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SODIUM BURNING AND AEROSOL RELEASE - AN EVALUATION
OF THE STATE OF THE ART

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Abstract

In a sodium-cooled reactor, the possibility of sodium fires and the resultant release of aerosols strongly influence containment vessel design and operating safety criteria. Qualitative investigations of sodium pool fires and spray fires have been carried out in conjunction with each major sodium-cooled reactor project. Recently, more quantitative studies were undertaken in these areas, and the problem of explosive injection of sodium sprays into containment volumes was investigated. Work is also under way to characterize aerosols released from sodium fires and to define their performance in the containment volume. This document briefly summarizes the work to date on sodium fires and aerosol release, reviews the scope of current programs, and then proposes additional effort to provide safety criteria for liquid-metal-cooled fast-reactor design.

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FOREWORD

This evaluation of the "state of the art" in the field of sodium burning and aerosol release investigations was prepared in the early part of 1967 at the request of the Liquid-Metal Fast Breeder Reactor Office at Argonne National Laboratory as background material for their evaluation of the research and development requirements in one area of fast reactor safety. An attempt was made to recognize all related work that had been performed up to that time, to assess work then in progress, and to recommend additional required effort beyond that being planned.

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CONTENTS

	<u>Page</u>
Abstract	1
Studies to Date	8
Pool Burning and Model Development	9
Ignition Effects	11
Burning Rates	12
Sodium Oxide and Fission-Product Release	14
Conclusions and Recommendations	15
Spray Burning and Model Development	18
Ignition Effects	18
Pressure Rise in Enclosures	20
Explosive Injections	21
Shock-Wave Formation	23
Conclusions and Recommendations	24
Aerosol Production, Transport, and Control	26
Theoretical and Experimental Program	27
Conclusions and Recommendations	28
References	30

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The introduction of sodium-cooled reactors and the resulting large-scale use of sodium has engendered continuing interest in the problems of sodium fires and attendant particulate-matter release. Each major sodium-cooled reactor project has sponsored at least a minimum investigation of the effect of sodium fires on reactor containment criteria, and recent plans to develop the technology for a liquid-metal-cooled fast breeder reactor (LMFBR) created a need for definitive design information. As a preliminary step in the LMFBR program, it is desirable to review the state of the art with respect to sodium burning and aerosol release. To this end, the available literature on investigations in these areas in support of liquid-metal-cooled reactor projects throughout this country and abroad has been reviewed and the pertinent facts summarized.

Sodium fires can be subdivided into those fires characterized by combustion of a sodium pool and those due to a leak or rupture that produces a burning spray. A special case of the spray fire is one in which sodium is introduced explosively into an oxidizing atmosphere.

Pool fires normally result from the accumulation of sodium on flat or recessed surfaces following either the rupture of sodium-containing equipment or the development of spray-producing leaks. The pool fire is characterized by a slower burning rate than that of a spray fire because of limited access of oxygen to the reaction zone. The rate of energy release is normally low, and extremes of temperature and pressure do not develop in an enclosed volume containing such a fire. Of major concern in pool fires involving a reactor coolant are the transport of sodium activity in the smoke from the fire and fission-product transport if conditions responsible for the fire have also caused failure of the fuel cladding.

The problem of damage to equipment by burning sodium or by corrosion products in fire residues is also serious. Sodium fires are characterized by dense sodium oxide aerosol evolution, and subsequent hydrolysis of this material to sodium hydroxide by atmospheric moisture can produce a widely spread corrosive medium. Therefore, fire- and smoke-suppression techniques are always of major interest. To date the standard technique in sodium-

cooled reactor design for suppressing burning rates and limiting smoke evolution has been to place all sodium-containing equipment in controlled atmosphere cells; that is, cells in which the oxygen concentration is controlled to below normal values.

The spray fire has all the problems associated with the pool fire; however, the problems are compounded by the increased reaction rate that results from more intimate contact between the sodium and air. The rate of energy release in a spray fire is a function of the reactant temperatures, the spray energy or degree of dispersion, the humidity, the oxygen concentration, and the geometry of the gas volume. The specific case of a spray fire that involves the explosive introduction of sodium into a surrounding atmosphere is of particular concern because the extremes of temperature and pressure would be expected in this case. There is experimental evidence¹⁻³ 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.⁴ Magnification of pressure pulses imposed on containment vessels by this mechanism must be considered.

Primary concern with sodium-air reactions traditionally has been expressed by those concerned with reactor design and hazards analyses. However, as the liquid-metal-cooled fast breeder program develops, the requirements for large component test facilities are sure to grow. Already, planning is under way for a pump test facility that will involve piping and components larger than any in use today. It is certain that these facilities will also generate requirements for design information on sodium fires and fire control. Although the absence of fission products makes the problem much less complicated, the designer of these facilities must still be concerned with the problems of fire suppression, minimizing equipment damage, and recovery and recycling of a large quantity of sodium.

Studies to Date

Early in the history of sodium-cooled reactors, a continuing series of research programs for investigating effects of sodium fires was begun.

In association with the Naval Reactors Program, Mine Safety Appliance Company⁵ and Knolls Atomic Power Laboratory⁶ 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 Associates⁷ and the Detroit Edison Company^{8,9} ran a series of injection tests in a 20-ft³ test chamber to evaluate sodium-injection effects 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 with a 70-ft³ test chamber.¹ These tests produced the most severe pressure surges that have been noted experimentally. Argonne National Laboratory recently conducted a foreshortened program¹⁰ with 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. 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 currently being 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 rates,¹¹⁻¹³ sodium oxide and fission-product release rates,¹³⁻¹⁶ and the characterization of aerosols released from fires.¹⁴⁻¹⁷

Pool Burning and Model Development

The burning of sodium from a molten pool and the accompanying release of energy, oxide fumes, and fission products are of primary concern in safety studies of sodium-cooled reactor systems. A recent consideration¹⁸ of design concepts for this type of reactor 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 system 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 facility covers the entire system. The primary containment vessel can be filled with inert gas.

Arrangements 1 and 3 are of specific interest, since they are currently under consideration for use in 1000-Mw(e) fast reactor designs. Arrangement 2 must be considered as an advanced proposal for application when the safeguard requirements for sodium-cooled reactors are thoroughly understood.

Based on these possible reactor containment arrangements, the following possible types of accident were formulated.¹⁸

1. Leakage of Sodium into a Cell Containing an Inert Atmosphere

Because of 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 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.

2. 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 (1) 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.

3. Direct Release of Sodium into the Containment Building Air Atmosphere

A reactor with 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 evaluation for EBR-II⁹ and FARET,¹⁰ although the former uses the first arrangement and the third arrangement is proposed for the latter. Again, both initial injection conditions and resultant accumulation and burning of sodium pools must be considered in evaluating the consequence of the accident.

With these accident models in mind, it is well to evaluate the work that has been done to date in the area of pool fire research. Most of the work has been done at Atomics International; and in support of that program, an excellent summary of sodium fires technology up to 1960 was presented by Droher and Gracie.¹¹ Some early tests were conducted by Atomic Power Development Associates⁷ in support of the Fermi reactor, and some related research has also been carried out by British investigators.²⁰⁻²²

Ignition Effects

Early British tests²⁰ indicated that the ignition of a sodium pool in the presence of air is a function of the circumstances under which the two reactants are brought together. Sodium that had been heated to approximately 500°F in the absence of air ignited on exposure to air. However, if heated in the presence of air, no ignition occurred until temperatures of 825 to 880°F were attained.

Droher and Gracie reported¹¹ the results of the pool fire tests to determine the minimum concentration of oxygen that would sustain combustion. A sodium charge of 0.2 lb was held at an elevated temperature in nitrogen, and oxygen was gradually admitted. At 800°F, burning nodules

appeared at 6.5 vol % oxygen concentration, while at 1000°F vigorous burning was noted at 4.6 vol %. In both cases, there was smoke formation at lower concentrations. In a subsequent test, the moisture content was increased by a factor of approximately 50 (from 3 grains H₂O per pound of air to 140 grains H₂O per pound of air) with no significant effect on the ignition temperature.

An examination of the effect of water vapor on the reaction of sodium and oxygen was carried out by Longton.²¹ He reported that variations in chemical reactivity of freshly distilled sodium and oxygen gas are directly related to water-vapor concentration. Dry oxygen does not react with sodium at a detectable rate at pressures up to 1 atm and temperatures up to 1000°F, while at a concentration of 0.25-mg H₂O per liter of oxygen at NTP (<1 grain H₂O per pound of air) the reaction proceeds smoothly even below 212°F. It was concluded that there were three distinct modes of reactivity controlled by the partial pressure ratio of oxygen to water, as indicated below:

	$\frac{P_{O_2}}{P_{H_2O}}$
No reaction	$4.5 \times 10^7 - 5.0 \times 10^5$
Ignition only	$4.0 \times 10^5 - 9.0 \times 10^3$
General reaction	$7.0 \times 10^3 - 4.0 \times 10^1$

In a companion study²² of the effect of hydrogen on the sodium-oxygen reaction, Longton concluded that hydrogen gives the same effect as water at one-half the partial pressure (or double the above ratio). Caution is necessary in applying these ratios, since the maximum water partial pressure used in the test program was 0.25 mm Hg (1 to 2 grains H₂O per pound of air at atmospheric pressure). Normal humidity gives water contents in air atmospheres well in excess of this value.

Burning Rates

The rate of combustion of sodium in a pool fire is also of interest. Considerable work has been done to define the problem and provide a basis

for the energy-release calculations used in hazards analyses. The British derived a theoretical burning rate of $4 \text{ lb/hr}\cdot\text{ft}^2$ for use in Dounreay containment vessel design.²³ Garelis and Nims⁷ conducted an experimental investigation of the problem and determined a burning rate of $4.9 \text{ lb/hr}\cdot\text{ft}^2$ for an enclosed sodium pool initially at 675°F when fed with approximately 100% excess air. In a subsequent test in open air, sodium that was initially at 850°F burned at a rate of $6.3 \text{ lb/hr}\cdot\text{ft}^2$.

A broad investigation of this problem is continuing at Atomics International. Early tests were qualitative, but attempts at quantitative definition of fire variables are now under way. Burning rates ranging from 15 to $33 \text{ lb/hr}\cdot\text{ft}^2$ were reported¹¹ in early tests with the rate increasing with depth of the pool and decreasing sharply with surface area for low area tests ($<80 \text{ in.}^2$). For larger surfaces, little effect of area on burning rate was noted.

There was considerable variation in reported bulk pool temperature and temperature distribution, surface temperatures, and flame temperatures due to wide variations in test techniques. Droher and Gracie report¹¹ surface temperatures of 1100 to 1250°F in four successive tests with an uninsulated container. With insulation applied, the entire sodium pool reportedly reached 1620°F , which is the atmospheric boiling point. They indicated that at lower burning rates in a depleted oxygen concentration, the insulated pool was at 1100 to 1200°F .

Lauben reports¹² that two insulated containers, one $10 \frac{1}{2}$ -in. in diameter and 18 in. high and the other $10 \frac{1}{2}$ in. in diameter and 36 in. high, produced similar results when 33- and 83-lb charges, respectively, were burned. The liquid surface reportedly was at 1600 to 1700°F , and the bulk pool temperature was 1200°F . Burning rates of 4 to $5 \frac{1}{2} \text{ lb/hr}\cdot\text{ft}^2$ were noted in these tests. Also observed was the tendency for oxides to sink to the bottom of the sodium pool as burning progressed and to accumulate gradually until they met the receding liquid surface and extinguished the fire.

Lauben further reports¹³ that burning rates cannot be properly evaluated without analysis of the fire residue. In nine tests, sodium charges of 35 to 170 lb were burned in 12- to 15-in. pots at rates of 1.4 to 8.4

lb/hr·ft². Subsequent analysis of the residue indicated that unburned sodium was not present to the extent thought and that the burn rates probably should be increased by a factor of 2.

In the most recently reported test,¹³ Lauben used a 2-ft-diam by 4-ft-high insulated vessel containing 615 lb of sodium. A burning rate of 6.2 lb/hr·ft² (based on pool-depth change) was measured between the 2nd and 17th hours, and this figure compared favorably with the rate based on analysis of the effluent oxygen concentration. The temperature measured near the pool surface with an optical pyrometer was 1600°F, and a thermocouple indicated a temperature of 1800°F a short distance above the surface. This fire (LF-1) and a recent fire (IF-10) in another facility with an 83-lb batch of sodium both showed similar burning rates (6 lb/hr·ft²) and similar surface temperatures in the pool (~1300°F).

Sodium Oxide and Fission-Product Release

Another major consideration in hazards evaluation 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 Koontz reported¹⁴ sodium burning experiments in a 40-ft³ chamber in which they used small sodium batches. In three experiments, release fractions of oxidized sodium ranged from 0.25 to 0.43. Burning of the sodium charge was incomplete in at least two tests.

Lauben has reported¹³ 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 5 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 falling 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, and a decrease from 360 to 12 ppm was noted. This indicated that the iodine had been selectively removed from the sodium, probably by absorption on sodium oxide. A supporting experiment was carried out in which 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.

Lauben also reported¹³ on oxide and iodine release measurements from the 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 Baurmash¹⁵ 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 ratios of iodine to sodium in the liquid and vapor phases were equal.

Lauben and Larson,¹⁶ in a continuing series of tests in the modified large fires apparatus (LF-2), found that iodine release showed temperature dependence. With a burning pool surface temperature of 1300°F, the released sodium contained iodine in one-half the concentration of the sodium in the pool. With a surface temperature of 1000°F, the released phase concentration was about one-fifth the concentration of the pool.

Conclusions and Recommendations

In spite of a lack of standard test techniques, the literature provides a few general conclusions applicable to pool fires. Pool fires in insulated burning pots tend to give burning rates of 4 to 6 lb/hr·ft². Initial burning rates are generally higher than average burning rates, discounting the period necessary to establish the equilibrium temperature profile in the system. A poorly defined temperature structure exists in the combustion zone. At the surface, temperatures from 1300 to over 1600°F

have been reported. Precise temperature measurement at this location is difficult, but some of the confusion in the literature appears to reflect differences in terminology. The flame temperature a short distance above the surface approximates 1800°F. Provision of an atmosphere containing no more than 4 vol % oxygen concentration will prevent ignition and sustained burning of a sodium pool. Oxides produced in the fire sink to the bottom of the burning pot and accumulate until the sodium surface recedes below the oxide level and the fire stops. Although measurements of release fractions of sodium oxide from the 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 to 40 percent. Iodine release fraction measurements are generally inconclusive at this time.

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 and his associates¹⁸ 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 currently in use for the characterization of aerosols released from fires and for the determination of activity, mass, and particle-size distribution. A laboratory test chamber is currently 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. The large fires apparatus is being used for burning rate and release rate studies, with the primary objective of obtaining an improved understanding of reaction region mass and energy release and transport. Preparations are being made for installation of an improved apparatus for modeling fires. This apparatus will consist of a burning pit simulating an equipment cell and a flexible dual chamber arrangement that can be mounted on the pit and arranged to simulate various interconnected primary and secondary containment volumes. Analytical models for mass and energy flow

throughout this enclosure arrangement are being formulated, and experimental verification is planned.

This coordinated program aimed at well-defined goals represents a significant milestone in sodium fires research. The excellent work done in the past has helped to define in a qualitative way the course of the future program. More is now known about instrumentation problems that must be solved. Many experimental techniques have been tried, and those that are generally most suitable are now evident. Problems with equipment have been uncovered, and appropriate modifications are being made.

The following recommendations are in order concerning the pool fires research program currently centered at Atomics International:

1. The five-year program¹⁸ presents a satisfactory outline of required research into the problem of sodium fires, mass and energy release and deposition, and modeling requirements for sodium-cooled and analytical techniques that will implement this plan and permit definition of sodium burning rates, sodium oxide release rates, and fission-product release rates. It is important that conditions existing in a sodium fire be completely defined so that energy and mass-balance calculations on the complete containment complex can be made with confidence during the course of reactor hazards analyses.

2. Further emphasis should be put on the definition of an experimental program for accident mitigation investigations. Techniques for reducing energy release have been suggested,¹⁸ such as the use of deep pits to reduce surface areas of sodium exposed to air and provision of in situ heat sinks to cool the released sodium. These concepts should be expanded while keeping in mind the requirements of nonnuclear, large-scale component test facilities. Of particular concern in this latter case are techniques that increase the probability of convenient recovery and reuse of large-scale spills.

3. Work to date has centered on static sodium pools. Because of the temperature structure, this geometry is basically free of strong convection currents. Consideration of a reactor system wherein the core and primary components are submerged in a large pool of sodium gives rise to the possibility of a fire on the surface of a sodium volume that

is being agitated by an internal heat source. The problems introduced by this case should be evaluated and any necessary experimental work incorporated into the program.

4. A comprehensive summary should be prepared of all sodium fires tests at Atomics International since 1960. This document should present the current and best interpretation of test results to date in order to supplement the summary issued by Droher and Gracie¹¹ at that time.

Spray Burning and Model Development

In considering spray fires, it is necessary to keep in mind the same reactor containment models and types of sodium leak as those introduced in the above discussion of pool burning. The discussions that follow summarize the work to date on spray fires and the special case thereof involving explosive injections into the containment volume. Because the development of adequate containment vessel design criteria prompted most of these investigations, the techniques used were oriented toward the system design of interest and the results are less fundamental than is desirable for broad application. However, many of the observations are generally useful in guiding preliminary design studies, and they form a useful base for future work. Droher and Gracie presented an excellent summary¹¹ of spray fire research up to 1960. More detailed information on much of the early work can be extracted directly from their document.

Ignition Effects

Ignition of a sodium pool when exposed to air has been shown to be a function of many variables. In the case of spray fires, the primary variables affecting ignition are oxygen content of the atmosphere and droplet size distribution. Cowen and Vickers²⁰ 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 at as low as 250°F, irrespective of moisture content in the test

environment. Other tests with moist and dry nitrogen resulted in no ignition.

Droher and Gracie reported¹¹ on tests in which 1000°F sodium was sprayed into an oxygen-depleted atmosphere. Most of the tests were conducted at a moisture content of 3 grains per pound of air. Some smoke was produced in 0.5 vol % oxygen concentrations, and incandescence of sodium was noted at 5 vol % oxygen. When the test was repeated at a moisture content of 210 grains per pound 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) were observed to ignite at room temperature in air, while drop-size particles ignited just above the melting point.

In tests conducted by Nelson and Nelson,⁶ 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 that was 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 (~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 with 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 evaluation²⁴ used 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 in the containment vessel of 30 psig 6 min after the start of burning of a pool

filling the entire basement area of the equipment cells. Spray fires were not considered because of the protective atmosphere enclosing the coolant system.

Pressure Rise in Enclosures

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 worse case, produce a sodium pool fire having less severe and more predictable time-dependent pressure and temperature characteristics. Tests of spray fires in air and controlled atmospheres have generally defined this requirement, but no systematic study of the spray fire has been made to date.

Mangold and Tidball⁵ conducted tests with a hemicylinder 26 ft in diameter and 15 ft long ($\sim 3900 \text{ ft}^3$). Eleven tests were carried out in which 30 to 200 lb of sodium were injected at rates of approximately 2 lb/sec. Oxygen levels were varied from normal air concentrations to 2.75 vol %. These tests indicated that control of the oxygen concentration in the enclosure and cooling of the chamber walls were very effective in limiting the fire. The maximum pressure encountered caused relief valves set at 13.3 and 15.3 psig to be opened. Most of the heat generated by combustion of the sodium was transmitted to the chamber walls. Maximum gas temperatures measured did not exceed 300°F.

Similar tests were conducted by Hines, Gemant, and Kelley^{8,9} in a cylindrical chamber 2 1/2 ft in diameter and 4 ft long ($\sim 19 \text{ ft}^3$). Four significant tests were reported in which oxygen was varied from normal air concentration to 0.6 vol %. Sodium was injected at 850°F. The maximum pressure encountered during these tests was 38 psig for the case of sodium burning in normal air. The maximum pressure decreased as the oxygen content and the injection pressure decreased. A theoretical time-dependent calculation was proposed that gave results in general agreement with the experimental measurements.

Further work along this line was carried out by Droher and Gracie¹¹ with a 3-ft by 4-ft by 4-ft box (48 ft^3) equipped with a relief valve set at 4 psi. The injection tests were made with 1/2-lb batches of

sodium generally at 1100°F. The sodium entered the box through a 1/4-in. tube that had been flattened to give an oblong opening 1 to 2 mils wide. This series of tests was designed to examine the effect of oxygen concentration in the test chamber between 0 and 5 vol % at a constant low value of humidity, injected sodium temperatures over the range 600 to 1200°F, injection pressures ranging from 30 to 60 psi, and moisture contents from less than 3 grains per pound of air to 300 grains at a constant oxygen concentration of 3 vol %. A brief test was also carried out on the ability of impingement baffles to reduce the mixing of sodium with air and thus reduce the resulting pressure.

These tests showed that both the maximum pressure achieved and the rate of pressure rise increased with oxygen concentration in the test chamber and with the temperature of the injected sodium. No significant effect of injection pressure was noted over the test range, although it was generally true that factors increasing the rate of admission and the degree of dispersion of the sodium led to increased rates of pressure rise. The rate of pressure rise was a direct function of the humidity in the enclosure, and it was shown that the use of a baffle to interrupt sodium travel minimized the pressure increase.

Explosive Injections

A specific case of a spray fire is that associated with the explosive injection of sodium into an enclosure. Two series of tests were run at Argonne National Laboratory to provide information for EBR-II and FARET hazards evaluations. Twenty-seven explosive injections were carried out by Humphreys¹ with the hydrogen-oxygen reaction as the driving force for the injection. Twenty-five of these tests were conducted with air in the test vessel and two with argon. The quantity of sodium varied from 200 to 1600 g, and it was normally injected at 750°F. Humphreys reports that he achieved experimental pressures that were approximately two-thirds of the theoretical maximum in some tests. Subsequent calculations by Argonne National Laboratory workers^{2,3} indicated that theoretical maximum pressures presented by Humphreys were probably high due to his assumption of complete reaction of the sodium and that, if thermodynamic limits on

the sodium-air reaction are considered, the theoretically maximum pressures correspond quite closely to the experimental results achieved by Humphreys in some cases. These computations also indicate a peak pressure in dry air of 133 psig at a sodium-to-oxygen molar ratio of 7. In air saturated with moisture at 25°C, the peak pressure of 122 psig occurs at a sodium-to-oxygen molar ratio of 6.

Grosse and Conway²⁵ also pointed out the importance of considering the dissociation of oxides in calculating thermodynamic limits on reactions. They indicate a maximum temperature of 2000°K for the adiabatic combustion of sodium to sodium oxide (Na_2O) in oxygen at 1 atm pressure. This is in general agreement with the ANL computations, which give a peak temperature of 2260°K for the sodium-dry air reaction at a sodium-to-oxygen molar ratio of 5.

In Humphreys' test, peak pressures in the reaction chamber were achieved in 30 to 200 msec after the start of injections, with the time increasing as the size of the injected sodium batch was increased. Maximum pressures occurred at sodium-to-oxygen molar ratios of 2.54 and 3.05. Although Humphreys' theoretical calculations indicated that maximum pressures should continue to increase until the sodium-to-oxygen molar ratio reached 4, the experimental results did not show this trend. This decrease in experimental yield at the higher molar ratios was attributed to a decrease in injection efficiency and to higher heat capacities in the system. Little effect was noted on the reaction when injection energy was varied. Humphreys attributed this to a secondary dispersion mechanism - a mild explosion triggered by the initial sodium reaction at the injector location, which caused progressive subdivision of the sodium due to reaction-induced turbulence. It was thought that the presence of water vapor contributed to improving this secondary dispersion and to increasing the yield from a test.

Subsequent small-scale experiments carried out by Smith¹⁰ at Argonne National Laboratory employed two types of reaction vessels, one a series of 159-in.³ right-circular cylinders having different height-to-diameter ratios and the other a 20-in. cube. This brief test program was based on the explosive introduction of small quantities of sodium at 1200°F

into air at injection velocities of approximately 100 ft/sec. The sodium-to-oxygen molar ratio was maintained at 4. Six of the tests carried out were recorded by high-speed cinematography.²⁶ For these tests, 0.5- to 2.5-g quantities of sodium were used, and the maximum pressure noted was 28 psi. The film taken during the course of these tests revealed generally poor dispersion of the injected sodium. This tendency toward poor dispersion is supported by Smith's observation that higher pressures were encountered in the smaller diameter test chambers. Such a result would be expected with axial injection and poor dispersion.

Shock-Wave Formation

A theoretical analysis was carried out recently by Moore⁴ on the effects of shock waves due to a sodium-air reaction following explosive introduction of sodium into a containment vessel. In this study, examinations were made of three proposed models applicable to a reactive spherical volume contained within a larger, totally enclosed spherical volume. The simplest and most pessimistic model was based on instantaneous combustion throughout the reactive volume. The second model had an advancing flame front in the reactive volume. The third model was based on uniform combustion at a finite rate throughout the reactive volume.

Results from models 1 and 3 are of primary interest, since model 2 produces only mild shock conditions. Pressures generated by shock waves at the enclosure wall were compared with the enclosure pressure that would result from adiabatic combustion of sodium over a range of sodium-to-oxygen molar ratios up to the stoichiometric ratio to produce Na_2O . Since the tests by Humphreys, as later interpreted by Argonne National Laboratory, indicate that an approach to this latter condition is experimentally possible, this comparison is of interest.

The "instantaneous-combustion" model produced pressures on the containment wall due to shock-wave impingement that were about double the adiabatic pressure. The "uniform-combustion" model predicts pressures at the wall only slightly greater than the adiabatic case over part of the range and slightly lower over the remainder of the range. For a

100-ft-diam containment vessel, the crossover point occurred when 600 lb of sodium was burned, with slightly higher pressures being predicted as the quantity of sodium was reduced. The model was not considered effective when more than approximately 1700 lb of sodium was burned in a containment vessel of the same diameter.

Moore⁴ recognized that for containment vessel design, the instantaneous combustion model would present pessimistic results, but he believes that it can be used when considering small reactive zones within a larger containment volume. In the case where the reactive zone begins to approach the size of the containment vessel, he recommends the use of the uniform-combustion model for the range where it is applicable.

Conclusions and Recommendations

Little of the work that has been done on spray fires to date is basic. Most of the effort has been supported by specific reactor programs and has had as its main objective solution of the design problems of the reactor involved. To date, a generally similar approach to the solution of the hazards of sodium leakage and spray fires has been adopted. Thus, most of the work performed has been usefully applied to each subsequent reactor design, and a gradually increasing technology has developed.

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 primarily a function of the size of the leak and the surface area of burning sodium.

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 system 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.

Sodium spraying into enclosed spaces containing oxygen 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 become less severe in that a 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 that would be created by simple temperature and pressure equilibrium 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.⁹ Such results are expected, since the basic mechanism is a time-dependent equilibrium 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 this case.⁹ However, the model is quite basic, and further refinement appears justified.

The following additional effort is required to permit reasonable definition of safety questions associated with spray fires:

1. Explosive Injections into Enclosures

- a. Extend the test program on explosive injection of sodium to cover representative cases of enclosure compartmentalization.
- b. Investigate analytically and experimentally the possibility of shock-wave generation due to the combustion of sodium in air and in atmospheres depleted in oxygen.
- c. Develop a model for explosive injection of sodium that will permit calculation of pressure and related transients necessary for containment vessel design. This model should be experimentally verified.

2. Spray Injection into Enclosures

- a. Experimentally determine the burning rate in spray fires as a function of the primary variables - particle size, oxygen concentration, injection energy, humidity, and sodium temperature.
- b. Investigate analytically and experimentally the heat transfer processes at work in a spray fire in representative geometries. Of particular interest are the radiative heat-loss processes that pertain.
- c. In conditions where explosive injections to the containment system are considered incredible, the spray fire could be the controlling consideration in containment vessel design. Therefore a model is needed for spray injection of sodium that will permit calculation of resulting pressures, temperatures, and related transients. The model should be experimentally verified.

Aerosol Production, Transport, and Control

In considering fast reactor safety, it is theorized that severe accidents could 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.

The sodium-cooled reactor program at Atomics International was responsible for the early formulation of a computer code to allow the

treatment of energy and mass transport in a containment vessel during a sodium fire. This code, SOFIRE, has continued in use and is being improved in degree of sophistication and range of applicability. However, much of the theory needed to predict the specific consequence of a severe accident is not available for incorporation into the code. The pool burning experimental program at Atomics International, which has been discussed previously, is currently producing much information of interest, and their proposed five-year program¹⁸ for characterization of sodium fires and fission-product release should go far toward providing the necessary analytical techniques and experimental verification.

Theoretical and Experimental Program

One major phase of this five-year program is concerned with aerosol production 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 four papers presented at the Ninth AEC Air Cleaning Conference of September 1966.

Lauben and Larson¹⁶ discussed the theoretical aspects of the sodium-oxygen reaction region heat and mass transfer. Pertinent experimental results presented have been mentioned previously.

Koontz, Nelson, and Baurmash¹⁵ discussed a laboratory device that is 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 device that has been developed will be an extremely useful tool in subsequent fundamental investigations of aerosols.

Nelson, Baurmash, and Koontz¹⁴ discussed the laboratory equipment that is being used in their aerosol studies and some of the early results.

A 40-ft³ chamber is available in which 2- to 6-in. burning pots can be installed. Instrumentation is available for monitoring temperature throughout the volume, the oxygen content of the contained gas, and the aerosol plateout 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 plateout 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 agreement between the electrostatic precipitator and the modified impactors, as was the case in work with the standard aerosol generator previously described.¹⁵ This problem must be eliminated before work on fission-product aerosol characterization can proceed.

Spiegler and his associates¹⁷ developed an analytical treatment of the processes involved in aerosol generation, agglomeration, and settling. The results from computer solutions of the pertinent equations are being compared with experimental results from the tests discussed above.

Conclusions and Recommendations

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 an analytical model that completely characterizes sodium fires and aerosols produced by these fires. As indicated before, rapid progress is being made at Atomics International in developing definitive experimental techniques for the controlled investigation of sodium fires. The work on aerosol characterization, as indicated above, is off to a good start. 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. This course is essentially the one outlined in the

five-year program for sodium fires research,¹⁸ and it is recommended that the work scoped by this document be continued.

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