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# Evaluation of Materials Interactions for Advanced Reactor Systems

Annual Progress Report

Prepared by D.G. Swanson, H.L.L. van Paassen

The Aerospace Corporation

Prepared for U.S. Nuclear Regulatory Commission

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#### ABSTRACT

This report consists of consultative support on a variety of topics related to the materials interactions that would occur following postulated core meltdown accidents in Light Water Reactors, Liquid Metal Fast Breeder Reactors and Gas-Cooled Reactors. The main topics that are addressed include (1) evaluation of the Floating Nuclear Plant core ladle for delaying the melt-through penetration of a molten core; (2) review of recent experimental results on the inveraction of molten UO<sub>2</sub> with magnesium oxide; (3) considerations in retrofitting existing reactors with core retention systems; (4) review of the Cas-Cooled Fast Reactor postaccident fuel containment program; and (5) ceview and evaluation of recent experimental information in the area of sodium-concrete reactions.

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#### I. SUMMARY

#### A. THE FLOATING NUCLEAR PLANT CORE LADLE (FNP)

A properly designed core ladle constructed from refractory materials such as MgO, as compared to concrete, can minimize gas evolution and reduce the rate of molten core penetration. Desirable characteristics of a core ladle include a high melting point, a high specific heat and a large heat of fusion. The thermal conductivity should be low, and the refractory should be miscible in molten fuel. Chemical compatibility, limited gas evolution and low cost are also desirable. The MgO core ladle proposed by Offshore Power Systems (OPS) for the FNP meets these requirements.

Controllable problems that will probably be encountered during the use of the proposed MgO core ladle include preaccident stability in a moist atmosphere and low density which could lead to brick floatation. The first problem can be controlled by the proposed steel shell that will encapsulate the MgO and through the use of moisture monitors that will warn when corrective action is required. Brick floatation can be controlled by the use of an inverted arch with a staggered bricking arrangement and a double keylocking tongue-in-groove design as proposed. It is not felt that thermal or mechanical shock are problems with modern MgO refractories.

The only potential disadvantage of MgO is its susceptibility to attack by iron oxide slag. A lack of experimental data for the interactions between iron oxide slag and MgO at high temperatures makes an assessment very difficult. Some relevant steel industry experience does exist; however, temperatures of 1650°C are employed in the steel industry, with an indication of the presence of 2550°C iron oxide particles in contact with the MgO liner in a basic oxygen furnace unit. In this temperature regime, attack by slag is not a problem in the industry's experience. At the postulated 2850°C temperatures in a core meltdown event, the rate of slag line attack will initially be much greater than at 1650°C until the slag is saturated with MgO. At temperatures between 1650°C and 2850°C, the amount of MgO needed to saturate the slag will increase as indicated by the iron oxide-MgO phase diagram. The extent of the problem will be determined by the temperature of the system and the quantity of slag as compared to the amount of MgO available.

Several more positive points sould be made, on the basis of metals industry experience, concerning the potential use of MgO in the core ladle. In the steel industry there is little concern about the response of modern MgO refractory bricks to mechanical shock. In addition, the MgO liners appear to be reasonably resistant to thermal shock, even when cold. Brick floatation does not appear to be a problem if an inverted arch design is used. It appears that MgO bricks, like the Harklase type, with a low lime concentration, are reasonably resistant to hydration in water. although some contradictory data have recently been developed. Also, with proper design, molten metal interactions with vet MgO bricks should not cause problems.

A hydrocarbon-impregnated brick, TOPEX-S has been proposed by OPS as the top layer of the core ladle to improve its thermal shock resistance. TOPEX-S brick generates hydrocarbon gas when heated and, consequently, this area requires further evaluation as to its suitability for use in the core ladle.

Mg0 or ZrO<sub>2</sub> will also be used to line the upper walls of the reactor cavity of the FNP. Concern has been expressed that the concrete may disintegrate if exposed to high temperatures, but a vertical, nonstructural wall of concrete of the height proposed will support its own weight at temperatures up to 900°C.

In conclusion, MgO seems to be the best material for the core ladle application, despite its slag attack problem. If the amount of available MgO is sufficiently large compared to the slag layer, the slag layer will eventually be saturated with MgO. None of the other materials suggested for this application appear any more promising than MgO. The core ladle proposed by OPS seems to meet the necessary core retention objectives with the exceptions noted previously and should be successful in delaying penetration by a molten core into the environment.

#### B. RECENT UO2-MATERIALS INTERACTION EXPERIMENTS

Recent experiments conducted at Sandia Labor tories in which gas evolution was observed from MgO brick heated alone at  $2750^{\circ}$ C, and from UO<sub>2</sub> and MgO heated together at 2600°C, indicate a possible detrimental reaction. In each case the specimens were heated on a tungsten grid. In our opinion, the gas observed is a consequence of a reaction between tungsten and MgO forming tungsten exides. This reaction has also been reported by other experimenters. We do not believe that the gas is being generated by a reaction between UO<sub>2</sub> and MgO, as some have suggested. An analysis of gas specimens in future planned tests at Sandia may resolve the question.

#### C. RETROFITTING EXISTING REACTORS WITH CORE RETENTION SYSTEMS

Materials were examined that could be laid in as bricks or poured in granular or chunk form onto the reactor cavity floor to ease the problems associated with retrofitting the containment building with a core retention device. The material property considerations included the need for high density to prevent floatation, a high melting point and a high boiling point to preclude vapor explosions. Other considerations were toxicity, fabricability and cost. It was concluded that high density materials, such as TaC, would probably be effective in core containment. However, the cost of the materials may be prohibitive, and extensive development may be required to develop a manufacturing technology. Another alternative, monolithic high alumina cement, probably represents an improvement over basalt concrete, but its melting point is too low to provide significantly greater penetration resistance. When cost, penetration resistance and gas generation are considered, MgO continues to look more attractive than the other suggested alternatives, providing radiation levels, space, and access problems permit proper installation so that floatation of the MgO bricks will not occur.

#### D. GAS-COOLED REACTOR PROGRAMS

General Atomic has been engaged for a number of years in an assessment of the postaccident containment capability of the prestressed concrete reactor vessel and a consideration of several design concepts for molten core retention. In the current design of the gas-cooled fast reactor (GCFR), it was

felt that graphite blocks would delay arrival of the heat front at the cooling system for ten hours. We feel that some consideration should be given to ways of restraining the graphite blocks to prevent possible floatation by molten core debris, followed by core debris coming in contact with cooling system. The effects of cooling system failure, the fraction of heat transferred downward and the prospects for a reaction between UO<sub>2</sub> and graphite are also unresolved.

Several core containment concepts are being considered by General Atomic. The beavy metal bath concept employs an alloy of uranium and iron with a low melting point. Unfortunately, however, the uranium metal is pyrophoric and is also easily corroded. With a proposed borax bath containment, the low boiling point of borax (1600°C) could be responsible for a vapor explosion if it comes in contact with molten  $UO_2$  at 2750°C.

Before judgments can be made on the interesting ideas under consideration at GA, additional information on the proposed designs is needed, supported by appropriate experiments. The designs in each instance have promising features but all of them could be improved through the addition of an underlying layer of a high temperature, chemically inert, sacrificial material to protect against cooling system failure. In each concept, General Atomic should provide evaluations of the consequences of a cooling system failure and the subsequent performance of materials.

#### E. SODIUM-CONCRETE INTERACTIONS

This area is of prime importance in the safety review of the Liquid Metal Fast Breeder Reactor (LMFBR) and is concerned with the analysis of sodium spill and core melt accidents. Major research programs at two national laboratories, Sandia Laboratories and Hanford Engineering Development Laboratory (HEDL), have generated fairly contradictory results which have raised new questions.

After reviewing sodium-basalt concrete experimental data from Sandia and HEDL, we have reached the following conclusions:

 A sodium-basalt concrete penetration rate of 1 in./hr is recommended. In the absence of firm evidence indicating limits of sodium penetration, we feel that a continuous penetration rate is appropriate.

- 2. Hydrogen recombination seems to be unlikely in significant amounts.
- 3. The firebricks employed in these tests are not a barrier to sodium. Indeed, this particular firebrick-liner combination may be detrimental if it comes into contact with liquid sodium. The pressure generated as reaction products form may cause the concrete to crack. Firebricks that are inert to sodium should be employed instead of the ones selected. Siliceous firebrick should be avoided.
- 4. It is difficult to determine whether basalt concrete is superior to limestone concrete in a sodium environment. Basalt concrete seems to be somewhat more resistant to penetration by sodium, but it also seems more apt to crack than limestone concrete.
- The questions of the effects of scale, geometry and mode of restraint remain unresolved.
- 6. Sodium pool temperature appears to affect the chemical reactions and formation of reaction products. A lower temperature sodium environment (600°C) seems to have a more severe effect on basalt concrete than the boiling sodium environment.
- Sodium pool depth may affect the time required for the occurrence of a reaction and the extent of reaction.
- 8. Dehydration of concrete may affect the extent of sodium penetration.
- The chemistry of the sodium-basalt concrete and sodium-limestone concrete interactions are speculative. Hypotheses have been proposed which seem reasonable but are as yet unconfirmed.
- The physical aspects (cracking, spallation) of the sodium-basalt and sodium-limestone concrete interactions are poorly understood.
- 11. Further research is needed on both the chemical and physical aspects of sodium-concrete interactions. Given the differences in the experimental conditions, it is difficult to accurately correlate the results. More detailed coordination in selecting experiments and experimental parameters for study would be desirable.

#### II. INTRODUCTION

This is an account of work performed for the Office of Nuclear Reactor Regulation of the United States Nuclear Regulatory Commission (NRC). We have reviewed and evaluated materials interaction technical information generated during the course of various safety reviews. The report is one of a series<sup>1,2,3</sup> prepared at The Aerospace Corporation concerned with the consequences of core melt accidents in nuclear reactors.

In the following sections, core-materials-interactions related issues concerned with the Floating Nuclear Plant (FNP) will be reviewed. The discussion includes references to relevant industrial experience from the metals refining industry. Industrial experience constitutes the only knowledge available for the prediction of the expected behavior of large-scale refractory structures. Next, recent molten core debris-materials studies will be reviewed and the problems likely to be encountered in retrofitting existing reactors with core retention systems will be discussed. A number of materials that might be used in the containment of a molten core have been examined.

The report continues with a discussion of materials issues in the gascooled reactor programs. Interactions between molten UO<sub>2</sub> and graphite are considered and several core containment concepts under consideration at General Atomic are reviewed. The latter have promising features, but there are a number of issues that can only be resolved by experimentation.

Sodium-concrete reactions are of considerable interest in the safety review of the Liquid Metal Fast Breeder Reactor (LMFBR) and are discussed at length in the final section of the report. Major research programs at Sandia Laboratories, under the sponsorship of NRC, and at Hanford Engineering Development Laboratory (HEDL), sponsored by the Department of Energy, have been established to study these interactions in a series of experiments stretching over several years. Unfortunately, the experiments at the two laboratories have generated fairly contradictory results and have raised many new questions.

#### III. THE FLOATING NUCLEAR PLANT CORE LADLE

#### A. REVIEW OF THE CONCEPT

In the original specifications for the Offshore Power Systems (OPS) FNP, a four-foot-thick layer of concrete was proposed as shielding below the reactor vessel. In Part III of the Final Environmental Statement (FES-III), the NRC staff concluded that this concrete basemat would be of limited value in containing a molten core in the event of a postulated core meltdown accident. The NRC Staff also concluded that alterations of the concrete shielding pad beneath the reactor vessel would be required to provide increased resistance to melt through in order to mitigate the environmental consequences of an accident. FES-III suggested that replacement of the concrete with a suitable refractory can delay penetration by molten core debris and increase the time available to mitigate the effects of a release into the environment.

#### 1. COMPARISON OF MOLTEN UO2 PENETRATION RATES

Relative ces of penetration of molten  $\rm UO_2$  and steel into concrete and a refractory material, MgO, are indicated in Table I. The molten  $\rm UO_2$  and low-temperature steel experiments were conducted at The Aerospace Corporation<sup>1,2,3</sup> and the high-temperature molten steel data were obtained at Sandia Laboratories. Molten  $\rm UO_2$  was observed to penetrate basalt concrete at a rate of approximately 14 cm/hr, and molten steel at approximately the same temperature penetrated concrete at a rate of nearly 130 cm/hr. The molten steel appears to be more penetrating because of its greater conductivity which results in greater heat transfer into the concrete. As expected, the 1600°C molten steel penetrates concrete at a significantly slower rate. From the limited experimental data available to us, it appears that an eight-foot concrete basemat might contain molten core debris for as little as two hours based on the 4-ft/hr erosion rate observed at Sandia.

Experiments have also been conducted with other refractory materials at The Aerospace Corporation (MgO), Westinghouse (MgO,  $Al_2O_3$ ,  $ZrO_2$ )<sup>5</sup> and at Argonne National Laboratory (MgO, steel, graphite).<sup>6</sup> Table I also lists the

Table I. Comparative Erosion Rates (cm/hr)

MOLTEN MATERIAL	CONCRETE	MGO
UO <sub>2</sub> (2800°C)	14 <sup>a</sup>	0.6 <sup>C</sup>
c∈el (2700°C)	130 <sup>a</sup>	P. A.
Steel (1600°C)	30	~0.

<sup>a</sup>Basalt concrete

<sup>b</sup>Limestone concrete

<sup>C</sup>With quiescent conditions (little stirring)

penetration rates observed in experiments at The Aerospace Corporation with MgO. Molten  $UO_2$  was found to penetrate MgO at a rate of 0.6 cm/hr under quiescent conditions where there was little stirring by the electric arc used to melt the  $UO_2$ . Under circumstances where vigorous stirring by the electric arc occurred, the penetration rate increased to as much as 5.0 cm/hr.

Low-temperature (1600°C) steel had only a very slight effect on MgO with negligible penetration. There is some evidence that molten steel at much higher `emperatures may cause substantial erosion of MgO; this possibility will be considered later.

Substantial gas evolution will also occur when molten core debris comes in contact with concrete. These gases can conceivably overpressurize the containment building. Furthermore, hydrogen generated from water in the concrete potentially accumulates in possibly unsafe concentrations.

From the experimental information available, it is unlikely that concrete will be able to resist attack by molten core debris for very long. It also appears that the molten core penetration rate and the extent of gas evolution can be significantly reduced through a suitable choice of refractory materials.

#### 2. POTENTIAL SACRIFICIAL MATERIALS

A number of high-temperature materials might be used as sacrificial materials in a core ladle. To better appreciate the choice made by OPS, it is useful to consider the alternatives.

While in many cases there is insufficient experimental data to make generalizations, it can be stated that oxides are more likely than carbides, borides and nitrides to be stable chemically in the presence of molten  $UO_2$ . These other materials tend to oxidize, especially in the presence of excess oxygen. In general, oxides have only minor chemical reactions with molten core debris. The oxides typically form eutectic mixtures with  $UO_2$ , which have melting points below that of either pure constituent. The oxides of magnesium (MgO) and aluminum (Al<sub>2</sub>O<sub>3</sub>) are used for crucible liners in the steel industry and consequently are excellent for the containment of low-temperature molten steel.

Magnesium oxide (MgO) has the advantages of a very high specific heat  $(0.31 \text{ cal/g}^\circ\text{C})$ , a high melting point  $(2800^\circ\text{C})$  and excellent stability with respect to molten UO<sub>2</sub> and low-temperature molten steel. It is easily fabricated and is relatively inexpensive. In large quantities, relatively pure Harklase MgO bricks are available at approximately \$1.80 for a 6-kg standard side brick with dimensions of 9-in. by 4-1/2-in. by 2-1/2 in.

A principal disadvantage of magnesium oxide, which is also a disadvantage for aluminum oxide  $(Al_2O_3)$  and graphite, is its low density compared to  $UO_2$  and its tendency to float in the molten core debris unless restrained in some manner. Another disadvantage of MgO is the potential for slag line attack. This will be discussed later.

Aluminum oxide  $(Al_2O_3)$  has a high specific heat, is chemically stable with respect to molten core debris and is available at relatively low cost. However, it has a low density and melting point (2037°C).

Zirconium oxide  $(ZrO_2)$  has the advantages of a high melting point  $(2730^{\circ}C)$  and density. It is chemically stable with respect to molten core debris. Its disadvantages include a low specific heat, structural instabilities and high cost.

Graphite has the advantages of high sublimation temperature, good specific heat, easy fabrication and low cost. It has two serious disadvantages, however. It acts as a neutron moderator, and thus increases the potential for

recriticality. Also, the reactions of graphite with  $UO_2$  generating CO,  $CO_2$  and UC are not completely understood and may be potentially serious problems.

Depleted  $0_2$  has the advantage of chemical compatibility and complete miscibility with the fuel (also  $U0_2$ ). Its high density will prevent it from floating away, it has a high melting point, and it is a good insulator. On the other hand, depleted  $U0_2$  is expensive and difficult to fabricate into structures because of its tendency to crumble into powder. If a jet of molten  $U0_2$  forms, it could easily sweep away  $U0_2$  in powder form, thereby exposing the underlying material.

#### 3. MAGNESIUM OXIDE

When the problems associated with the use of the potential sacrificial materials are considered, it appears that MgO is the best choice. Some lesser-known material might serve as a better sacrificial layer; it may be desirable to fund an extensive research and development program to examine alternatives. However, it does not appear at this time that any other material would offer significant advantages.

The advantages of MgO include:

<u>High melting point (2750°C)</u>. This reduces the rate of advance of the melting front of the core debris into the sacrificial bed.

High specific heat and heat of fusion. These properties reduce the rate of advance of the melting front by absorbing heat from the molten core debris.

Low thermal conductivity. This will also reduce the rate of advance of the melting front by slowing the rate of transmission of energy into the sacrificial bed material.

<u>Miscibility of molten fuel and sacrificial layer</u>. Since the fuel and sacrificial material are miscible, the volumetric heating rate of molten fuel will be decreased by dilution.

<u>Chemical compatibility with molten core debris</u>. The sacrificial material should not react chemically with molten  $UO_2$  or steel. It is especially important that no exothermic reactions occur between the sacrificial material and the molten core debris.

Limited gas evolution. Gaseous reaction products, such as CO,  $CO_2$  and  $H_2$ , should not be generated by chemical reactions between the molten core debris and sacrificial materials. The generation of these gases can create a potential for an explosion and can increase the sparging of fission products from core melt debris. The only gases that have been experimentally observed to evolve in  $UO_2$ -MgO interations are gases trapped in the pores (19 percent porosity).

Low cost and availability. MgO is relatively inexpensive and can be manufactured into mechanically stable structures, such as bricks, or larger cast structures at reasonable cost.

#### 4. CONTROLLABLE PROBLEMS WITH MgO

Resistance to thermal shock. A desirable characteristics of the sacrificial material is that it be sufficiently resistant to thermal shock so that it does not fail mechanically when exposed to the rapid heating transient of a core meltdown accident. Very pure magnesium oxide is not generally regarded as a good shock resistant material. Some commercial MgO firebricks contain intentionally added impurities to improve resistance to thermal shock. In the steel industry, where firebrick liners are used hundreds of times, the firebricks are successfully used in certain applications without significant spallation from thermal shock. (This will be discussed subsequently under steel industry experiences.)

However, in OPS application, the core ladle would be used only once if it is ever required. In the steel industry, spallation of an inch of material would be unacceptable because the liner must be reused. In the core ladle, loss of an inch or two of MgO from an eight-foot layer should not be unacceptable. In any event, in our small-scale tests with molten UO<sub>2</sub> and low temperature molten steel, MgO behaved acceptably without spallation when molten material was poured on cold firebrick.

It should be noted that OPS is proposing to use a chemically bonded form of MgO firebrick, rather than fired MgO, in order to enhance the shockresistance capability of the core ladle. These bricks, manufactured by Harbison & Walker Refractories under the tradename TOPEX-S, will be employed as the

top layer of bricks in the core ladle. Certain adverse consequences of using these bricks will be discussed later.

<u>Mechanical Shock</u>. Although twenty years ago, an effort was made to cushion MgO firebricks with a bottom layer of small scrap pieces before larger chunks of iron were added. Today large chunks are commonly dropped directly on the bricks in steel mills with little noticeable effect. Mechanical shock will probably not be a significant problem with the more advanced refractories available at present.

<u>Preaccident stability</u>. The sacrificial bed should be stable in the environment of reactor cavity for the expected life of the reactor. MgO will fulfill this requirement as long as the steel liner keeps it dry. However, if MgO is allowed to remain in contact with water for long periods of time (probably years), it will hydrate and change its dimensions, with serious consequences for the core ladle. Moisture level in the ladle should be monitored, and there should be provisions for dealing with moisture.

Low density. A sacrificial material with a lower density than the fuel may float to the surface of the melt if core material gets underneath it by way of cracks, seams or flaws. As will be discussed later, this problem can be controlled by proper design.

#### 5. SLAG-LINE ATTACK-A DISADVANTAGE OF MGO

Slag line attack is the one potential disadvantage in the use of MgO over which there is little control. Because of a lack of experimental data at this time, it is not possible to assess the seriousness of the problem.

Iron oxide slag may form an eutectic mixture with MgO at temperatures as low as 1500°C. Consequently, potential exists for erosion of MgO by slag at temperatures far below the melting point of pure MgO. Factors that may serve to mitigate low-temperature dissolution of MgO by iron oxide include saturation of the slag layer and a slow dissolution rate. In the steel industry, MgO is successfully used at temperatures about 1700°C without problems. In the basic oxygen process, MgO is employed in an environment where the temperature can reach 2500°C. However, there is an indication of a problem with attack by iron oxide near the boiling point of iron.

At lower temperatures, in an experiment at Sandia, a substantial degree of slag line attack was observed at 1700°C in a MgO liner for an induction furnace used to melt stainless steel. The chemical composition of the MgO in the liner of the induction furnace was different from that of the Harklase MgO proposed for use in the core ladle. This is important because there is evidence for a substantial difference in some of the physical properties of MgO as a function of impurity level.

At this time it is not possible to reach a conclusion about the extent of slag line attack at very high temperatures. Very little information exists about the interactions between MgO and iron oxide above 1700°C. Experiments planned at Sandia will examine this problem and should provide much needed data.

#### 6. DESIGN-RELATED ISSUES

<u>Flotation of Firebricks</u>. Since the brick is less dense than the molten fuel, there is the potential for floatation of firebricks if they become loose or crack. The proposed inverted arch design with a double keylock should prevent floatation. Furthermore, since each row of bricks forms an independent arch, loss of one row would not lead to loss of an entire layer of firebricks. Designs of the type suggested for the core ladle have been used for many years in the metal refining industries with only very rare instances of brick floatation. Although the core ladle environment would be more severe than that normally encountered in the metals industry, there is at present no reason to believe that arch design would be unsuccessful.

Passage of Molten Core Debris Between Bricks. It has been suggested that molten material might flow through the spaces between bricks and penetrate far into core ladle. Since the material is heat generating, it might not freeze immediately on contact with the colder firebricks. OPS has responded with calculations indicating that the rate of heat loss from the molten core debris to the brick will exceed the heat generation rate with the core debris. Consequently, the molten core debris should penetrate only a short distance before freezing. The double keylock design should also make channeling through the spaces between bricks more difficult by making the path long and more curved.

<u>Need for Increased Sidewall Thickness</u>. Sidewall thicker than that proposed by OPS would be desirable for two reasons. First, evidence indicates that molten core debris may penetrate sideways as far as it penetrates downward. Consequently the side of the core ladle should be as thick as the bottom. In addition, if the barge should list substantially during an accident, the containment would be ineffective if the sidewalls were thin.

Additional Instrumentation Is Needed. Since long-term exposure to water would be deleterious to the MgO firebricks, a provision should be made to include moisture sensors in the core ladle. Although the ladle will be encased in a thin shell of metal, water could be admitted through imperfections or cracks that may develop during operation. With moisture sensors imbedded in the core ladle, its condition could be monitored and remedial work conducted if necessary. The core ladle should also contain thermocouples or similar detectors to monitor the progress of a molten core in attacking the ladle. This information would be useful in determining the time available for remedial measures and the need for evacuation.

#### 7. CONCLUSION

A properly designed core ladle constructed from refractory materials such as MgO, as compared to concrete, can reduce the extent of gas evolution and reduce the rate of molten core penetration. The core ladle proposed by OPS seems to meet these objectives, with the exceptions noted previously, and should be successful in delaying penetration by a molten core into the environment.

#### B. RELATED INDUSTRIAL EXPERIENCE

The current practices of the metal refining industry are of interest because OPS has proposed to contain a molten core in a postulated core meltdown in a ladle that will be similar in design to ladles used to contain molten steel. Furthermore, since molten steel from the reactor structure will be a major constituent of the expected molten core debris, the experience of the metals refining industry in handling and containing molten steel is highly relevant. Industrial experience in the metals industry provides the only detailed body of engineering data available on the expected behavior of large scale refractory structures.

Before considering the applicability of the metals refining industry's experience to the FNP core ladle, it is useful to briefly review the chemistry of the steel refining process. The chemistry explains the way in which refractory bricks must be selected for compatibility with slags and why the bricks encounter higher temperatures in certain processes than are normally associated with the steel industry.

Iron ore is reduced to a high-carbon alloy of iron, called pig iron, in a blast furnace. Coke is burned in the blast furnace forming carbon monoxide which reduces the ore.

$$Fe_2O_3 + 3CO = 2Fe + 3CO_2$$
 (1)

Some of the freshly reduced iron, in turn, reacts in the blast furnace to form pig iron, either by physical contact with hot carbon or by the reaction

$$3Fe + 2CO = Fe_3C + CO_2$$
 (2)

(3)

Impurities present in the ore and coke ash must be removed. Limestone is added to the materials in the blast furnace to "flux" or convert the impurities into slag. Slags are basic if they contain a preponderance of lime (CaO) or magnesia (MgO) or acidic if they contain a preponderance of silica (SiO<sub>2</sub>) or alumina (Al<sub>2</sub>O<sub>3</sub>).

At the next stage of the steel refining process, the metal enters the basic oxygen furnace (BOF) unit where it is convert i into steel. Oxygen from a lance or probe is injected at supersonic velocity into a mixture of iron scrap and molten iron from the blast furnace. No heat is added to the metal at this point; instead, chemical reactions suprly all the energy required to melt the scrap and burn off impurities. Oxygen entering the BOF unit burns the first pig iron it encounters, forming FeO at temperatures as high as 4500°F. The FeO is quickly distributed throughout the molten pool, Initially, the silicon and manganese impurities are removed by the reactions:

$$Si + 2FeO = 2Fe + SiO_2$$

$$Mn + FeO = Fe + MnO$$

These oxides are insoluble in molten iron and accumulate, forming a layer of slag. From 5 to 30 percent of the iron in the BOF unit is converted to FeO and collects in the slag layer. The reactions generate much additional heat and raise the temperature of the remaining metal. Gradually the carbon also begins to burn off

$$FeO + Fe_3C = 4Fe + CO$$
(5)

In a few minutes, the carbon is gone. Inside the BOF unit, the molten material consists of a layer of iron, practically free of carbou, silicon and manganese, covered by a layer of slag.

Once all the impurities have been removed, charcoal and manganese can be added to produce steel with the properties desired. For example, high carbon steels can be made by adding charcoal to molten steel in the ladle.

1. MECHANICAL SHOCK

In a core melt down, large churks of metal from the reactor structure may fall considerable distances before impacting the core ladle. There has been concern about the effect of mechanical shock from this impact on the integrity of the ladle. The only available data in this area are from the metals refining industry; consequently, it is useful to review their experience.

During each heat in a BOF unit, from 20 to 30 percent of the initial charge, or from 40 to 60 tons, consists of scrap metal. The remainder is molten steel from the blast furnaces. Pieces of scrap metal as large as ten tons are dropped from a height of 20 ft onto the refractory in the BOF unit

As previously noted, there was concern about the effects of mechanical shock on the MgO refractory at one time. Twenty years ago, a bed of smaller sized scrap metal was placed on top of the refractory to protect it from the impact caused by larger pieces. With the improved refractories available today, there is no concern about mechanical shock caused by the impact of scrap.

(4)

Areas of the furnace impacted by scrap do weal slightly faster but the hot strength of the material is quite good.

It also should be noted that supersonic velocity oxygen from the oxygen probe in the BOF unit blows the steel scrap around in the furnace. Even so, wear on the furnace walls from mechanical shock is minimal.

This information indicates that an MgO core ladle will be unaffected by the mechanical shock that would result if large pieces of the internals of the reactor vessel fell on the MgO.

2. PREHEATING OF MgO, CRACKING AND THERMAL SHOCK

Another area of concern regards the thermal shock and cracking that could occur when molten  $UO_2$  or steel comes into control with the core ladle. Again, a review of industrial experience is relevant in considering the resistance of the core ladle to cracking and thermal shock.

Usually, MgO liners in steel mills, with certain exceptions noted below, are preheated from 3 to 4 hrs with gas jets prior to use to reduce potential thermal shock problems. After preheating, the liner is at a temperature of 2000°F. No attempt is made to heat the liner to the approximately 3000°F (emperature of the metal.

In certain applications, high purity MgO is used successfully without preheating. An example is the "slide gate" used to control the flow of molten steel from a ladle. This device consists of two blocks of MgO containing apertures. When the MgO blocks are moved so that the apertures coincide, molten steel can pass through and leave the ladle. The design and required condit ons of operation make preheating of the slide gates an impossibility. The temperature of the slide gate prior to contact with molten metal is about 100°F.

No attempt is made to preheat the MgO liner in the BOF unit prior to use, even when it is cold. During normal operation, the furnace cools to room temperature from two to five times during the life of a liner. At times the BOF units have been operated only one or two shifts per day because of a lack of demand for steel. Industry studies suggest that liner life will be reduced by

25 percent if the liner is allowed to cool to room temperature every day and is not preheated prior to use.

Molten steel has accidentally been poured into cold MgO ladles when steel industry personnel have forgotten to preheat the ladles. In these situations, damage to the ladle has been minimal rather than catastrophic. The only damage that has been observed is cracking to a depth of 2 to 3 inches. Below that, the liner remains undamaged. Since the cracked material remains in place, it continues to contain the molten metal.

Emergency ladles, used to hold molten steel when "breakouts" or leaks occur, arc lined with inexpensive firelay and are not preheated. Even so, the ladles will hold molten metal without difficulty.

The cracking of cold MgO ladles from molten steel pours on cold liners should not be a problem in the case of the OPS core ladle. Unlike the steel industry ladles which must be used repeatedly, the FNP core ladle will be used only once if it is ever needed. If the molten core debris affects the core ladle, along the lines of steel industry experience, the loss of a few inches of MgO from an eight-foot-thick layer as a resu<sup>+</sup>: of cracking should not pose a problem. Furthermore, in industrial experience, the cracked bricks remain in place and thus continue to provide protection.

Although the  $UO_2$ -steel mixture in a core meltdown will be at a higher temperature than is normally encountered in the steel industry, it should be noted that the  $UO_2$ , at least, is a poorer conductor. There is evidence from experiments performed several years ago at The Aerospace Corporation that cracking induced by molten  $UO_2$  will not be worse than the steel industry experience. In those experiments, there was little evidence of significant damage from cracking induced by pours of molten  $UO_2$ . Whether  $5200^{\circ}F$  molten steel would cause significantly greater thermal shock than molten  $UO_2$  is not known. Experiments with molten steel- $UO_2$  mixtures were performed at The Aerospace Corporation, but the yields of molten material were too small to provide a good test of thermal shock resistance.

From many years of experience, the steel industry has learned which compositions, grain sizes and densities provide the optimum for thermal cycling.

With a controlled microstructure, better thermal resistance can be achieved. The industry experience has been that 98-percent pure MgO is more shock resistant than either 93-percent MgO or 100-percent pure MgO. A mixture of small and large grains is typically used, and the best shock resistance is found to be obtained with a mixture of 60-percent course grains (-0.5-in. down to -20 mesh) and 40-percent fine grains (-60 mesh down to micron size). The porosity is maintained from 15 to 20 percent, as fully dense MgO cannot withstand thermal shock. The pores are believed to serve as crack terminators. Chromepericlase bricks have been found to have a better thermal resistance than MgO and are sometimes used in applications where thermal shock is felt to be important.

All of this indicates that there is little reason for serious concern about the resistance of the MgO core ladle to thermal shock. The ladle will only be used once, and even if cracking occurs, the bricks will remain in place.

#### 3. BRICK FLOATATION

Since Mgo bricks have a lower density than the molten core debris, it is possible that MgO bricks could float away in the more dense molten steel and  $UO_2$ . Consequently, there was interest in learning whether the steel industry has had any adverse experience with brick floatation. It appears that instances of brick floatation have been extremely rare. Usually an inverted arch design employed in refractory brick structures has proved successful in the restraint of the bricks, indicating that the similar design selected for the core ladle should be adequate to prevent brick floatation. Brick structures with flat bottoms and diameters in the 15 to 18 foot range have been constructed without the benefit of an inverted arch design. Floatation of bricks has generally not occurred with these designs either. The inverted arch design and double keylocking of bricks proposed for the FNP should be sufficient to prevent brick floatation.

#### 4. FUSING OF JOINTS

Representatives of Harbison and Walker Refractories and OPS have suggested that the joints between bricks in the core la le will fuse together

during contact with molten core debris. In the steel industry, the joints remain after contact with molten metal at 3000°F. When the metal cools, the bricks are still discrete. Since the molten core debris would be at a much higher temperature, fusion of bricks may occur. The experience of the steel industry here is not applicable to the reactor accident scenario.

#### 5. SLAG LINE ATTACK

Concern exists that slag from molten steel from the reactor structure could attack and seriously erode the effectiveness of the MgO core ladle. Although the temperatures in a reactor accident are much higher than those found in the steel industry, enhancing a slag attack, it is still useful to review steel industry experience, as there is little other available data for evaluation of the problem.

The lining used in a BOF unit is determined by the chemistry of the expected slag. In the units at U.S. Steel, for example, the slags are basic, with lime to silica ratios greater than 1.5. Lime and MgO are added to the molten mixture to control basicity and slag attack; the added MgO saturates the slag layer. MgO bricks are selected with lime to silica ratios similar to the slag for compatibility. A compatible brick is more resistant to slag line attack and more unlikely to be wet by the slag. Consequently, in the FNP, the refractory must be chosen to be compatible with the expected slag.

Eightly percent of the liner of a BOF unit consists of "tempered" brick, in which the MgO grains have been impregnated with pitch and baked at a low temperature (500°F). The tempered brick contains 96 to 97-percent MgO and some intentionally added carbon to reduce penetration by slag. In addition to a 2-percent carbon contribution from the pitch, very fine carbon totaling 2 percent is added to the MgO. The reasons for the effectiveness of carbon are not understood. According to one theory, the slag cannot wet or penetrate the MgO pores when carbon is present. It has also been suggested that the permeable MgO can volatilize, combine with carbon and redeposit forming an impermeable layer.

The remaining 20 percent of the liner consists of a more expensive "burnimpregnated" brick prepared by passing the MgO through a firing process prior

to the addition of pitch. This material has a greater hot strength than the tempered brick and is used in regions where it will be subject to impact from falling iron scrap. The composition of the burn-impregnated brick is similar to the tempered brick. Again, carbon-bearing material is added to the brick to reduce slag attack. The presence of carbon reduces the slag penetration from approximately 5.0 cm to a few mm per heat. Carbon cannot be employed in an oxidizing environment. The BOF environment is reducing as a result of the CO and CO<sub>2</sub> formed as carbon is oxidized.

In the BOF units at U.S. Steel, the slags contain a high concentration of iron oxides. The slag thickness tends to be from 12 to 18 in. in a 40 to 48-in. layer of molten material. The slag is quickly saturated with MgO and penetrates the MgO liner to a depth of only a few mm. The usual operating temperature is about 3000°F. No heat is added to the molten metal in the BOF unit; rather, energy is supplied by chemical reactions with the supersonically introduced oxygen. Oxidized particles of iron in contact with the MgO walls are thought to be at temperatures as high as 4500°F because of the oxidation reaction. Thus, there is some relevant industrial experience at temperatures approaching those of a nuclear accident scenario. In this environment, the liners last for 1200 to 2000 heats until they eventually must be replaced because of slag attack. A typical heat lasts about one hour; thus, a total of 24 in. of MgO are eroded over 1200 to 2000 hr.

Slag is never completely eliminated, even when refined metal is heated. The slag from melting refined metal in air will amount to less than one percent of the total. In an environment containing water, or where air bubbles through molten iron, additional slag can be generated from refined steel. Thus, in a reactor, some slag can be expected when the metal structures melt.

As temperature increases, the amount of slag attack rises substantially. At 3200°F, the highest tap temperature used in steel mills, the erosion rate is twice what it is at 3000°F. The industry tries to avoid temperatures above 3000°F because leaks in ladles and crucibles increase dramatically at higher temperatures. Also, continuous casting equipment is designed to handle lower temperature metal. With excessive superheating the skin on the castings will

treak. In addition, alloys from superheated steel will not have the desired properties. The industry has no experience with near-boiling iron in the temperature range that may occur in a nuclear reactor core meltdown.

Harklase MgO is somewhat less slag resistant than the high lime to silica ratio MgO bricks used in the working liner of the ladle. However, the difference is small, about ten percent.

Chrome-periclase bricks are occasionally used in steel mills in place of MgO. These bricks are more resistant to thermal shock than MgO. They are also better insulators and have a lower thermal expansion constant, and thus are better suited for certain applications, such as the roofs of electric furnaces. However, these bricks are considerably less resistant to attack by basic slags than is MgO. In a neutral or accidic slag environment, chrome-periclase bricks are more resistant to slag attack than MgO. For the core ladle, the refractory selected must be tailored to the expected chemical environment.

#### 6. HYDRATION RESISTANCE

When MgO is hydrated, it swells and becomes less dense. Substantial differences have been observed in the hydration resistance of various types of commercially available MgO bricks. These differences appear to be caused principally by the quantity of lime present in the bricks. In water, a high lime MgO brick will swell within 48 hours. Harklase, which has a relatively low lime concentration, is considerably more resistant to swelling even after months of exposure.

Hydration resistance seems to be the principal advantage of the relatively expensive Harklase brick and was probably the motivating factor in its selection as the core ladle material. Hydration resistance is important in the FNP, as water may accumulate in the core ladle over long periods of time. Although the ladle will have a steel jacket, imperfections in its construction could allow water to control the MgO. In the steel industry, Harklase bricks are generally used only in the safety liners of ladles because of the relatively high cost of the brick. A ladle typically has 12 in. of Harklase as a safety liner next to the metal wall with a 24 to 27-in.-thick working liner of

a cheaper grade of MgO in contact with the molten steel and slag. Harklase is used for the safety liner so that the ladle can be sprayed with water to cool it quickly when it becomes necessary to reline the working liner.

The MgO bricks that have been impregnated with pitch tend also to be more resistant to hydration. Some of these bricks have been stored at U.S. Steel for as long as 5 years without becoming hydrated. Impregnation of bricks with boric acid has also been used to improve hydration resistance. Acids generally increase the hydration resistance of MgO bricks for reasons that are not understood.

Problems have been reported with the hydration of MgO in steam. An American Society for Testing Materials (ASTM) test procedure<sup>7</sup> to compare the hydration resistance of MgO bricks has been developed.

It appears that hydration resistance can be a problem with certain kinds of MgO bricks since water may accumulate in the core ladle despite its steel jacket. For this reason the core ladle should be equipped with moisture sensors and a method of water drainage.

7. THE EFFECT OF MOLTEN STEEL ON WET MgO BRICKS

The effect of molten steel on wet MgO bricks is of interest because of the possibility that water from the reactor will accumulate in the core ladle. Although a steel shell will cover the ladle, imperfections in the shell could develop over time and admit water to saturate the bricks.

Molten steel has been poured on wet MgO bricks at U.S. Steel without the serious consequences that might be expected. Steam forms in the brick and is driven back through the refractory. It leaves the ladle through vent holes or drains provided for this purpose at the bottom of the ladle.

Serious consequences can result when molten steel is poured on top of pools of water in crucibles, trapping the water. Steam can be released with explosive force and can cause sprays of molten metal as it escapes from the molten pool.

With proper design, molten UO<sub>2</sub> and steel from a core melt accident will have little effect on wet refractory bricks. This problem is controllable unless pools of water are allowed to accumulate.

#### 8. GAS EVOLUTION

Substantial quantities of gas are generated when molten steel is poured on pitch-impregnated MgO bricks. This may be of concern if OPS proceeds with plans to employ a top layer of shock resistant TOPEX-S brick (see Section III-1). This material is impregnated with the waste liquor from paper mills and appears to generate large quantities of a hydrocarbon gas, which is at least potentially flammable and will increase the pressure in the reactor containment building.

#### 9. CONCLUSIONS

It should be noted again that the temperatures employed in the steel industry are around 3000°F, with an indication of the presence of 4600°F iron oxide particles in contact with the MgO liner in the BOF unit. At the postulated 5200°F temperatures in a core meltdown event, the rate of 3lag line attack will initially be much greater than at 3000°F until the slag is saturated with MgO. At temperatures between 3000°F and 5200°F, the amount of MgO needed to saturate the slag will increase as indicated by the iron oxide-MgO phase diagram. It is this slag attack at higher temperatures that is the main concern regarding the use of MgO in the core ladle.

Several more positive points should be made about the potential for the of MgO in the core ladle. Steel industry experience suggests that there should be little concern about the response of modern MgO refractory bricks to mechanical shock. The MgO liners appear to be reasonably resistant to thermal shock, even when cold. Brick floatation does not seem to be a problem with proper design. It appears that MgO bricks, like Harklase, with a low lime concentration, are reasonably hydration-resistant in water, although we would like to see more long-term data. Also, with proper design, molten metal interactions with wet MgO bricks should not cause problems.

In summary, although there is a slag attack problem with MgO, it still seems to be the best material for the core ladle application. If the amount of available MgO is sufficiently large compared to the slag layer, the slag layer will eventually be saturated with it. None of the other materials suggested for this application appear at this time to be more promising than MgO.

#### C. EXTENT OF WATER ABSORPTION BY MgO BRICK

The absorption of water by MgO bricks was determined in an experiment in our laboratories. Initially, a specimen of Harklase MgO brick was sectioned into five pieces using a diamond saw and water coolant. The brick pieces were then measured by taking two readings each of the length and width and four readings of the thickness. The volumes were calculated and are provided in Table II.

During the sectioning, the bricks absorbed water from the coolant for the saw. Approximately 18 hr were required before the bricks dried. At that time, the damp brick pieces were placed in an oven which had been preheated to 300 C and kept at approximately this temperature for 6-1/2 hr. For the first half hour, steam from the bricks cooled the oven somewhat. The remaining six hrs were uneventful.

At the end of the 6-1/2-hr drying period, the bricks were removed from the oven and weighed. The dry weight and calculated apparent density are given in Table II.

The bricks were subsequently submerged in water for a period of 16 hr, then dried of excess water and weighed again. The specimens were weighed within 30 seconds after removal from the water since they appeared to be drying rapidly. The weight of the wet bricks, the calculated weight of water absorbed and volume percent of water absorbed are provided in the table.

After weighing, the wet bricks were left on a laboratory bench for two days and then were reweighed. Weights were identical with measurements made when the specimens were first removed from the oven, indicating that the Harklase brick dries out completely in air at room temperature (20 C, and from 50 to 70 percent relative humidity).

As noted earlier in this report, steel industry experience indicates that Harklase is resistant to swelling by water-induced hydration because of its low lime concentration. However, MgO will hydrate readily in steam. In recent experiments at Sandia Laboratories, substantial hydration has been observed in Harklase after only a few weeks exposure to water (not steam).

Brick	Volume, cc	Weight Dry, gm	Weight Wet, gm	Density, gm/cc	Weight H <sub>2</sub> 0 gm	Weight % H <sub>2</sub> O	Volume % H <sub>2</sub> 0
1	49.091	141.0	148.3	2.893	6.3	4.4	12.7
2	58.904	169.95	177.4	2.884	7.5	4.4	12.7
3	58.226	166.8	173.8	2.865	7.0	4.2	12.0
4	57.478	154.95	172.0	2.871	7.0	4.2	12.1
5	159.263	461.1	480.8	2.895	19.7	4.3	12.4
Average				2.882		4.3	12.4

Table II.	Water	Absorption	by	MgO	Brick	Submerged	in	Water	
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Experiments have recently been conducted at our facilities in which Harklase MgO was left in a humid atmosphere and submerged in both tap water (pH = 6.0) and distilled water (pH = 6.0) for 60 days. At the end of that period, visual observation indicated no swelling or apparent loss of strength.

#### D. TOPEX-S MgO BRICK

Manufactured by Harlison and Walker Refractories, this brick has been proposed by OPS for use as the top layer of bricks in the core ladle. It is essentially an unfired MgO brick with a chemical bonding agent added to hold it together. Although the composition of the bonding agent is proprietary, a principal constituent is the waste liquor from paper pulp mills.

A TOPEX-S brick was sectioned into specimens for density, porosity and water absorption measurements. Since the brick was cut using a water coolant, the specimens were initially weighed after soaking at least three hrs in water.

The brick specimens were placed in a horizontal quartz tube furnace and heated. A flow of 1.5 SCFH of a 2-percent  $O_2$  and 98-percent  $N_2$  mixture passed through the tube during the period of heating. At temperature of 140°C, steam came from the tube and some water condensed in the cool part of the tube. Ultimately approximately 10 cc of water previously collected in the tube also evaporated. During heating, the water acquired a straw color which became progressively darker. As heating continued, the water evaporated and was replaced by a dark oily residue which collected in the cool downstream portion of the tube. In addition, a large quantity of a noxious blue-grey smoke was emitted from the tube for over a half-hour. Eventually, the temperature reached 825°C and the oily residue deposits were oxidized and removed from the tube. During this part of the heating cycle, the exit gas emerged from the tube in pufis, suggesting some low-level deflagrations.

After cooling, the specimens were weighed, then soaked in water for 18 hr and weighed again. The two wet weights were approximately the same. When the samples were removed from water after soaking for 18 hr, they emitted a strong odor of H<sub>2</sub>S. The heating procedure changed the color of the brick samples from a dark tan color with light tan inclusions to a light grey color (the

Sample Number	First Measurement	Second Measurement	Dry Weight gms	Volume, cc	Weight H <sub>2</sub> 0, gms	Weight %, H <sub>2</sub> 0	Volume %, H <sub>2</sub> O	Density, gms/cc
1	69.9	70.0	66.3	23.17	3.55	5.35	15.3	2.83
2	62.1	62.2	58.9	21.46	3.20	5.44	14.9	2.79
3	114.1	114.2	108.1	38.68	6.05	5.60	15.6	2.74
4	128.7	128.7	122.2	43.13	6.50	5.32	15.1	2.86
Average						5.43	15.2	2.81

## Table III. Water Absorption by TOPEX-S Brick

Wet Weight,

gms

color of cigarette ash). Some of the surfaces also had black stains from what appeared to be unoxidized carbon.

Table III provides the measured and calculated values of density and water absorption.

The porosity and density of TOPEX-S chemically bonded MgO brick were measured by mercury porosimetry. Results are in Table IV for a series of measurements. The greater density and lower porosity of this brick as compared with the Harklase-fired MgO brick is probably due to the addition of the hydrocarbons employed to provide chemical bonding. When the temperature in the brick exceeds the relatively low boiling point of these hydrocarbons, they boil away, although a carbonaceous residue remains.

Sample Number	Bulk Density, g/cc	Apparent Density, g/cc	Percent Void Volume	Volume of Voids/gm, cc/gm
1	3.01	3.22	6.5	0.022
2	2.53	2.70	6.5	0.026
3	2.98	3.18	6.4	0.022
Average	2.84	3.03	6.5	0.023

Table IV. Porosity of TOPEX-S Brick

#### E. COMPRESSIVE STRENGTH OF FNP CONCRETE AT HIGH TEMPERATURES

Some concern has been expressed by the ACRS over the effect of high temperature on the nonstructural concrete sidewalls. The area is question is located behind the MgO bricklayers above the FNP core ladle. The concern centers on the possibility of the wall collapsing if it is heated sufficiently.

The physical properties of concrete are a function of its temperature. The coefficients of expansion of the various types of concrete depend upon the material used as the aggregate as well as on the cement. The relative coefficients of expansion of the constituents influence the extent of cracking as the material is heated. The compressive strength depends strongly on the cement and, to a much lesser extent, on the aggregate used. The most important effect on compressive strength is the dehydration of the cement paste.

In the range between room temperature and 300°C, both limestone aggregate and basalt aggregate concretes retain their initial compressive strengths,<sup>8</sup> to within ±5 percent (see Figure 1). At 550°C, both types of concrete retain approximately 70 percent of their initial compressive strengths. Limestone retains approximately 40 percent of its compressive strength while basalt retains only 30 percent of its initial strength at a temperature of 700°C. At 800°C, limestone aggregate concrete is down to 25 percent and basalt aggregate is down to 15 percent of its initial strength.<sup>8</sup>

The survival of a concrete surface at elevated temperatures depends not only on the temperature of the concrete but on the rate of heating. Steam generated within the concrete must escape and tends to weaken the concrete, but spalling is generally not observed. Some cracking is to be expected under these circumstances.

Figures 2 and 3 indicate the strength of basalt concrete at different temperatures after exposure for periods of as long as four hours.<sup>9</sup> In the worse situation for which we have data, ordinary concrete was exposed to a temperature of 927°C (1700°F) for four hours. The ultimate load strength was 113 psi in the weakest specimen and the average was 130 psi.

Although this is not the same material as the basalt concrete proposed for the FNP, it is useful to look at its behavior further in the absence of other data. Since the pressure exerted by the weight of a one foot thick slab of concrete with a density of 2.6 g/cc is 1.13 psi, the maximum height concrete wall that would be self-supporting is 100 ft high. The wall in the reactor room that is of concern has a height of 35 ft, leaving a safety factor of 3. Consequently, at 927°C, after four hours, the FNP wall would continue to be self-supporting.

In conclusion, a vertical, nonstructural wall of concrete should hold together without difficulty at temperatures of about 900°C. As long as the concrete retains a significant percentage of its original strength, it will not disintegrate.

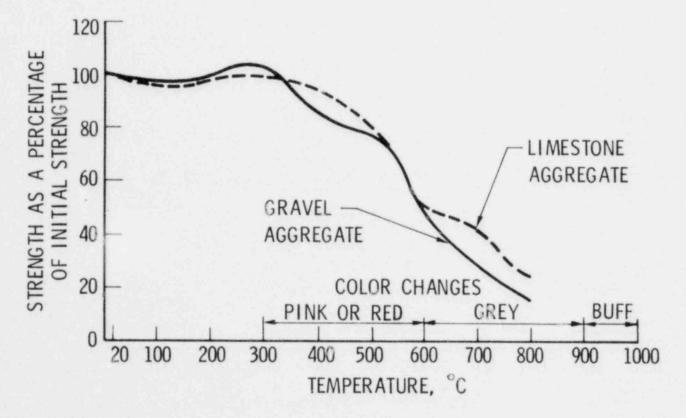
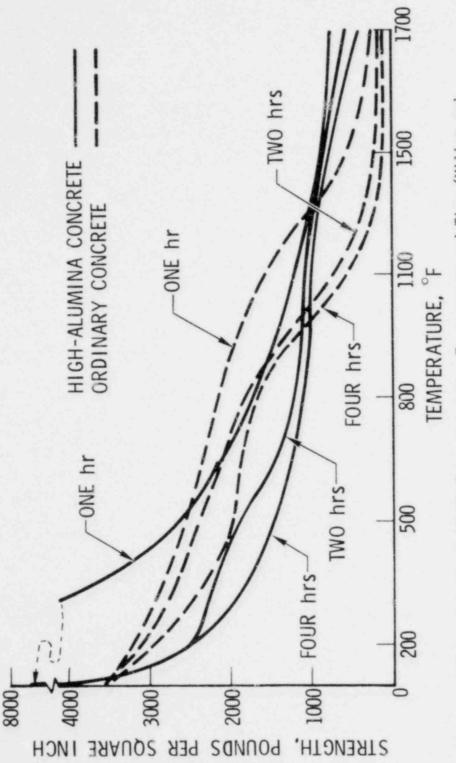


Figure 1. Compressive Strength of Concrete After Heating to High Temperatures (A. M. Neville, "Properties of Concrete," John Wiley and Sons, Inc., New York, 1963)



Strength of Concrete versus Temperature and Time (Miller and Faulkner, The Effect of High Temperature on Concrete) Figure 2.

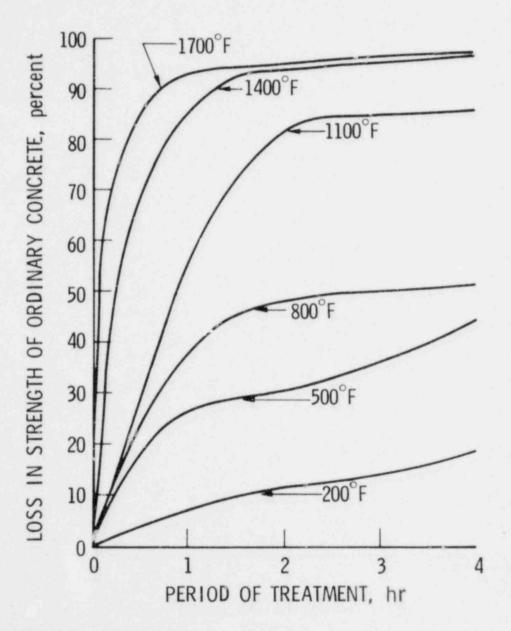


Figure 3. Loss in Strength (Percent) of Ordinary Concrete versus Time and Temperature (Ref. 9)

### IV. RECENT UO2 - MATERIALS INTERACTION EXPERIMENTS

#### A. SANDIA LABORATORIES MgO SURVEY EXPERIMENTS

Recently, experiments have been conducted at Sandia with MgO by itself and with MgO, and molten  $UO_2$ . When MgO was heated by itself, gas evolution was observed at 2750°C.

In the molten UO2 - MgO test, a 0.5-in.-thick disk of MgO with a diameter of 2 in. was heated in a furnace in contact with 30 g of  $UO_2$  in an argon atmosphere. At 1900°C, the UO2 penetrated entirely through the 0.5-in.-thick disk during an unknown period of time. The penetration at a temperature below the melting point of UO2 suggests the formation of a low melting mixture. During the course of the UO2-MgO experiment, gas evolution from the brick was observed at 2600°C. At 2500°C, there was no evolution of gas. The gas producing reaction was described as turning the molten mixture into a foam. Concern has been expressed that a gas generating reaction could affect the containment capability of a core ladle by stirring the molten mixture and thereby altering its heat transfer properties. Consequently, gas evolution could enhance the rate of penetration. It has been suggested that MgO is unsuitable as a core retention material on the basis of this observed gas evolution which has been ascribed to a reaction between  $UO_2$  and MgO generating  $UO_3$ . We believe that there are other reactions that can account for the observed gas in the experimental environment of Sandia's tests.

Apparently, in both tests, the experimental heating apparatus contained a resistively heated tungsten grid. In each test, the specimens were supported by this grid. In both tests significant amounts of tungsten and magnesium dioxide disappeared from the reaction zone. It has been reported in various papers 10,11,12 that when MgO is heated in contact with tungsten, a volatile oxide of tungsten, WO<sub>3</sub>, is formed. Many of these papers 11 describe reactions which occur around 2000°K (1727°C) at which point the equilibrium pressure of WO<sub>3</sub> is only  $10^{-5}$  atm. At this low temperature, the reaction proceeds too slowly to produce a visible reaction. Foaming is not observed at these temperatures because the MgO is still solid. However, in Brewer's paper<sup>11</sup> it was reported that MgO reacted rapidly with tungsten at 2750°K.

A temperature of 2750°K (2477°C) is below the temperatures at which the Sandia reactions were observed. It is significant to note in the Sandia experiments that both the magnesia and the magnesia-urania mixtures melted before foaming was observed. The molten magnesia makes better contact with the tungsten than does a solid and can act as a flux.

It appears that the foaming reaction of magnesia on a heated tungsten grid can adequately be explained by the tungsten-magnesia reaction described in the literature. The foaming at a lower temperature in the  $UO_2$ -magnesia experiment probably results from the lowering of the liquidus temperature by  $UO_2$  since formation of a liquid is a prerequiste to foaming (solids will not foam). Accordingly, it is our view that these experiments do not provide evidence for asserting that MgO is unsuitable as a core retention material.

#### B. ARCONNE NATIONAL LABORATORY EXPERIMENTS

Experiments at Argonne National Laboratory (ANL) with molten  $UO_2$  have recently been resumed including an experiment with limestone concrete mixed according to the Clinch River Breeder Reactor formulation<sup>13</sup>. These experiments provide an opportunity for comparison and verification of early Aerospace Corporation tests.<sup>1,2</sup>

The test cell was a 10 kv 10 by 61-cm-rectangular parallelepiped with sides and bottom constructed c water-cooled, U-shaped brass segments. Graphite electrodes were employed at each end of the cell. The apparatus was filled to a depth of 30 cm with limestone concrete; this in turn was covered with 9.5 cm of powdered UO<sub>2</sub>. An embedded tantalum wire was used to heat the UO<sub>2</sub> powder for 45 min. The UO<sub>2</sub> was then resistively heated by applying 15 kW to the powder. Molten UO<sub>2</sub> remained in contact with the concrete for about 45 min and produced a maximum penetration of about 7 cm.

After the test the concrete was examined, and two different zones were identified. In one region the concrete was both degassed and dehydrated and the zone consisted principally of CaO. The CaO remained after  $CO_2$  evolved from the CaCO<sub>3</sub> in the aggregate. The second, lower, zone consisted of dehydrated concrete. There was an indic tion of a vigorous or even violent gas evolution with molten material spewed in all directions.

The ANL results are reviewed here because they provide one of the very few opportunities for comparison and confirmation of results obtained in earlier work at The Aerospace Corporation. In tests conducted at Aerospace in 1977, a 1.4-cm penetration by molten  $UO_2$  into Clinch River limestone concrete was observed. The  $UO_2$  was heated by an electric arc. Although the experiment lasted a total of 45 mir, the  $UO_2$  was probably molten in the region of the concrete only for 15 to 25 minutes. Given the differences in the method of heating and other differences in the experiments, the agreement between the ANL and Aerospace experiments is quite reasonable.

#### V. RETROFITTING EXISTING REACTORS WITH CORE RETENTION SYSTEMS

It has been suggested that the concrete-lined cavity below existing reactors be modified to improve the resistance of the containment structure to penetration by molten core debris. Various experimenters have estimated this penetration rate to be 1 to 4  $ft/h^{1,4}$ . If this estimate is correct, reduction of the penetration rate would be highly desirable.

For some reactor systems, feasible modifications to the area below the reactor vessel are restricted to comparatively simple procedures because of limited physical access and economic considerations. Physical access is limited by radiation levels and by the absence of entry ports or manholes of sufficient size to admit a worker. Economic considerations probably do not allow the excavation of the foundations of a reactor plant. Since the available space is probably limited, dilution of a molten core in a large volume of material is presumably impractical and, consequently, only sacrificial bed core retention systems will be considered.

The subsequent emphasis of this section will be on examination of materials that could be poured in granular or chunk form onto a reactor room floor to retrofit a reactor with a sacrificial layer. Such a layer might be economically installed, without substantial modifications to either the access ports or reactor cavity floor, if a suitable material is available at reasonable cost. The disadvantage of this kind of layer, as compared to individually placed bricks of a refractory material, is its susceptibility to the channeling of molten core debris between the granules or chunks.

Since these granules or chunks would be unrestrained, the first priority in the selection of a material is a density at least as high as that of the  $UO_2$  fuel. It is impractical to construct a floor of unrestrained low density material that could reliably withstand the mechanical and thermal shocks of a core disruptive accident without floating away in the molten  $UO_2$ .

 $UO_2$  has a maximum density of 10.96 g/cc at room temperature and 8.7 g/cc at its melting point of 2860°C. To a first approximation, most materials

expand with the same coefficient of expansion as that of UO<sub>2</sub> so that the choice of sacrificial materials is limited to those with densities greater than 11 g/cc at room temperature. This density requirement limits the passible candidate materials to 14 heavy metals and 15 of their compounds. There are no compounds of less dense metals whose density exceeds 11 g/cc. A comprehensive list of materials with densities greater than 11 g/cc is provided in Table V.

This list of candidate sacrificial materials does not include, for obvious reasons, the transuranic elements and their compounds. Other considerations include a low boiling point, which eliminates gold, lead, mercury, thallium and tungsten oxide. Materials with a high toxicity, such as osmium dioxide, are also unacceptable. The list of candidate materials cannot include those materials whose cost is prohibitive (gold, palladium, rhenium, rhodium, ruthenium and their compounds). If we further stipulate that the candidate sacrificial material have a melting point at least as high as that of uranium dioxide, the list is reduced to eight possible choices; these are listed in Table V<sup>1</sup>.

The sacrificial material selected should have a high enthalpy so that as much of the molten core's heat as possible will be used in heating and melting the refractory. The enthalpy of several materials studied thus far is provided in Table VI.

The final consideration of the candidate materials in Table VI should take into account their chemical compatiability with molten  $UO_2$  and other materials expected to be present in a core meltdown.  $UO_2$  is, of course, compatable with itself, but it has been reported to form a lower oxide ( $UO_{1.86}$ ) and oxygen at temperatures above 1600°C under vacuum or an inert atmosphere. The high temperature chemistry of the other candidate materials with molten  $UO_2$  is not well known and must be investigated before any sacrificial material can be recommended. One potential reaction is the oxidation of tungsten and its compounds to form a volatile oxide of tungsten. The metal carbides could oxidize to form CO, which would readily escape. These reactions are speculative and should be thoroughly investigated.

Compound or Element	Density, g/cc	Melting Point, °C	Boiling Point, °C
Gold	19.32	1064	2807
Hafnium	13.31	2227	4602
Hafnium Carbide	12.2	3890	ant out out into
Lead	11.34	327.5	1740
Mercury	13,59	-38.87	356.6
Mercuric Oxide	11.1	d(500)	
Osmium	22.48	3045	5027
Osmium Dioxide	11.37	d(500)	
Palladium	12.02	1552	3140
Rhenium	20.53	3180	5627
Rhenium Oxide	11.4	d(1000)	
Rhodium	12.4	1966	2727
Ruthenium	12.3	2310	3900
Tantalum	16.6	2996	5425
Tantalum Boride	11.15	3000	
Tantalum Carbide	13.9	3880	5500
Tantalum Nitride	16.3	3360	
Thallium	11.85	303.5	1457
Thorium	11.7	1750	
Tungsten	19.35	3410	5660
Tungsten Carbide	15.63	2870	6000
Tungsten Dicarbide	17.15	2860	6000
Tungsten Dioxide	12.11		800 (sub)
Uranium	18.95-19.05	1132	3818
Uranium Boride	12.7	2365	
Uranium Dicarbide	11.28	2350-2400	4370
Uranium Hydride	11.4	Unstable	
Uranium Nitride	14.31	2630	
Uranium Dioxide	10.96	2878	3880

Table V. Elements and Compounds with Densities Greater than 11.0 g/cc

Compound	Density, g/cc	Melting Point, °C	Boiling Point, °C
Hafnium Carbide	12.2	3890	?
Tantalum	16.6	2996	5425
Tantalum Boride	11.15	3000	?
Tantalum Carbide	13.9	3880	5500
Tantalum Nitride	16.3	3360	?
Tungsten	19.35	3410	5660
Tungsten Carbide	15.63	2870	6000
Tungsten Dicarbide	17.15	2860	6000
Uranium Dioxide	10.96	2860	3880

Table VI. High Density, High Melting Point Compounds

Table VII. Enthalpies of High Density, High Melting Point Materials

Material	cc melted/kcal	Melting Point, °C	Density, g/cc
TaC	1.6	3927	14.5
MgO	2.4	2853	3.5
Zr02	2.3	2760	5.7
U02	3.7	2860	8.7 (liquid)
ThO <sub>2</sub>	4.1	3300	9.95

The most promising of these high density compounds is tantalum carbide, (TaC) with a melting point of 3927°C and a density of 14.5 g/cc. Expressed in terms of cc of refractory material melted per kcal of heat input, the enthalpy value of TaC is the lowest of any of the materials studied. Its high temperature chemistry in the presence of molten core debiis is unknown at this time. A major problem with TaC is its cost, which is about \$70/kg. The material also is not currently available in large quantities, and production techniques may require expensive, time-consuming development.

High alumina cement (HAC) has been suggested as a candidate sacrificial material for retrofitting existing reactors. It is relatively inexpensive and forms a monolithic layer, preventing floatation. It is more refractory than basalt increte, and some experimental evidence indicates that it is fairly resistant to penetration by molten steel at 1900°C. However, molten core debris is expected to react temperatures on the order of the melting point of  $UO_2$  (2860°C). At higher temperatures, HAC may not provide much protection against core penetration, and it presumably will generate substantial quantities of gas, including hydrogen. Gas generation can lead to overpressuization of the containment. It also is expected to form a low melting eutectic with iron oxide slag.

Multilayer . "stems might offer cost reduction advantages. For example, consider a "sandwich" of HAC, TaC, HaC and MgO pellets. The TaC and MgO granules could be poured into the cavity and the HAC would be an inexpensive way of installing a material to hold the TaC and MgO in place. The HAC, as a monolithic layer, would resist floatation, and TaC would not float because of its density. The top three layers would tend to hold the MgO pellets in place.

Another suggestion that should be explored, where space is limited, is the possibility of channeling the molten core debris to another location where more space might be available.

#### VI. THE GAS-COOLED FAST REACTOR POST-ACCIDENT FUEL CONTAINMENT PROGRAM

This program at General Atomic is concerned with safety issues in the Gas Cooled Fast Reactor (GCFR) program. Its objectives include an assessment of the postaccident fuel containment capability of the prestressed concrete reactor vessel (PCPV) and a consideration of several design concepts for molten core retention.

In the current design, if molten fuel relocates to the bottom of the GCFR reactor vessel, it will initially contact the lower preshield. This complicated structure consists of approximately 30 cm of stainless steel covering a lower main shield. The main shield consists of large blocks of graphite encapsulated in stainless steel. There is a layer of fused silica that acts as a thermal barrier between the lower main shield and the cavity liner coo'ing system.

Without a sacrificial layer present, calculations indicate that twothirds of the heat leaving the molten pool will travel upward and one-third downward into the liner and cooling system. Furthermore, it is felt that the graphite blocks will delay the arrival of the heat front at the cooling system for ten hr. There are two problems with this scenario in our view. The feasibility is dependent on the proportion of heat transferred upward, which may differ substantially from the calculated values due to uncertainties about thermal properties. A larger proportion of heat transferred downward would increase the rate of penetration of the lower shield. In addition, there is the possibility of penetration of the lower main shield by molten core debris, followed by flotation of the low-density graphite blocks. If floatation of the blocks occurs, or if the molten core penetrates between the blocks, only the thin thermal barrier would remain to prevent the core from penetrating the cooling system and entering the concrete vessel. Some consideration should be given to ways of restraining the graphite blocks in this eventuality. The proposed design does not indicate any restraints that could prevent flotation.

Other issues of concern include the possibility of a reaction between graphite and molten UO<sub>2</sub>. General Atomic has indicated that such a reaction

was believed to occur and probably was endothermic. Further study is clearly needed. If the reaction proceeds to an appreciable extent under the conditions of an accident, the result could be overpressurization of the PCRV. In that event, any carbon monoxide generated would have to be handled carefully as it could burn or even explode if it came in contact with oxygen, forming CO<sub>2</sub>. Whether such a potential exists is not clear from the information available.

Another issue concerns the possibility of a failure in the cooling system. In that event, it would be desirable to have a passive sacrificial layer present to delay the attack of the molten core on the concrete. The graphite blocks proposed in the design could serve this purpose if there is not a serious chemical reaction and if the blocks are designed to minimize problems of floatation and penetration between the blocks.

Additional information on construction details, dimensions, core construction and expected quantity of debris is needed for further consideration of safety-related issues in this reactor system.

Molten Core Containment Concepts. The molten core retention systems discussed consisted of a high-temperature crucible, a heavy metal bath and a borax bath. The high-temperature crucible was only briefly discussed and seemed to be essentially similar to the preshield and main shield discussed previously. Once again, graphite blocks were the main line of defense. Consequently, the preceding comments apply equally to the high-temperature crucible.

<u>Heavy Metal Bath</u>. The heavy metal bath concept consisted of a layer of graphite bricks covering another 2-m-thick layer of bricks of an alloy of uranium and iron lining the bottom of the PCRV. The cavity liner underneath was cooled. The advantage of this system is that the molten  $UO_2$  cannot sink through the metal once it has melted because of its high density (p =16.0 g/cc). The material has a low melting point (1100°C - 1250°C) and should freeze in the vicinity of the cooling system. The bricks should be easily fabricated, although the uranium constituent  $\frac{1}{2}$  the alloy is currently quite expensive.

Some though should be given to the maintenance of a controlled environment for this system, since the alloy may be both pyrophoric and easily corroded. If the alloy is pyrophoric, and molten core comes into contact with oxygen or water (heat could drive water out of the concrete), the result could be a metal fire or worse. At this time, there is insufficient information available to us to determine whether this possibility is a problem. Further exploration of this issue is desirable and will be considered for future study.

The concept employs metals with a low heat capacity and high thermal conductivity. The low heat capacity of the metal means that it will be easily melted by the molten core without absorbing much heat. Since the metal is also a good heat conductor, most of the heat load will be transmitted directly and quickly to the cooling system. A thin thermal barrier will be employed to reduce the heat load on the cooling system. However, in the event of a cooling system failure, this sytem would provide little protection to the PCRV concrete from a high heat flux transmitted from the molten core. The heat flux could cause concrete decomposition or even lead to attack on the concrete by the molten metal of the bath. Nothing was said by General Atomic of the consequences of a cooling system failure.

Borax Bath. The borax bath system employed a 2-m-thick layer of borax bricks sealed in stainless steel cans covering the bottom of the cooled PCRV. Calculations indicated that this material would provide ten hr of protection. Its advantages include high neutron absorption, low cost and high heat capacity.

There are two main concerns about a borax system. Its low boiling point  $(1600^{\circ}C)$  makes a vapor explosion possible if it comes into contact with molten  $UO_2$  (melting point, 2750°C). In addition, it has low density (2.5 g/cc), so that it could float on top of a molten  $UO_2$  layer. Also, it might be particularly susceptible to attack by a jet of molten  $UO_2$  or steel. Many of these concerns may be unfounded, but the expected behavior can only be resolved by experimentation.

#### CONCLUSIONS

Before judgments can be made on the several interesting ideas presented, additional information on the property designs is needed, and the concepts should be supported by appropriate experiments. The designs in each instance have promising features, but all could be improved by the addition of an underlying layer of a high temperature, chemically inert, sacrificial material co protect against cooling system failure. In each concept, General Atomic should be specifically asked about the consequences of a cooling system failure.

#### VII. SODIUM-CONCRETE INTERACTIONS

As of April 1979, 44 sodium-concrete interactions tests had been performed at Hanford Engineering Development Laboratory (HEDL) and Sandia Laboratories. These tests examined concretes similiar to those used at the Fast Flux Test Facility (FFTF) and proposed for use at the Clinch River Breeder Reactor Plant (CRBRP). The information developed is of considerable interest in LMFBR safety-related programs.

The experimental results varied considerably, even under what appeared to be similar initial conditions. This variation is not completely understood and is the principal reason for concern about predicting long-term effects of a large sodium spill.

#### A. REVIEW OF SODIUM-CONCRETE FXPERIMENTS

The small-scale HEDL experiments have investigated the penetration of liquid sodium into approximately equal numbers of limestone, basalt and magnetite concrete specimens. Usually, 50 lb of liquid sodium were poured on 1-sq ft area specimens. Typically, the HEDL pours were made at higher temperatures than those at Sandia; in a majority of the HEDL tests, sodium was poured at 871°C. HEDL has conducted one large-scale, 1000-lb sodium pour on a faulted liner in which firebricks separated the sodium from the basalt concrete.

Although Sandia initially concentrated on studies of the interactions between sodium and limestone concrete, recent experiments have examined basalt and magnitite concrete interactions with sodium. Fewer tests have been performed at Sandia than at HEDL, although the experiments have been on a significantly larger scale. Typical experiments have involved pouring approximately 180 kg of liquid sodium on specimens with areas of about 1 sq m. The pouring temperature of the sodium in the experiments has varied from 450°C to 760°C. Recent experiments have included steel liners and FFTF firebricks covering the concrete.

Results from the HEDL experiments are summarized in Table VIII, and results from Sandia's tests are shown in Table IX. The tables include all

(All tests are listed by type of concrete)

Test Number	Concrete Type/ Thickness, CM	Surface Area Orientation m <sup>2</sup>	Temperature Ave/Max °C	Sodium Mass, kg/ Depth, cm	Exothermic Reaction	Reaction Time (hr)	Hydrogen Evolven kg	Penetration Max/Ave, CM	Noises (Pops, Bumps, etc)
SC-4	Limestone 30	0.092 Horizontal	677 802	22.7 25.4	Yes	8	0.18	8.4/6.1	Yes
SC-5	Limestone 30	0.092 Horizontal	871 871	22.7 25.4	No	2	0.10	4.3/2.5	No
SC-6	Limestone 30	0.092 Horizontal	871 871	24.4 27.3	No	8	0.26	4.8/3.8	No
SC-8	Limestone 30	0.092 Horizontal	871 871	23.6 26.4	Yes	24	0.28	5.3/4.4 15 cm dam.	No
SC-10	Limestone 30	0.092 Vertical	871 1093	18.1 20.3	Yes	8	0.25	13/8.9	Yes
SC-12	Limestone 30	0.092 Vertical	871 954	19.1 21.4	Yes	24	0.23	14/9.2	Yes
SC-13	Limestone 30 3.15 Kg NaOH	0.092 Horizontal	871 932	22.7 25.4 NaOH 24 Hrs	Yes	26	Yes	7.6/4.4	Yes
SC-14	Limestone 30 3.15 Kg NaOH	0.092 Horizontal	871	22.7 25.4 NaOH 0 Hr	-	24	Yes	7.6/5.7	?
SC-18	Limestone 30	0.092 Horizontal	871	3.15 NaOH + con- crete chunks	~	8	No	5.0/2.5	?

## Table VIII. HEDL Sodium-Concrete Interaction Tests (Continued)

Test Number	Concrete Type/ Thickness, CM	Surface Area Orientation m <sup>2</sup>	Temperature Ave/Max °C	Sodium Mass, kg/ Depth, cm	Exothermic Reaction	Reaction Time (hr)	Hydrogen Evolven kg	Penetration Max/Ave, CM	Noises (Pops, Bumps, etc)
SC-19	Limestone 30 3.15 Kg NaOH	0.092 Horizontal	677	22.7 25.4		8	Yes	6.4/3.8	7
SC-15	Basalt 30	0.092 Horizontal	871	22.7 25.4	÷ .	2	Yes	4.4/3.2	No
SC-16	Basalt 30	0.092 Horizontal	871	22.7 25.4	-	8	Yes	7.6/7.0	No
SC-17	Basalt 30	0.092 Horizontal	871	45.4 ? Na leak	-	24	Yes	7.0/5.7	- 1
SC-20	Basalt 30 3.15 Kg NaOH	0.092 Horizontal	500	22.7 25.4 NaOH@O hr	•	8	Yes	0.6	,
SC-22	Basalt 30	0.092 Horizontal	871 829	22.7 25.4	-	24	Yes	7.6	No
SC-23	Basalt 60	0.092 Horizontal	871 860	45.4 25.4 Na leak		16		4.4	
SC-24	Basalt 30	0.092 Horizontal	593 No Heating	22.7		Cooled rapidly		1.3	-
SC-25	Basalt 60	0.092 Horizontal	871 871	45.4 Na leaks	-	100		Significant side wall attack 16.5	-

Test Number	Concrete Type/ Thickness, CM	Surface Area Orientation m <sup>2</sup>	Temperature Ave/Max °C	Sodium Mass, kg/ Depth, cm	Exothermic Reaction	Reaction Time (hr)	Hydrogen Evolven kg	Penetration Max/Ave, CM	Noises (Pops, Bumps, stc
SC-1	Magnetite 30	0.092 Horizontal	677 677	22.7 25.4	No	8	0.18	2.8/1.9	No
SC-2	Magnetite 30	0.092 Horizontal	871 871	23.6 26.4	No	2	0.08	4.1/1.6	No
SC-3	Magnetite 30	0.092 Horizontal	871 960	24.0 26.9	Yes	8	0.28	4.8/3.4	Yes
SC+7 30	Magnetite Horizontal	0.092 871	871 27.4	24.5	No	24	0.29	6.9/4.5	No
SC+9	Magnetite 30	0.092 Vertical	760 996	19.5 21.8	Yes	8	0.31	14.7/7.7 and 15 cm hole	Yes
SC-11	Magnetite 30	0.092 Vertical	871 927	18.1 20.3	Yes	24	0.36	12.4/8.6	Yes
LFT-1	Basalt under 1.6 cm steel liner 0.3 cm gap	0.002 hole Horizontal (5 cm hole in plate)	871	22.7	-	8	2	3.8	
LFT-2	Morex firebrick Insulating brick Mortar Basalt	0.002 hole Horizontal cm hole in plate	871	22.7	-	8	1	6.4 cm into 7.6 cm firebric	k -
LFT-3	Limestone Steel liner with 4-in, gap	0.002 hole Horizontal 3/4-in. Mg0 aggregate	871	22.7		8		5.7/1.9	
	1.6 cm steel 0.6 cm gap 7.6 cm firebrick 0.6 cm mortar Basalt	0.018 hole (0.85) 5 gauge	704 860	454 56	100 hrs Na cons @70-80	umed	H <sub>2</sub> @113/min max	14 cm max@ctr	

Table IX. Sandia Sodium -- Concrete Interaction Tests

(All tests are listed by type of concrete)

Test Number	Concrete Type/ Thickness, CM	Serface Area Orientation m <sup>2</sup>	Temperature Ave/Max °C	Sodium Mass, kg/ Depth, cm	Exotuermic Reaction	Reaction Time (hr)	Hydrogen Evolven kg	Penetration Max/Ave, CM	Noises (Pops, Bumps, etc)
1	CRBRP Limestone 30.5	0.29 Horizontal	550 800	21 8.6	Yes	22 min	Yes	8.3	Explosion
2	CRBRP Limestone 38.1	1.17 Horizontal	550 800	108	Yes	45 min	Yes	9.1/7.6	Yes
3	CRBRP Limestone 38.1	1,47 Horizontal	550 740	186 15	Yes	3 hr	Yes	15	Explosion Large spalled chunk
4	CRBRP Limestone 38.1	0.55 Horizontal	450	188 30	No	4 hr (8 min)	Yes	0.5	Yes (once)
5	CRBRP Limestone	1.17 Horizontal	540 460	186	No	2 Hr	7	slight (0.5)	No
6	No. 5 reused + 36 Kg NaOH		700	186	No	×	?	slight	No
8	CRBRP Limestone	1.17 Horizontal	550	127 15	Moderate	-	7	-	90 ( B. ).
9	CRBRP Limestone	Horizontal	600	182 35	Yes		Yes-H <sub>2</sub> expl. T + 2 min		Sec. 1973
10	CRBRP Limestone	Horizontal	625	182	-	-	Yes-H <sub>2</sub> expl. T + 30 sec		- 1

# Table IX. Sandia Sodium -- Concrete Interaction Tests (Continued)

Test Number	Concrete Type/ Thickness, CM	Surface Area Orientation m <sup>2</sup>	Temperature Ave/Max °C	Sodium Mass, kg/ Depth, cm	Exothermic Reaction	Reaction Time (hr)	Hydrogen Evolven kg	Penetration Max/Ave, CM	Noises (Pops, Bumps, etc)
7	Balsalt	Horizontal	485	45	No	-	Yes	Slight	No
11	Basalt 38.1	1.6 Horizontal	760 600	128 15	Yes	4.5 hr Na limited	Yes	18/5	Yes
14	Magnetite +Bricks +Liner	1.17 Horizontal	639 870	128 13	Yes	1.8 hr Na limited	Yes	? severe > 15	Yes
12	l cm steel liner + insulating firebrick + Basalt concrete	0.65 Horizontal	600 865	69.9-45.5 30+20	Yes	10.5 hr (exp) Na limited	Yes	?severe cracks leaks	Yes
3	Steel liner Firebrick-14 cm Basalt-38.1 cm	0.55 Horizontal	600 925	182 25	Yes	10 hr ? 12 hr Na limited	Tes	14 cm brick +38.1/25 cm (total penetration) 52.1 cm total	Yes

tests conducted prior to April 1979. Blanks in some columns for certain tests indicate unreported data. In the two tables, concretes are grouped together by type (limestone, basalt, magnetite). The specimen thickness, area and orientation are provided. Sodium temperatures, masses and depths are also shown. Information is given on the test duration, whether or not an exothermic reaction was observed, hydrogen evolution, extent of penetration and whether or not noises were observed suggesting a vigorous interaction.

Historically, there appeared to be a substantial difference in the results obtained by Sandia and HEDL. In the first three large-scale Sandia tests, highly energetic reactions were observed which quickly consumed all the sodium and penetrated as far as 15 cm into the limestone concrete within 3 hrs. In the first seven HEDL small-scale tests (1 ft<sup>2</sup>) on magnetite and limestone concrete, penetration of less than half this depth occurred over 24 hr with an excess of sodium. All tests were made on horizontal surfaces.

The differences in results were initially attributed to scale effects, since the surface areas in the Sandia tests ranged from 3 to 15 times greater than those of HEDL. S'gnificantly, the sodium pool depths in the initial Sandia sodium pours were approximately half those used by HEDL. Also, the initial HEDL sodium temperatures were 300°C higher than those used by Sandia; it was thought at the time that the higher temperatures would produce more energetic reactions.

Following these initial tests, the experimental results changed. The fourth Sandia limestone concrete test employed a  $0.5-m^2$  surface area and twice the previous sodium depth. A penetration depth of only 0.5 cm was observed. This was followed by six more tests that produced little penetration but some experimental difficulties, such as hydrogen deflagrations or explosions. HEDL conducted two limestone and two magnetite concrete tests using vertical test surfaces and observed penetrations that came close to 15 cm. HEDL observed that the penetration into a vertical surface exceeded that into a horizontal surface by a factor of two. On the other hand, Sandia observed greater penetration downward than radially.

During this period, sodium-limestone concrete interactions were of considerable interest because limestone concrete had been proposed for use in the CRBRP. At this point, Sandia proposed a mechanism to explain the different results obtained at Sandia and HEDL. It was suggested that NaOH rather than sodium was primarily responsible for the observed attack on concrete. The NaOH formed when sodium reacted with water driven from the concrete by heat. It was suggested that the concrete would experience little attack during the period before the sodium became saturated with NaOH. When additional NaOH was formed, the liquid NaOH formed a separate layer, which in turn attacked the concrete. It was argued that the shallow pools used in the first three Sandia limestone concrete tests quickly saturated with NaOH allowing the energetic reaction to begin promptly. The deeper sodium pools used by HEDL, and in Sandia test No. 4, required time to reach saturation with NaOH. In addition, it was suggested that reaction products would provide some shielding from further attack for the concrete. This poposed mechanism led both Sandia and HEDL to perform tests in which NaOH was added to the liquid sodium before it came into contact with limestone concrete in order to achieve immediate saturation. Sandia's test with NaOH (test No. 6) showed a lesser reaction than before, and HEDL's tests (tests SC-13, SC-14 and SC-19) showed about the same penetration. These results did nothing to confirm the NaOH attack hypothesis.

#### 1. BASALT CONCRETE

Recent emphasis has shifted towards study of the sodium-concrete interactions expected in the type of basalt concrete that is being used in FFTF. HEDL has conducted nine small-scale basalt concrete tests at temperatures from 500°C to 871°C. With one exception, the total penetration in these tests varied from 3.2 to 7.6 cm but could not be correlated to either test duration or temperature. All the successful sodium tests were performed at 871°C. The total penetration data for the 2.8- and 24-hr tests are conflicting, with the 24-hr data showing less penetration in one case than was indicated in the 8-hr tests. However, the 100-hour test showed a 16.5-cm penetration, suggesting continuing attack for a long period. Furthermore, the sodium leaked out before the conclusion of the experiment so the test was probably sodium

limited. HEDL also conducted two lower-temperature tests with an initial charge of NaOH added to the sodium. The penetrations observed in these tests were shallower than penetrations in tests with sodium alone.

When Sandia test No. 11 is considered, the basalt concrete data look much worse. In that test, penetrations as deep as 18 cm were observed in a 4.5-hr test at 600°C. The average depth of penetration over the entire bottom of the crucible was 5 cm. Sandia also has conducted two tests with basalt concrete protected by firebrick and a steel liner. Substantial penetration and severe cracking occurred during both tests. All the Sandia basalt tests were limited by exhaustion of the sodium supply, so it is not possible to say how far penetration might have proceeded.

Posttest examination of the reaction products has shown differences that indicate two different reaction mechanisms are at work. In the quiescent HEDL high-temperature horizontal tests, a dense, hard layer - apparently  $Na_2O$ formed between the unreacted sodium and the concrete below. This layer is believed to provide the concrete some degree of protection from further attack. On the other hand, the reaction zone in the lower temperature Sandia tests is a porous, cinder-like sodium silicate.

Until April 1979, nine tests had been conducted at HEDL with sodium and bare basalt concrete, and three with liners. Sandia had completed one bare basalt concrete test and two tests with liners and firebricks providing protection from sodium.

It is useful to compare the effect of different parameters on these tests. The following sections give our interpetations of the preceding test results.

#### a. Sodium Temperature

The temperature of the sodium pool in most of the relatively quiescent HEDL tests was much higher than that of the relatively energetic Sandia test when the exothermic reaction started. The HEDL tests were maintained at 871°C, whereas the Sandia basalt test was conducted at 760°C. At both of these temperatures, water is driven from the concrete. However, it has been

suggested by Sandia that, at the lower temperature, water reacts with sodium to form NaOH and NaH. At higher temperatures, Na<sub>2</sub>O alone is formed. NaOH is a liquid at these temperatures and attacks concrete. In addition, the reaction products are soluble in NaOH so that there is little opportunity for a protective layer to form. On the other hand, at higher temperatures, the reaction generating Na<sub>2</sub>O is apparently favored. Although Na<sub>2</sub>O can attack concrete, its potential is limited as it is a solid at these temperatures. This seems to be one instance where the chemical environment is more severe at lower temperatures than a higher temperatures. It should be noted that the temperature of the sodium is elevated to 800°C or higher by the exothermic reaction once it starts. There is also some evidence from thermocouple data that the areas in which the reactions occur can reach temperatures in excess of 1000°C.

There is a flaw in the foregoing explanation of the difference between the HEDL and Sandia tests. In HEDL tests SC-20 and SC-21, 3.15 kg of NaOH were added to sodium at temperatures of 500°C and 649°C, respectively. This quantity should have saturated the sodium. Under the hypothesis advanced, penetration should have been extensive. However, the observed penetrations were only 0.6 and 2.5 cm, respectively. There are other factors that may affect the extent of penetration; one of these is sodium pool depth.

#### b. Sodium Pool Depth

If the concrete is principally attacked by NaOH, rather than by sodium, a longer time will be required for attack to occur in a deeper pool. A longer time is required in order to saturate the sodium with NaOH when the pool is deep. The NaOH has only limited contact with the concrete until the pool is saturated. At that time, a separate NaOH layer forms in contact with the concrete allowing the attack to proceed expeditiously. In the Sandia shallow pool test, this saturation would have occurred relatively quickly, leading to rapid penetration. In tests with deeper sodium pools, a longer time should be required for saturation in this temperature range.

A problem exists if this hypothesis is used to explain the lack of a reaction in HEDL tests SC-20 and SC-21, which were the only HEDL tests in the same temperature range as Sandia's tests. In these tests, NaOH was added to the sodium in a quantity that should have assured initial saturation. Consequently, the results in tests SC-20 and SC-21 remain unexplained.

#### c. Failed Liner Tests

Experiments have been conducted to simulate the effects of a failed steel liner. The steel liner is part of a system consisting of the liner and firebricks and is intended to protect the underlying basalt concrete from liquid sodium. Failure of the steel liner has been simulated by the intentional introduction of a hole in the liner. In each of the tests, sodium reacted with the firebricks and, in some of these tests, the sodium was consumed before the bricks were totally penetrated. In some of the later tests at Sandia using more sodium, the reaction continued into the concrete, apparently until all the sodium was consumed. In tests at HEDL, less concrete penetration was observed, and excess sodium remained.

The reaction products expanded considerably, exerting extreme pressures on the containing surfaces. The pressure bent the "failed" steel plate upward and forced reaction products through the hole. In addition, the expansive forces produced under these conditions can cause serious cracks in the concrete, allowing sodium to penetrate to the bottom of the concrete crucible. In the last tests at Sandia, the pressure caused failure of all the rebars as well as severe cracking in the basalt and magnetite concrete structures.

Further, the reaction products appeared to be porous and provided little or no barrier to the flow of sodium to the reaction zone. In Sandia test No. 12, additional sodium was added to the top of the reaction mass, and the exothermic reaction was restarted shortly thereafter. This demonstrates clearly that the reaction products are porous to sodium and offer no protection to the concrete below.

In reviewing the sodium concrete data, it appears that a major constituent in the fire-resistant brick and insulating brick is involved in the chemical reactions with liquid sodium. The first choice candidate is  $SiO_2$ , which is also present in the basalt concrete. Sodium reacts with silica and forms a host of complex chemical compounds involving the rest of the components of

concretes. Some of these compounds have low melting points, while the behavior of other compounds is not easily predicted owing to their complex phase diagrams. It would appear best to avoid, to the extent possible, the inclusion of siliceous materials in the reaction zone and to replace the fireresistant and insulating bricks with a material such as MgO, which is inert in liquid sodium.

#### d. Cracking

HEDL and Sandia have observed differing degrees of cracking. Cracking is critical because it reduces strength, provides a pathway for sodium to reach and attack fresh concrete, and provides a potential escape route for gases, such as hydrogen, generated by sodium reactions.

HEDL's tests with basalt concrete indicate little or no evidence of cracking except for SC-25. On the other hand, Sandia observed substantial 1/8-inch radial cracking in the bare basalt test (test No. 11) and very severe cracking in test Nos. 12 and 13, where firebricks and liners were also present. In these tests, sodium preferentially penetrated into cracks as deep as 20 cm.

HEDL has asserted that the cracking observed by Sandia results from thermal expansion in the Sandia crucible geometry. In test No. 11, the HEDL interpretation has some merit. However, since only hairline cracks were observed in similar crucibles of limestone concrete (admittedly a different material), factors other than thermal expansion may be involved. In tests No. 12 and No. 13, the pressure exerted by the voluminous reaction products trapped between the steel liner and the concrete almost certainly was a major contributor to the severe cracking.

In the actual reactor system, firebricks and a steel liner are present and, consequently, reaction products may possibly form, generating pressures that could cause cracking. Cracking is also possible at the point where the reactor room floor and sidewalls meet. Consequently, the problems that cracking may cause should be given further consideration.

#### e. Penetration Rates

Under the conditions of the 2-, 8-, and 24-hour HEDL tests, the depth of sodium penetration into concrete ranged from 3.2 to 7.6 cm. In HEDL test SC-25, however, a penetration of 16.5 cm was observed over 100 hr at 871°C. Sodium leaked away at some point during the test, so that it is not possible to determine if the penetration would have proceeded further.

In Sandia test No. 11, a penetration of 18 cm was observed after only 4.5 hr at 600°C. There is no way of knowing how far the sodium would have penetrated in this test because the supply was exhausted. In Sandia test No. 13, sodium completely penetrated through 14 cm of firebrick and 25 cm of concrete in ten hours at 600°C. Although the reaction was sodium limited, some sodium penetrated entirely through the 38-cm-thick basalt specimen along a crack and was found underneath the crucible. From information supplied by thermoccuple temperature excursions, Sandia determined that the reaction front advanced into the concrete in test No. 13 at a rate given by D = 1.6 t<sup>1/12</sup>, where D is the pentration distance and t is time in minutes.

#### f. Hydrogen Generation

The explosions at Sandia demonstrate the problems that can occur if hydrogen escapes through cracks or other paths and collects elsewhere. When a proper air mixing ratio and ignition source appear at the collection point, a deflagration or explosion can result.

Substantial hydrogen generation can be expected from reactions between sodium and water from the concrete. The hydrogen data obtained in HEDL test LFT-5 are not conclusive because of the limited sodium penetration beyond the firebricks into the concrete.

#### 2. COMPARISON OF BASALT AND LIMESTONE CONCRETES

The penetration rates for sodium attacking limestone and basalt concrete under the conditions of the experiments at HEDL are similar. Some experiments at Sandia indicated that sodium will penetrate limestone concrete more rapidly than basalt concrete under certain conditions. However, the basalt test specimen (test No. 11) at Sandia displayed greater cracking than similar limestone specimens. Although arguments have been made that this cracking was caused by the effects of geometry and thermal expansion, these arguments are speculative since both the limestone and basalt specimens at Sandia had the same geometry.

Any comparison of the concrete's behavior during a postulated accident should also include consideration of concrete attack by molten core debris. Experiments with molten  $UO_2$  on both concretes suggested that limestone concrete was more resistant to attack by molten core debris, probably because of its higher melting point and the extensive evolution of  $CO_2$ , which can carry away some of the heat.

Data available today do not conclusively indicate that one concrete is better than the other. Limestone concrete seems to be more crack-resistant than basalt concrete, but it may be more easily penetrated by sodium. Limestone concrete appears to be more resistant to molten  $UO_2$ .

#### B. RECENT EXPERIMENTS AT SANDIA AND HEDL

A few additional tests have been completed after the completion of the previous section. In Sandia liquid sodium-concrete test No. 15, 527 lb of liquid sodium at 688°C were poured into a square cavity in a basalt concrete cylinder containing two experiments. On one wall of the cavity, a 3/8-in... thick low carbon steel liner covered the basalt concrete. The liner contained a flaw which admitted limited quantities of sodium. At the opposite wall, another intentionally flawed liner covered a double layer of Kaiser Morex fire-resistant firebrick (used in FFTF), which in turn covered the concrete wall. The two other walls and the bottom of the cavity were covered by the steel liner and a layer of MgO, which served to protect the concrete from the sodium. An exothermic reaction was observed during the course of the experiment, but the interaction appeared to be more benign than in previous tests.

HEDL has conducted a series of experiments<sup>7</sup> to examine the iteractions between liquid sodium and dehydrated basalt concrete. In the first of these "special effects tests," SETI, a basalt concrete specimen was dried for 24 hr at 427°C to eliminate 80 percent of the moisture initially present in the concrete. When 24 kg of liquid sodium at 879°C was poured on the specimen, a penetration of 27 cm in 2 hr was observed. An exothermic reaction accompanied the rapid penetration and the reaction product was a loose, friable material.

In a second test, SET2, the concrete specimen was dried for 48 hr at 200°C. This process removed from 30 to 50 percent of the water originally present. Again, 24 kg of sodium at 870°C were poured on the specimen. Rapid penetration occurred to a depth of 7.5 cm during the first hour. Thereafter, the experiment was quiescent for 6 hr when rapid penetration occurred down one side of the specimen, perhaps because of sodium penetration between the specimen and its holder. The reaction products were again a loose, friable material.

HEDL conducted SET3 to study what was thought to be a protective reaction barrier phase. Fifteen kg of NaOH were placed on a dehydrated concrete specimen like the one in SET1, from which 80 percent of the water originally present had been removed. The NaOH was gradually heated over 6 hr to 760°C and penetrated 4 cm into the concrete during an 8-hr test. An ll-cm-thick layer of reaction products formed with a consistency described as being similar to sandstone. This hard reaction product, very different from the products that formed in SET1 and SET2, had to be chipped away with a chisel.

Finally, in SET4, sodium was maintained at 600°C for 50 hr and then at 870°C for 50 hr on a hydrated concrete specimen. A maximum penetration of 7.7 cm was observed after 100 hr. Large amounts of hydrogen were generated and, as in SET3, the reaction product was like stone.

It was concluded by HEDL that the reaction between sodium and water to form NaOH and hydrogen was the dominant reaction. Water, deep inside the concrete, was thermally released and migrated by a pressure-driven flow to the hot surface where it reacted with sodium, forming NaOH, the major reaction product. HEDL has proposed that the rate of heat transport into concrete, rather than the rate of sodium penetration, determines the rate of water transport and hydrogen release.

Using hydrated concrete and an excess of reactants, only limited penetration has been observed with the formation of a hard stonelike reaction product. HEDL believes that a protective barrier limited penetration; a steampressure barrier phase was present caused by water driven from the concrete.

The steam then reacted with sodium to form a NaOH layer and large amounts of hydrogen. HEDL further believes that "mechanical effects" - presumably cracking and spallation-may allow penetration of the protective barrier in some tests, so that sodium can rapidly penetrate into the dehydrated concrete. This stage of the process would produce little hydrogen or energy release.

The explanation suggested by HEDL for the greater penetration observed in some tests is the opposite of that proposed by Sandia. HEDL suggests that NaOH does not rapidly penetrate basalt concrete and that rapid penetration occurs when sodium comes in contact with dehydrated concrete. The Sandia position most recently has been that basalt concrete is attacked to a greater degree by NaOH than by sodium. However, in tests with NaOH added to sodium on comprete to insure the presence of a NaOH layer, only limited penetration was observed. Although the Sandia results appeared to show that saturation of sodium with NaOH was important in reactions with limestone concrete, the chemistry in the case of basalt concrete is presumably different and data are insufficiant to support a similar conclusion for basalt concrete.

The experimenters at Sandia have found evidence that contradicts the explantion of the available sodium-concrete data proposed by HEDL. In test No. 13, Sandia found that 24 percent of the reaction product inside the liner was NaH. They feel that NaH could only be formed if hydrogen from the sodium-water reaction (forming NaOH) was present; therefore, attack by sodium on dehydrated concrete cannot account for the observed erosion in their experiments.

If the sodium-water reaction is dominant, as proposed by HEDL, then the water content of the concrete may be highly significant in determining the penetration rate and may contribute substantially to the observed differences. At both laboratories, water was added to obtain a certain "slump" in the "slump test" widely employed in the construction industry. However, obtaining consistent results in these tests will presumably require better control over the concrete water content. The water content of the constituents should be determined prior to mixing, rather than by use of the slump test, and additional water should be added so that the laborato is have the same initial total water content in their concrete.

#### C. CONCLUSIONS

After reviewing sodium-basalt concrete interactions, we have reached the following conclusions:

- A sodium-baselt concrete penetration rate of l-in/hr is recommended. In the absence of firm evidence indicating when sodium penetration will end, we continue to feel that a continuous penetration rate is appropriate.
- 2. The firebricks employed in these tests are not a barrier to sodium. Indeed, this particular firebrick-liner combination may be detrimental if it comes into contact with liquid sodium. The pressure generated as reaction products form may cause the concrete to crack. Firebricks that are inert to sodium should replace those selected. Siliceous firebrick should be avoided.
- 3. It is difficult to determine whether basalt concrete is better than limestone concrete in a sodium environment. Basalt concrete seems to be somewhat more resistant to penetration by sodium, but it also seems more likely to crack than limestone concrete.
- The questions of the effects of scale, geometry and mode of restraint remain unresolved.
- 5. Sodium pool temperature appears to affect the chemical reactions that occur and the reaction products that form. A lower temperature sodium environment (600°C) seems to have a more severe effect on basalt concrete than the boiling sodium environment.
- Sodium pool depth may affect the time required for the occurrence of a reaction and the extent of reaction.
- Dehydration of the concrete may affect the extent of sodium penetration.
- The chemistry of the sodium-basalt concrete and sodium-limestone concrete interactions are speculative. Hypotheses have been proposed that seem reasonable but are as yet unconfirmed.
- The physical aspects (cracking, spallation) of the sodium-basalt and sodium-limestone concrete interactions are poorly understood.
- 10. Further research is needed on both chemical and physical aspects of sodium-concrete interactions. Given the differences between the experimental conditions, it is difficult to coherently correlate the results. More detailed coordination in selecting experiments and experimental parameters for study would be desirable.

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