabri reactor. In any case the Doppler effect is capable of restricting any power excursion corresponding even to a total melting of the fuel. Moreover the main and safety tanks should resist the most serious accident (energy below 100 kg TNT equivalent).

8.2.8 Southwest Experimental Fast Oxide Reactor (SEFOR)

The Southwest Experimental Fast Oxide Reactor (SEFOR) is a 20 Mw(t) fast sodium-cooled reactor with nuclear characteristics similar to the large, soft spectrum, fast breeder reactors fueled with mixed $\text{PuO}_2\text{-UO}_2$. SEFOR will be used to obtain physics and engineering data concerning reactor operating conditions. In particular, SEFOR is designed for the systematic determination of the Doppler reactivity coefficient at temperatures up to the vicinity of fuel melting by means of a series of static, oscillating, and subprompt and superprompt transients.

The SEFOR project³⁹ consists of two parts: the design and construction of the reactor facility and the research and development program. The design and construction is supported by Southwest Atomic Energy Associates (a group of 17 investor-owned utility companies located in the southern and southwestern parts of the United States), Karlsruhe Laboratory in Germany, Euratom, and General Electric Company.

SEFOR safeguards considerations are discussed by G. Billuris et al.⁴⁰ For SEFOR, the radiological effects are calculated on the basis of release of fission products and plutonium corresponding to melting of two-thirds of the core, and an initial pressure of 10 psig within the inner containment barrier. Calculations for the maximum hypothetical accident indicate that fuel melting would be limited to 33% of the core and that the initial pressure of the inner containment would be around 6 psig. The calculation for the radiological effects also assumes leakage rates of 20% of the contained volume for the inner containment, and 2-1/2% of the contained volume for the outer barrier, as compared with the design leakage rates of 1% and 1/2%, respectively. The reasons for choosing the higher rates to evaluate the radiological effects are to allow for uncertainties in periodic

testing that will be performed and the possibility that small leaks may develop in areas of difficult accessibility.

The dual-barrier arrangement has proved to be quite effective in reducing the consequences of plutonium release, which could occur only within the inner containment barrier, if it should occur at all. The interposition of a low-pressure region between the containment barriers reduces the leakage rates by several orders of magnitude over that which would be attainable with the outer barrier alone. The current lack of knowledge of aerosol behavior introduces large uncertainties in predictions of the plutonium that would remain airborne, so that the reduced leakage provides a desirable additional margin for the estimates of the radiological effects of the maximum hypothetical accident.

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9.0 DISCUSSION AND RECOMMENDATIONS

In this chapter the release of fission products from fast reactor fuels and their transport through liquid metal coolant, blanket gas, and containment is reviewed, following in general the sequence of the preceding chapters. In each section recommendations for needed research and development work are given.

9.1 Fission Product Release from Fast Reactor Fuels

The first step in analyzing the release of fission products from core to containment is to determine the amount of fission products present at any given time in core life and at any time after shutdown. The following information is required to permit the calculation of a given isotope formed during fission: composition of fuel, time of irradiation, flux density of the reactor or facility, relative fission yields of each isotope along the decay chain of the mass number of interest and any branching ratios involved, half-lives (or decay constants) of the isotopes along the decay chain, and cross sections for neutron capture by the nuclides.

Several computer codes are available to make these calculations (e.g., ISOGEN at BMI). The most hazardous fission products released from reactor fuels, in order of decreasing volatility, are xenon and krypton; iodine; tellurium and cesium; ruthenium; strontium and barium; and plutonium. The three main types of uranium-plutonium fuels for fast power reactors are mixed oxides, mixed carbides, and U-Pu-Zr or U-Pu-Ti alloys.

The metals and alloys are generally characterized by melting points below 2000°C. In the case of the oxides dispersed in a metallic matrix, the melting points of the fuel are still determined by that of the matrix. The oxide fuel materials are generally characterized by melting points above 2000°C and by a relatively high degree of chemical inertness. The carbides, in general, have high melting points but are characterized by vigorous chemical reaction with a number of materials. The release of fission products is also affected by chemical reactions, solubility in the fuel or its environment, ratio of fuel surface area to fuel volume, duration of molten condition, composition and pressure of cover gas, and other parameters.

The degree of melting is always difficult to postulate, since uncertainty exists concerning the rate of heat loss from the reactor core. The melting of fuels with reasonable burnup levels invariably leads to high release rates

for Xe, I, Te, and Cs. The lowest-melting fuels may also release some Ru and Cs, in addition to Xe, I, and Te; the higher-melting ones release all of these and may also release some Sr and Ba.

Recommendations: Laboratory and pilot plant investigations of the effects of fuel composition and physicochemical characteristics of fission products on the release of hazardous fission products from fuel should be extended.

Fission products can be released from the reactor fuel under various conditions, ranging from leakage and washout by the coolant to a major release during melting or a massive release during a violent nuclear excursion. An extensive long-range research program should be conducted to identify fission-product species and fractions released for each set of circumstances and for degrees of violence to be anticipated in a nuclear excursion.

9.2 Vented Fuel Elements

Realization of the potential of fast ceramic reactors for achieving low fuel-cycle cost requires fuels of high burnup and high power density in order to minimize fuel fabrication, recovery, inventory costs. These fuels, however, release considerable quantities of gaseous fission products, which must be either contained within the fuel cladding or released into the coolant and/or blanket gas. The fission gases can be retained in the core by thick cladding or by low density fuel, but both methods result in poor neutron economy.

Extension of the cladding to provide a plenum (reservoir) above or below the core reduces the pressure buildup in the fuel element by increasing the volume within its cladding. This allows a decrease in cladding thickness, but necessitates an increase in fuel element length, which is limited in practice by economic considerations. Furthermore, the buildup of fission gas pressure can still become quite high, because although the plenum is outside the core it is still an integral part of the cladding.

Buildup of fission-gas pressure within the fuel element cladding can be avoided by venting the fuel element into the coolant or into the coolant blanket gas. If the fuel elements are vented to the coolant, the volatile fission products that escape from the fuel will be released into the coolant

from every vented element, greatly increasing the volatile fission product burden of the primary coolant. On the other hand, to the extent that venting of fuel elements reduces cladding ruptures, the amount of non-volatile fission products introduced into the coolant by leaching of the fuel will be reduced.

The objective of venting a ceramic fuel to coolant is to release selectively the long-lived fission gases so that the pressure stress on the fuel element can be reduced; hopefully, this will lead to a less frequent failure of fuel elements, thus reducing contamination of the coolant through leaching. Venting to coolant may actually reduce, rather than increase, the burden of fission product contamination.

Recommendations: Present investigations of various types of vented fuel elements should be more strongly oriented toward impartial evaluation of the relative merits and deficiencies of each type; vented-to-coolant fuel elements must be compared in detail with nonvented and vented-to-plenum fuel elements, with particular regard to fuel element integrity and to degree and types of fission product release to coolant and blanket gas. For effective utilization of high burnup fuels, venting of fuel elements may become standard practice in sodium-cooled fast breeders.

9.3 Fission Product Transport

The transport of fission products from the fuel element into the primary coolant and blanket gas can be of any degree of severity and can also differ in regard to what fission products are released to the coolant. These differences are dependent not only on the amounts of fission products generated in the fuel but also on the extent of plateout or sorption on the inside walls of the fuel element, the extent of containment by filters or traps within fuel elements of certain designs, the size and location of the orifice through which they escape the cladding, and the fuel temperature at the time of release.

Fission product releases by diffusion through intact cladding or by leakage through pinholes or very small cracks in the cladding are usually an operational problem only. Releases from larger cracks become a safety problem when the cumulative product of crack sizes and number of defective fuel elements contaminates the primary coolant to a level that significantly increases the hazard of a rupture in the primary coolant system.

Radioactive decay, together with containment in a closed volume or dilution and discharge, or both, is the only really effective means now available of handling the noble gases.

Radiological study of the relative importance of the various fission products indicated that below about 1% release of ^{131}I from sodium (and assuming a nominal fuel release fraction of 40%) other isotopes begin to acquire controlling significance with respect to hazard determination. Further studies are required to substantiate this premise; but clearly the role of other isotopes normally considered less significant must be evaluated when iodine retention is demonstrated. Both in-pile and out-of-pile studies of fission product retention by sodium have demonstrated the retention of iodine. Experiments have shown significant retention by liquid sodium of zirconium-niobium, ruthenium, barium-lanthanum, and cesium. Following the appreciable reduction of iodine and other fission products by sodium retention, strontium, cesium, and plutonium become (after noble gases) the most significant radiological isotopes to be considered in reactor accidents.

Recommendations: Pilot-plant and semi-engineering-scale experiments should be accelerated in order to evaluate the applicability of small-scale experiments in fission product transport to realistic accident conditions. These studies include the extent of reaction of hazardous fission products with sodium, the ability of sodium to retain them during and after accidents of the types and degrees of severity considered credible in sodium-cooled fast reactors, the extent which fission products are carried through the sodium coolant into the blanket gas by bubbles of fission gases from the fuel, and the extent of plateout and other means of retention.

More information is needed concerning operations involving cladding failures under high burnup conditions, and attention should be given to the behavior of fuel elements that are operated following failure. A safety problem will occur if fuel jacket failures cause a sudden general injection of gross fission products into the coolant. The program should also examine conditions anticipated in vented fuel element operations, particularly during fuel reloading periods.

9.4 Plutonium and Other Fission Products in Sodium Aerosols

Fission products are likely to be present in the sodium coolant as dissolved metals. Exceptions to this are the rare gases, which escape unreacted, and iodine, which would be predominantly in the form of sodium iodide. The concentrations of iodides other than sodium iodide depend on the temperature and concentrations of the reactive metals in the sodium. Upon exposure to air, fission products at the surface will be oxidized if the surface temperatures are high enough.

Large quantities of sodium oxide aerosols containing plutonium, other fission products, and ^{24}Na may be dispersed as a result of a primary coolant sodium fire following certain postulated accidents in a sodium-cooled fast reactor. Sodium fires can be divided into combustion of a sodium pool and combustion of a sodium spray. A special case of the spray fire is one in which sodium is introduced explosively into an oxidizing atmosphere.

Of major concern in pool fires involving a reactor coolant are the transport of ^{24}Na activity in the smoke from the fire and, if conditions responsible for the fire have also caused failure of the fuel clad, fission product transport.

The sinking of oxide particles in a burning pool of sodium could result in formation of fission product oxides which tend to have very low volatilities. On the other hand, failure of fission products to oxidize at the surface because the gas is depleted in oxygen could result in substantial volatilization of fission products as metals. This is very dependent on the rate of diffusion of fission products to the surface. These metals may form oxides as they rise through the air-burning region (assuming the burning process occurs predominantly in a gas-phase reaction zone). If such volatilization and oxidation occurs, the fission product oxides will behave similarly to the sodium oxide which is formed, and may subsequently agglomerate, fall out, or plate out. Iodine is an exception to this. Its release could occur by vaporization of the iodide or by entrainment. The entrainment mechanism must be considered for the release of sodium and dissolved fission products which adhere to particles carried up by convective air currents.

Recommendations: There should be additional work on sodium aerosol sampling techniques and devices, to facilitate the study of the behavior of sodium liquid, sodium vapor, and sodium oxide aerosols under accident conditions.

The equipment developed and the experience gained with water-cooled reactors on aerosol behavior and filtration by high-efficiency filters may be adaptable to sodium-cooled reactors. However, to prevent excessive loading of the high-efficiency filters, high concentrations of sodium oxide aerosols must first be drastically reduced by baffles, scrubbers, and/or graded deep-bed filters of stainless steel fibers. Design of an air cleaning system for such a facility requires basic data defining the aerosol characteristics of sodium and plutonium released during a fire or an explosion. All reports of particle size data should specify whether agglomerates are included.

9.5 Engineered Safeguards

High-power high-burnup fast power reactors present some new safety considerations because of the very much larger inventory of radioactive fission products, such as noble gases and plutonium.

In liquid metal-cooled reactors, a high coolant temperature (500 to 650°C) is possible without pressurization. This is a considerable safety advantage in cases of rupture of the coolant system. Sodium has certain disadvantages, however. It reacts chemically with oxygen and water and must therefore be isolated from them. Further, exposure of sodium to neutrons produces ^{24}Na , a strong gamma emitter with a half-life of about 15 hr, and this creates a serious problem in nuclear power plant applications. For these reasons, in the present conceptions, the steam boiler has been separated from the primary sodium coolant by a secondary sodium coolant system, which does not become radioactive. The vault necessary to contain the radioactive primary sodium provides the shielding required and also acts as a container for the inert blanket gas which surrounds the primary system.

The loss of coolant in a power reactor would inevitably lead to fuel melting, and it is important to show both theoretically and experimentally that isolated trouble spots will not spread progressively throughout the whole reactor. Any defect must be identified quickly by the detection of incipient sodium boiling flow or outlet temperature in a subassembly, or increase in fission product concentration in the bulk coolant, and followed by corrective action to prevent formation of a serious void in the coolant or the spread of fuel melting.

Removal of the large fission product afterheat could be difficult if the primary system were disrupted in an accident. Special provisions will be required to prevent or limit a non-nuclear accident (e.g., a sodium-water reaction) that could disrupt the entire secondary system if no alternate afterheat removal system were available. The afterheat removal problem may be related to the choice of a basic primary system design.

Recommendations: Further research should be conducted in fission product release under core vaporization conditions, directed to large-scale tests of engineered safeguards, and these tests must be related to realistic accident conditions. Laboratory and pilot plant experiments are required to design these tests and to interpret the results. Engineered safeguards will be required to prevent or control fires, to control aerosols of sodium and fission products, to detect coolant voids, to remove fission product afterheat in case of disruption of the primary system, and to contain all radioactivity within permissible limits.

The development of engineered safeguards for liquid-metal-cooled reactors will benefit from studies of the applicability of the concepts that have evolved in the water reactor safety programs. For example, the protection of the air cleaning system in a nuclear installation against loss of efficiency or complete failure is essential in the design of the system. Practical methods for the retention, storage, and disposal of noble gases (krypton, xenon) in large quantities under accident conditions should be developed. The merits and disadvantages of relatively new types of engineered safeguards must be evaluated early in the safety programs, since they may influence the basic designs of liquid metal fast reactors.

9.6 Accident and Fission Product Release Models

The study of reactor accidents by the use of sequential analytical models has been proposed as part of the American Nuclear Society's Standards Program for some time. The research effort on plutonium and fission product release and transport in sodium systems has not been adequate for accident evaluations, and acceptable information is not currently available. Furthermore, the models of reactor accidents for water-cooled power reactors can be used only as preliminary guides to estimating fission-product release.

Fission product release assumptions typically evolve from studies that begin in a series of laboratory experiments, progress through intermediate-size studies, and finally are predicated upon the experience gained in the operation of full size systems. However, safety analyses of sodium-cooled fast reactors have been largely conducted by extrapolations from small capsule experiments. This procedure may be inadequate until the scaling applicability of such things as plating and other surface effects is determined.

It is important to show by a sensible theoretical analysis with reasonable experimental support that isolated trouble spots in a reactor would not spread progressively throughout the whole reactor. For the fast reactor the safety limitation may be the time taken to sense local void formation or blockage of coolant flow and shut down the reactor to prevent spread of damage. If fast-reactor safety in this situation could be assured, it might be possible to show that no substantial release of fission products could occur and hence, that containment would not be needed.

Severe accidents in sodium-cooled fast reactors might produce aerosols consisting of sodium, sodium oxides, fission products, and possibly fuel particles that would be distributed within the containment system. It is important in considering such an accident to have available a theoretical model substantiated by experimental evidence that can be used to evaluate the hazards created and the safeguards necessary to meet containment criteria.

Sodium fires may be generally categorized into three types: (a) a fire in which the integrity of the equipment cell is maintained, the initial oxygen content is controlled, and the oxygen is depleted during the burning period; (b) a fire in which integrity of the cell is not maintained (or fails due to the high energy release during burning), thus allowing a greater fraction of ^{24}Na or fission products to escape; and (c) transport of the oxide and fission products into a secondary containment region, where either absolute containment or dynamic cleanup must be provided in order to minimize the further transport of activity.

Recommendations: Experiments in various laboratories and in reactors have provided much data on release of fission products under conditions that simulate reactor accidents. The effects of important variables on fission-

product behavior have generally been recognized, but interpretations of the data have been at best empirical. Analytical models of fission product release and transport need to be developed for realistic accident conditions.

Modeling and simulation techniques are needed for explosive reactions, vault mockups, and spray ejections. Model experiments and large-scale experiments will be required in a long-range development program. Model research into the problem of sodium fires should include mass and energy release and deposition, definition of sodium burning rates, sodium oxide release rates, and fission product release rates. Conditions in a sodium fire should be completely defined so that energy and mass balance calculations on the complete containment complex can be made with confidence during reactor hazards analysis.

The future extension of capsule experiments should include mockup and simulation studies of intermediate size and complexity. The first steps in the sodium study comparable to the water- and gas-cooled efforts have been taken. Equipment and techniques are available, but considerable work must be done to provide data upon which the acceptance of the capability of the sodium coolant for fission product retention may be based.

10.0 CONCLUSIONS

Most liquid-metal fast breeder reactors of current design will use ceramic fuels (mixed oxides, carbides, or nitrides of uranium and plutonium). These fuels will be operated to high burnup, and thus will accumulate large fission-product inventories and will release large fractions of the volatile fission products. Expansion of high-burnup fuels due to solid fission products must be accommodated by low effective fuel density (compacted powders, pressed and sintered pellets, or axial cores), or by annular expansion volumes between fuel and cladding. The annulus can be filled with gas or sodium.

The buildup of pressure from volatile fission products could be contained by thick cladding, but this is expensive in terms of breeding ratio. The cladding can be extended to provide a plenum (reservoir) above or below the core, which reduces the pressure buildup in the fuel element by increasing the volume within the cladding. This allows a decrease in cladding thickness, but necessitates an increase in fuel element length, which is limited in practice by the additional sodium coolant required and by increased coolant pressure drop and consequent increase in coolant pumping power.

Gaseous fission product pressure could become quite high in vented-to-plenum fuel elements, because although the plenum would be outside the core it would be an integral part of the cladding. Increased stresses on the cladding due to buildup of gas pressure can be avoided by venting the fuel element into the primary sodium coolant. Current development work on venting devices emphasizes holdup within the fuel element long enough for short-lived fission-product gases to decay and for their decay products to deposit within the cladding; thus the long-lived fission-product gases would be selectively released. Venting should lead to less frequent failure of fuel elements and thus reduce contamination of the coolant through leaching, and it should minimize sudden uncontrolled release of noble gases from rupture during operation or meltdown, with the possible exception of releases resulting from fast excursions.

The advantages of venting fuel to the sodium coolant must be weighed against the possibilities of increased shielding requirements, additional decontamination problems in normal maintenance, and increased hazard to

personnel from primary coolant system leaks involving few or no damaged fuel elements. In the case of vented sodium-bonded carbide or alloy fuels, the possibility of loss of bond before bulk coolant boiling occurs must also be considered. Fuels that were vented to a plenum, rather than to the coolant, would not be subject to these problems as long as the cladding remained intact. Venting of fuel elements to the coolant may become standard practice in sodium-cooled fast breeders, but it is still under evaluation.

During normal operation and after minor meltdowns, most of the iodine and other fission products released from vented or ruptured fuel elements will be retained in the sodium coolant system. The major fraction of the long-lived noble gases and ^{134}Cs , and some of the ^{137}Cs and other fission products, daughters, and activation products diffuse into the argon blanket gas and may be largely removed by blanket-gas purification systems. Purification equipment is necessary in both the sodium coolant system and the blanket-gas system, to keep radioactivity from exceeding safe levels during operation, and it may reduce the amounts of fission products available for release to the containment system in the event of a major accident.

In the event of rupture of the primary coolant system, sodium containing radioactive ^{24}Na , fission products, and possibly fuel materials would be released into the nitrogen-filled containment enclosing it. If the containment integrity were breached by the pressure of sodium vapor expansion, a pressure surge, or a postulated nuclear excursion, these materials would escape into the air atmosphere within the secondary containment, the sodium would ignite, and aerosols of sodium oxide and other materials would be produced.

Noble gases released into the secondary containment system under accident conditions must be retained until they have decayed to acceptable levels, followed by controlled release from a tall stack. The release must be gradual enough to prevent rapid contamination of the outside atmosphere by ^{85}Kr , which has a half-life of ten years and therefore would not decay significantly during a postaccident decay period of economically feasible length.

After a major accident involving the burning of sodium containing fission products and fuel materials, a large part of the fission products, UO_2 , and PuO_2 might be occluded by, attached to, or sorbed by the sodium

oxide aerosols, most of which would agglomerate rapidly into particles that could be removed by filtration. The filtration problems created by fine, uncoagulated aerosol particles might be aggravated by the presence of radioactive ^{24}Na . On the other hand, most of the iodine would probably be in the form of NaI or otherwise associated with sodium; this might reduce or possibly eliminate the necessity for removal of gaseous iodine and methyl iodide by charcoal beds.

The equipment developed for and the experience gained with water-cooled reactors on aerosol filtration by high-efficiency particulate air filter systems may be readily adaptable to sodium-cooled reactors. However, to prevent excessive loading of the high-efficiency filters, high concentrations of sodium oxide aerosols must first be drastically reduced by baffles, scrubbers, and/or graded deep-bed filters of stainless steel or other fibers.

Much additional research and development and extensions of present work are needed in the control and containment of fission products and other radioactive materials in liquid-metal-cooled fast breeder reactor plants. Examples of areas that require additional work are the sampling, characterization, agglomeration, and filtration of aerosols produced in the combustion of sodium containing fission products, activation products, and fuel materials; scaleup of the results of small-scale experiments; accident models, fission-product release models, and simulation techniques; further evaluation of vented fuel elements; reliability of engineered safety features; transport of fission products in the primary sodium coolant-blanket gas system; and release of fission products from fuel to coolant and blanket gas. These recommendations are discussed in more detail in the preceding chapter, "Discussion and Recommendations".

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Appendix A

DISCUSSIONS ON FAST REACTOR SAFETY

The decision to declare a reactor safe or unsafe is very difficult. The many factors may be evaluated on the basis of probability, but even with an evaluation of this type the final decision must be based largely on judgment of the highest quality. There are a variety of opinions concerning accidents in fast reactors and concerning the types and amounts of containment necessary to guard against these accidents. Some of the more authoritative are reflected in the information below. The first two sections were extracted from articles on reactor safety by two outstanding authorities; the last section is an abstract reproduced in its entirety.

- Al. Basic Safety Considerations for Fast Reactors (Extracted from "Some Basic Safety Considerations in the Development of Power-Producing Reactors," by F. R. Farmer)¹

This article discusses the range of the more serious accidents possible in large power-producing reactors and the effect the limitations imposed by safety considerations have had on the development of various reactor systems. Reactor safety provisions are then reviewed with the objective of seeking simplification of the present complex systems.

In the normal operation of a reactor, the heat generated by the fuel is balanced by the heat removed by the coolant. This situation might be changed as a result of the following:

1. The addition of reactivity through movement of fuel or absorber or a change in neutron moderation.
2. A reduction or loss of coolant flow either throughout the core or locally to some region.

The complete loss of coolant flow in the whole or in part of any power reactor must inevitably lead to fuel melting if the reactor remains at full power. It is important to show by a sensible theoretical analysis with reasonable experimental support that isolated trouble spots in a reactor would not spread progressively throughout the whole reactor.

With so little experience available, it would be presumptive to present an analysis purporting to cover all fast-reactor systems. Some general points have emerged, however, from the studies made already in connection with the UK program.

An attempt has been made in some designs to prevent the complete loss of coolant flow by the use of multiple circuits and by the diversification of power supplies. For example, in the Dounreay fast reactor, there are 24 primary coolant circuits and 24 primary coolant pumps powered in pairs by 12 diesel generators. It is doubtful whether complications of this type will find general favor. It is commonly accepted, however, that a loss of power to the pumps is possible.

In the sodium-cooled system, the important effect of a loss of flow is that the total amount of heat removed from the reactor decreases and the bulk sodium temperature increases. If bulk boiling occurs, both coolant flow and rate of heat transfer decrease drastically and give conditions locally that are almost equivalent to a complete loss of cooling. The design must ensure that bulk boiling will not occur at any time during a loss-of-flow accident. With some values of reactivity coefficients, designing the circuit for a high degree of natural circulation and designing the pumps to have suitable rundown characteristics will ensure that a reactor scram is not required and that the reactor power will decrease safely to a value appropriate to the residual flow. These inherent safety features may not be adequately proven or economically provided in early fast-reactor designs. Safety will then depend on a reactor trip based on coolant temperature or the provision of auxiliary means for maintaining adequate flow to remove heat in step with decreasing neutron power. Both these features may be included in some designs.

A further fault condition to be considered is the loss of cooling in a limited region of the reactor, such as a single fuel assembly. Severe damage would be expected in the region directly affected, but this would not be a major hazard if the condition did not propagate throughout the whole core. This has been recognized as a major problem in fast reactors, and an extensive investigation of this situation is underway. It is recognized that damage could spread through thermal effects, through mechanical distortion of adjacent core members owing to pressure generated by sodium boiling, or through circulation of debris by the coolant. There is reasonable assurance that melting induced by local blockage would not propagate throughout the core provided the reactor was shut down rapidly. The problem is then one of identifying the defect sufficiently quickly, as, for example,

by the detection of either sodium boiling, variations in flow or outlet temperature in every subassembly, or fission products in the bulk coolant.

A sodium-cooled reactor with oxide fuel dispersed in a stainless steel matrix and canned in stainless steel was taken as an example for discussing the depressurization accident. In normal operation the averaged temperature of a fuel element at the core center may be about 600°C , and the failure temperature for this type of fuel cannot be higher than the melting point of stainless steel, 1430°C . It is easy to show that a reactor of this type cannot survive a complete loss of coolant. For a reactor rating at 200 Mw/ton, the complete loss of coolant would lead to a fuel temperature rise of 500°C per second at full power and a rise of over 700°C within the first 10 sec after shutdown. Sodium-cooled reactors operate at a low pressure, however, and use the coolant at temperatures well below the boiling point. Although reactor designers face some new problems in vessel designs for high temperatures and temperature gradients, the vessels are not at the same time required to retain high pressures. Designs in which the reactor and heat exchangers are housed in close-fitting compartments or are suitably double skinned should eliminate the possibility of exposing the core through loss of coolant. It may well be that the development of fast reactors and the limitation of their rating will depend on the development of suitable sensors to give rapid and clear indications of local faults.

An attempt was made on the Dounreay fast reactor, where personnel access to the containment vessel was required, to maintain acceptable working conditions by using a self-contained air-cleaning system. This proved to be difficult, and the reactor is now fitted with the normal arrangement of a fresh-air ventilating system and isolating valves that will seal the containment vessel on demand. Thus, if the containment structure is to provide the protection required after an accident, a number of valves must operate and seat perfectly on piped services and on the ventilation plant.

The simple concept of a perfect steel enclosure is, in practice, transformed into a more complex structure. Various alternatives to full containment have been under investigation for some years. These aim at reducing the amount of fission products available for leakage or reducing the pressure causing the leakage. Some fission products may be absorbed on

charcoal, copper, or other active surfaces or combined in solutions. Water or solution sprays can be used to remove heat, to condense steam, and to trap fission products. Combinations of these methods can be used.

One type of containment is illustrated by the "confined" reactor, e.g., the Oak Ridge Research Reactor (ORR) at Oak Ridge, Tennessee. The reactor building has a reasonably low leak rate under atmospheric pressure conditions and is ventilated through a scrubbing plant. This system may be acceptable for those reactors which have a low-energy primary circuit that cannot accidentally release considerable volumes of steam or gas.

A second system, called "vapor suppression", has been under investigation in the United States for several years for use with water reactors. In the case of a fractured circuit, this system seeks to reduce the peak pressure to which the containment is subjected by rapidly condensing the steam released. In one form the escape path of the contents of the primary circuit is arranged so that the steam will pass through a water trap and the inner building will thus be subject to a lower total pressure pulse of short duration.

Another system uses the idea of "vented containment" and exploits the fact that a significant interval could elapse between the fracture of the primary circuit and the melting of fuel, with its accompanying release of fission products. Thus the containment structure would allow the initial release of the gas or steam-water-air mixture, but the valves in the structure would be arranged to close before there was a significant release of activity. The containment structure is designed for relatively low pressures.

These developments in the containment concept, representing a move away from the idea of full pressure containment, are to be welcomed as a first step. If release of fission products from the reactor can be positively prevented by simple emergency procedures of the type discussed in this article, no containment would be necessary. Structures around reactors would serve such functions as preventing ingress of air in a gas-cooled reactor and maintaining an inert atmosphere in a sodium-cooled reactor. Simple filtration systems fitted to these structures would deal with fuel-handling accidents, spillage of active coolant, etc.

For the fast reactor the safety limitation may be the time taken to sense local blockage of coolant flow and shut down the reactor to prevent spread of damage. If fast-reactor safety in this situation could be assured, it might be possible to show that no substantial release of fission products could occur and hence, that containment would not be needed. In the present development of pressurized-water reactors, the fundamental approach to safety by necessity differs from that outlined above. Although the system has inherent characteristics that assure safety in the event of most types of accident, it is not yet possible to guarantee that fuel melting will not occur in a depressurization accident. The provision of containment is a natural consequence of this finding.

A2. Safety in Fast Reactor Design (Extracted from "A Look at Fast Reactor Safety," by David Okrent²)

This article reviews briefly some of the major safety features in the design of early fast reactors, discusses the "maximum accident" approach taken in their safeguard analyses, and mentions a few safety-related operating experiences encountered during their operation. The different safety characteristics and safety problems entering into large reactor design are then discussed.

The energy release and damage potential for hypothetical nuclear accidents leading to explosive disassembly remain to be evaluated fully for large fast reactors. Other things being equal, larger cores would lead to larger energy releases.^{3,4} However, calculations by Nicholson⁵ and by Wolfe, Friedman, and Riley⁶ indicate that negative Doppler effects can sharply reduce the magnitude of such a nuclear accident, as well as the probability of its occurrence.

Recently, model experiments with chemical explosives have confirmed the ability of the primary reactor-containment structure of the Rapsodie reactor to withstand practically all the mechanical effects of the worst credible calculated incident. Therefore the quantity of sodium released into the reactor building would be limited, and the risk of a large sodium fire would be eliminated.^{7,8} It is expected by these authors that similar containment would be possible for larger fast reactors based on a similar generation of energy density during the burst.⁸ Experiments and analyses pertaining to the primary containment of explosive nuclear accidents have been performed in

Europe and in the United States⁹⁻¹². References (13) through (21) contain studies of the stability of fast reactors.

Detailed specification and examination of credible accidents that could lead to explosive disassembly in large fast reactors have not been completed. It is likely that sophisticated analyses of the motion of coolant and fuel will be necessary so that reactivity insertion rates and core shutdown characteristics can be estimated more realistically. The magnitude of the burst depends primarily on the reactivity insertion rate when passing through prompt critical, rather than on the total amount of reactivity available^{3,4}, and hence this is a crucial parameter. Similarly, if shutoff requires the generation of sizeable pressures, then the density of the fuel, the temperature at which pressure generation rises sharply, and the equation of state of the material all play an important part in determining the energy release required for disassembly. One question that has arisen, particularly for the larger, more loosely coupled fast reactor cores, relates to the possible effects on the remainder of the core when only a portion of the large core is initially involved in an energy release large enough to generate sizeable pressure.

The high-power high-burnup fast power reactors contemplated for the future present some new safety considerations. The very much larger radioactivity inventory is an obvious hazard. If a credible accident were capable of releasing the bulk of these fission products into the reactor building, the previously used single containment shell might not prove adequate for many sites. Most methods developed for reducing the building pressure and fission-product concentration in the containment building following a bad accident in water-cooled reactors are probably not directly applicable, and a new approach to engineered safeguards may be required.

Removal of the large fission-product afterheat could be difficult if the primary and secondary systems were disrupted in the accident. Special provisions might be required to prevent or limit an accident (e.g., a sodium-water reaction) that could disrupt the entire secondary system unless an alternate means of afterheat removal were available.

A large amount of fissionable material representing several critical masses if reassembled at full density would be present in the core. The guided dispersal of such fuel into a subcritical configuration in the event

of a slow meltdown accident and the maintenance of an appropriate amount of coolant on this material may prove to be a difficult problem if such recourse is necessary.

Ease of accomplishment of afterheat removal and primary containment, if necessary, may be related to the choice of basic primary system design. Of four 1000-Mw(e) liquid-metal-cooled fast reactor designs, one used the submerged primary system concept of EBR-II and the other three employed the more conventional arrangement of double-walled reactor vessel, piping, and components in the primary system.

A problem not new to large reactors, but perhaps aggravated by their higher flux levels, is related to the growing awareness of the possibility of undesirable effects on structural materials of long-term irradiation by neutrons. Both reactor vessels and major structural components within the vessel may be subject to increasing scrutiny in this regard.

A further problem, which may be introduced by operation with higher sodium temperatures, is the evaporation of sodium and its subsequent condensation and freezing in cooler regions, possibly on mechanisms or in small clearances. Freezing of control-rod drives and the consequent loss of scram capability with extended operation are obviously to be avoided, even if only a fraction of the total number of rods is needed for shutdown. Two distinctly separate mechanical designs for the control rods in simultaneous operation in each reactor may be desirable to provide the degree of redundancy now accomplished in many other reactors.

None of the four design studies went to sufficient depth to identify the maximum credible accident or to show how containment and afterheat removal would be accomplished. Since afterheat may be generated at the rate of roughly 100 Mw the first day, the absorption or removal of this heat from possibly disrupted primary and secondary systems can present significant problems.

In a paper presented at the Third Geneva Conference, Vathaire, Vernier, and Pascouet⁸ gave some of the French views on fast reactor safety in support of the design concept discussed by Zaleski²² and Vendryes²³. They state, "It would appear that in the case of realistic incidents, the effect of a positive sodium coefficient may be more than compensated by that of a negative Doppler coefficient." They go on to state that "Accidents due to general

failure of the cooling system (pump breakdown, loss of coolant) may be prevented by design (number and inertia of pumps, double circuit containment, coolant reserves). The prevention of local coolant incidents (due to disturbances in the flow around the fuel itself) raises rather more difficult problems. An effort must be made to ensure rapid detection of such irregularities whose undetected development is particularly dangerous. The possibility of sodium-water reactions in steam generators must also be carefully examined."

Concerning the containment of large future fast reactors, Vathaire says "Studies made so far lead to the conclusion that the very pessimistic accident conditions taken as the basis for the definition of early prototype fast reactor containments could be made much more optimistic. It may perhaps already be said that the mechanical effects of credible power excursions could be contained by a smaller primary enclosure immediately surrounding the reactors, and that the probability of a sodium fire after such an excursion can be practically excluded by suitable designs, and finally that only a very small proportion of radioactive substances would be released in the reactor building.

We may consider that large fast reactor power houses of the future may be accommodated in simple containment buildings equipped with filters and ventilation."

A3. Reactor Safety and Siting (Abstract of a Paper, "Reactor Safety and Siting - A Proposed Risk Criterion." by F. R. Farmer²⁴)

The building of a reactor implies the acceptance of some finite degree of risk. No engineering plant and no structure is entirely risk-free and there is no logical way of differentiating between "credible" and "incredible" accidents. The incredible is often made up of a combination of very ordinary events - for example, the breakdown or deterioration that occurs in normal plants and their measuring instruments; and the credible may be exceedingly improbable. The logical way of dealing with this situation is to seek to assess the whole spectrum of risks in a quantity-related manner, and the purpose of this paper is to show how this may affect the choice of sites for nuclear power stations.

Some assumption of plant failure or maloperation provides the starting point for a safety analysis. A measure of risk can be obtained by estimating the probability of the failure and assessing the consequences. Any initiating event sets up an accident sequence which can follow many paths where the likelihood of following any one path depends upon the performance of many items of plant. From knowledge of this performance, a probability can be assigned to each pathway and a full safety evaluation would then be comprised of events with associated probabilities and associated consequences. A convenient scale for probabilities would be the average time interval between events, e.g., reactor years, and for consequences, the equivalent ground-level release of ^{131}I . A safety criterion can then be defined by an upper boundary of permissible probability for all fault consequences.

The line defining the safety criterion will reflect three main considerations:

1. Possible public reaction after an accident,
2. The estimated number of casualties likely to arise in the population affected by the release,
3. The increased risk incurred by any individual.

To establish the collective risk in terms of casualties or individual risk it is necessary to define a population distribution and this has been chosen to be typical of that around suitable industrial sites in or near large cities in the UK. Considering either the individual or the community, a line can be chosen so that the risk can be a very small fraction of that occurring naturally. It is likely that the anticipated public reaction applies a greater restriction than that obtained by risk estimates.

Some conclusions of a general nature are presented:

1. There is no need to obtain greater precision in the estimation of fission-product release than the precision that can be attributed to the probability of a release.
2. If any single feature of the reactor is used to reduce a potential release of fission products by many orders of magnitude (as by filtration containment or reactor control), then the reliability of this feature should be established to at least the same order of magnitude.

3. The final presentation of reactor risk is inexact and may, at best, be claimed to represent the reactor within a factor of 10. This is not an outcome of the present method, but a consequence of shortcomings in knowledge of plant performance, equally, or more so, than any shortcomings in knowledge of reactor and fuel behavior.

Those sites in the UK which might be used in a power network vary by a factor of 10 at most in population density. The position of the boundary line may be placed by informed opinion also within one order of magnitude. Hence, reactors cannot be matched to sites with any precision. There is obviously a development period in all countries in which site selection is very much a matter of political wisdom. It is already clear in the UK that reactors must attain a standard of safety that will permit complete freedom in siting; a similar situation will be reached shortly in a number of countries. This paper has proposed a means by which an appropriate safety standard may be defined by any country, and a means of assessing the ability of a reactor to meet the standard.

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Appendix B

PHYSICAL AND CHEMICAL DATA ON FISSION PRODUCTS AND FUEL COMPONENTS

Table B.1 Free Energy of Formation of Oxides of Fissile, Fertile, and Fission-Product Elements

Compound	$-\Delta F_T^O$ (kcal/gram-atom of oxygen)				
	500°K	1000°K	1500°K	2000°K	2500°K
ThO ₂	135	124	113	101	89
Ce ₂ O ₃	134	123	112	100	90
La ₂ O ₃	132	123	112	101	91
SrO	129	117	106	87	67
BaO	122	111	100	87	70
ZrO ₂	119	108	97	86	75
UO ₂	96	86	76	dec	
PuO ₂	128	102	92		
Cs ₂ O	54	34	7	dec	
MoO ₂	55	46	36	26	dec
MoO ₃	50	40	32	29	26
TeO ₂	28	16	5	dec	
RuO ₂	15	5	dec		
RuO ₄	11	6	2	dec	

dec = decomposes.

Table B.2 Yields and Characteristics of Important Fission Product Isotopes Arranged Approximately in the Order of the Volatility of the Elements^a

Isotope	Half-life	Activity [Kilocuries/Mw(thermal)]		Total Weight [g/Mw(thermal)]		Boiling Point ^c (°C)	Probable Release and Transport Form ^d	Hazard
		After 1 yr of Irradiation ^b At Shutdown	1 Day After Shutdown	Isotope	Element			
High Volatility (Group I)								
Kr	Stable	-	-	4.9		-153	Elemental gas	External radiation,
-83m	114m	3	0					slight health
-85	10.27y	0.1	0.1	0.3				hazard
-85m	4.4h	8	0.2					
-87	78m	15	0					
-88	2.8h	23	0.1					
-89	3m	31	0					
-90	33s	38	0		5.2			
Xe	Stable	-	-	55.4		-108	Elemental gas	External radiation,
-131m	12d	0.3	0.3	0.003				slight health
-133m	2.3d	1	0.7	0.003				hazard
-133	5.27d	54	47	0.3				
-135m	15.6m	16	0					
-135	9.2h	34	14	0.01				
-137	3.9m	48	0					
-138	17m	53	0					
-139	41s	61	0		55.7			
Br	Stable	-	-	0.03		59	Elemental Br ₂ or HBr	External whole
-83	2.3h	3	0					body radiation,
-84	32m	6	0					moderate health
-85	3m	8	0					hazard
-87	56s	15	0		0.3			
I	Stable	-	-	0.45		183	Elemental gas	External radiation,
-129	1.7x10 ⁷ y	10 ⁻⁶	10 ⁻⁶	7.92			I ₂ or HI;	internal irradiation
-131	8d	25	23	0.20			variable	of thyroid, high
-132 ^e	2.3h	38	0	0.004			gaseous	thyroid, high
-133	21h	55	26	0.05			organic	radiotoxicity
-134	52m	63	0	0.002			compounds;	
-135	6.7h	55	4.4	0.01			adsorbed forms	
-136	86s	53	0	0.000	2.6		on small particles	
Cs	Stable	-	-	22.7		685	Elemental Cs	Internal hazard
-134	2y	0.029	0.29	0.03			converting to	to whole body
-136	13d	0.147	0.147	0.002			CsOH	
-137	26.6y	1.28	1.3	12.2	35.0			

Table B.2 (continued)

Intermediate Volatility (Group II)						987	Elemental Te above 1000°C, converting to TeO ₂	External radiation, moderate health hazard 132Te contributes a health hazard from 132I daughter
Te	Stable	-	-	5.1				
-125m	58d	0.005	-	0.0003				
-127m	105d	0.414	0.5	0.04				
-127	9.4h	2.1	0.5			to		
-129m	34d	2.9	2.3	0.2				
-131m	30h	3.9	2.2	0.0007				
-131	25m	26	0	0.1				
-132	77h	37.3	30.1	0.1				
-133m	52m	54	0					
-134	44m	57	0					
-135	2m	36	0	5.4				
Volatile Under Highly Oxidizing Conditions (Group III)						4230	Volatile oxide, RuO ₄	Internal hazard to kidney and GI tract
Ru	Stable	-	-	17.4				
-103	41d	25.7	25.7	0.8				
-106	1y	1.54	1.54	0.5	18.7			
Tc-99	2.12x10 ⁵ y	10 ⁻²	10 ⁻²	9.3	9.3	(4600)	Volatile oxide, Te ₂ O ₇	Internal hazard to tract and lung
-99m	6.04h	5.5	0.1	0.001				
Mo	Stable	-	-	34.3	34.4	(4800)	Volatile oxide, MoO ₃	Internal hazard to GI tract and lung
-99	67h	51.5	40	0.1				
Low Volatility (Group IV)						1366	Elemental Sr converting to SrO	Internal hazard to bone and lung
Sr	Stable	-	-	6.5				
-89	54d	39	39	1.5				
-90	28y	1.2	1.2	8.7				
-91	9.7h	51	9.7	0.0	14.6			
Ba	Stable	-	-	14.6		1635	Elemental Ba converting to BaO	Internal hazard to bone
-140	12.8d	53	48	0.7	15.3			
Sb-125	2.7y	0.04	0.041	0.10	0.10	1640	Elemental Sb or Sb ₂ O ₃	Internal hazard to GI tract, lung, total body, and bone
Refractory (Group V)						1602	Elemental Sm converting to Sm ₂ O ₃	Internal hazard to bone, lung, and GI tract
Sm	Stable	-	-	5.2				
-151	93y	0.01	0.01	0.42				
-153	47h	1.4	1.0	0.003				
-156	~ 10h	0.1	0.02	0.00	5.6			

Table B.2 (continued)

Pm-147	2.6y	4.8	4.8	5.0	5.0	(2700)	Elemental Pm	Internal hazard to
-149	54h	11.8	8.7	0.02			converting to	GI tract, bone and
							Pm ₂ O ₃	lung
Pr	Stable	-	-	11.4		3020	Elemental Pr	Internal hazard
-143	13.7d	52	45	0.8			converting to	to GI tract
-145	6h	36	2.34	0.01	12.2		Pr ₂ O ₃	
Y	Stable	-	-	4.9		2783	Elemental Y	Internal hazard to
-90	64.5h	1.2	0.9	0.002			converting to	GI tract
-91	58d	53	52.4	2.2			Y ₂ O ₃	
-92	3.6h	52.6	0.5	0.006	7.1			
Nd	Stable	-	-	39.9	40.2	3090	Elemental Nd	Internal hazard to
-147	11.3d	23.2	20	0.3			converting to	GI, liver, and
							Nd ₂ O ₃	lung
La	Stable	-	-	12.9		3370	Elemental La	Internal hazard to
-140 ^f	40h	54	36	0.1	13.0		converting to	GI tract
							La ₂ O ₃	
Ce	Stable	-	-	23.9		3470	Elemental Ce	Internal hazard to
-141	32d	55.3	55.3	1.8	35.2		converting to	liver and lung
-143	33h	54	33	0.053			Ce ₂ O ₃	
-144	290d	30.0	30.0	9.5	36.8			
Zr	Stable	-	-	43.5	45.9	4325	Elemental Zr	Internal hazard to
-95	63d	53	53	2.4			converting to	GI tract, total
							ZrO ₂	body and lung
Nb	Stable	-	-	0.0		4930	Elemental Nb	Internal hazard to
-95m	90h	0.5	0.5	0.001			converting to	GI, bone, lung,
-95	35d	53	53	1.3	1.3		Nb ₂ O ₅	total body

- a. This table is an expansion of a similar table published by J. R. Beattie in "An Assessment of Environmental Hazard from Fission Product Releases," AHSB(S)R-64 (May 16, 1961).
- b. Assumed thermal neutron flux, 5×10^{12} neutrons/cm².
- c. See J. O. M. Bockris, J. L. White, and J. O. Mackenzie (eds.), "Physicochemical Measurements at High Temperatures," Appendix VI (By J. L. Margrave), Academic Press, New York, 1959; also see A. N. Nesmeyanov, "Vapor Pressure of the Chemical Elements" (R. Gary, ed.), Elsevier, New York, 1963.
- d. Chemical states of released fission product elements are affected by temperature and by composition of the ambient atmosphere during and after release. Those shown in the table are selected to represent most probable chemical states during typical accident conditions.
- e. 38.0 kilocuries of I-132 are produced by decay of Te-132 in the reactor. Te-132 released in an accident will produce more I-132.
- f. See Ba-140.

Table B.3. Classification and Melting Point of Fuel Components and Mixtures

Fuel Component	Composition ^a (wt %)	Melting Point ^b (°C)	Reference
Class I: Metals and Alloys			
Uranium		1133	d
Uranium-aluminum eutectic	87-13	640	
Uranium-molybdenum	90-10	~ 1150-1250	e
Uranium-zirconium	98-2	1150	e
Uranium-niobium	10-90	2400	f
Uranium-thorium eutectic	72-28	1370	e
Uranium-plutonium	95-5	1070-1100	g
Plutonium		640	g
Plutonium-zluminum	3.7-96.3	650	g
Thorium		1750	h
Uranium-zirconium hydride	8-92	1170-1300	e
Zircaloy-2 (cladding material)	98.5 Zr-1.5 Sn	~ 1850	e
		659	
Class II. Oxides Dispersed in a Metallic Matrix			
Aluminum-UO ₂		659	d
Aluminum-U ₃ O ₈		659	d
Stainless steel-UO ₂		1370-1425	e
Michrome-V-UO ₂		~ 1390	e
Class III. Oxide Fuel Materials			
Uranium dioxide ^c , UO ₂		2860	i
Uranium oxide, U ₃ O ₈		2860 (UO ₂)	j
Plutonium dioxide, PuO ₂		2280	k
Thorium-uranium dioxide (solid solution)	93.6-6.4	~ 3200	e
Uranium-plutonium dioxide (solid solution)	95-5	2700	l
Uranium oxide-beryllium oxide	5-95	2150-2500	j
Beryllium oxide, BeO		2550	d
Thorium dioxide, ThO ₂		3300	e

Table B.3 (continued)

Class IV. Carbides in Graphite or Pyrolytic Carbon

Uranium monocarbide, UC	2500	m
Uranium dicarbide, UC ₂	2480	m
Thorium monocarbide, ThC ₂	2625	m
Thorium dicarbide, ThC ₂	2655	m
Plutonium monocarbide, PuC	1654	k
Plutonium dicarbide, PuC ₂	2250	k
Zirconium monocarbide, ZrC	3535	m
Niobium monocarbide, NbC	3500	m
UC-UC ₂ (eutectic)		
UC ₂ -C (eutectic)	2450	m
ThC ₂ -C (eutectic)	2500	m
ZrC-C (eutectic)	2430	m
NbC-C (eutectic)	3250	m

Class V. Miscellaneous Compounds

Uranium nitride, UN	2850	n
Uranium monosulfide, US	2460	n
Uranium disulfide, US ₂	1680	
Thorium nitride, ThN	2630	

Class VI: Fuel Systems Liquid at Reactor Operating Temperatures

NaF-ZrF ₄ -UF ₄	53-40.5-6.5	530	e
LiF-BeF ₂ -ZrF ₄ -UF ₄	65-29.1-5.0-0.9	450	o
Pu-Fe	90.5-9.5	411	k

- Compositions for Class VI are given in mole %.
- Melting points given for Class II are melting point of matrix.
- A wide range of values for the melting point of UO₂ is found in the literature. The value given here is believed by the authors of this chapter to be accurate within the uncertainty assigned by the investigators (+ 45°C or + 81°F). The melting point of irradiated UO₂ has been reported (11) to vary with burnup from 2790°C at zero burnup to a maximum of 2930°C at 0.09 at. % burnup and then to decrease to 2760°C and 2660°C at 8.4 and 11.3 at. % burnup, respectively.

(Footnotes continued on next page).

Table B.3 (continued)

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