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RESPONSE OF SECONDARY CONTAINMENT TO
PRESENCE OF SODIUM AND HYDROGEN

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SUMMARY

As part of an effort to demonstrate that the risk to the public from extremely low probability events in liquid metal fast breeder reactors is bound within an acceptable envelope, containment pressurization by sodium and hydrogen was evaluated. Temperature and pressure histories are presented for typical sodium spray and pool fires and sodium vapor reactions. A review of mechanisms for hydrogen generation and recombination as well as limit for flammability and autocatalytic recombination is provided, and general containment design options to reduce risk are discussed.

1. Introduction

Within the safety research and development program for liquid metal fast breeder reactors, directed by the U.S. Department of Energy, an effort is made to demonstrate that even for extremely low probability core disruption or melting events, the reactor containment and confinement building provide sufficient retention and natural mitigation so that the risk to the public from releases of radioactivity is bound within an acceptable envelope. It is recognized that the major contribution to risk, given a core-disruptive event, would arise from the failure of containment and uncontrolled releases of radioactive materials to the environment. Therefore, containments are designed to accommodate loading conditions of extremely low probability events. The following loading conditions are expected to present the major challenge to containment integrity: (1) short-term pressurization by sodium spray or pool fires following energetic ejection of sodium into the containment and (2) long-term pressurization by sodium vapor and hydrogen after failure of the reactor vessel and guard tank. In this paper, the pressure and temperature responses of a typical LMFBR containment to the presence of sodium and hydrogen are described and scenarios and phenomena that could lead to containment pressurization or pressure mitigation are reviewed. In the concluding section, some design measures are discussed that could be effectively used to improve containment reliability, if necessary.

2. Accident Scenarios

Maximum containment loading conditions are expected from extremely unlikely accident sequences for which neutronic shutdown occurs by core disruption. The energy generation and rate at which such mechanisms proceed can vary significantly depending on the rate of reactivity insertion. At the high end of the spectrum are energetic core dispersions, at the lower end are nonenergetic melting events. The common characteristic of these events is that core debris and sodium may penetrate through the primary system boundary and interact directly with the containment atmosphere and concrete structures. The following scenarios may exist.

Energetics:

During the energetic core-dispersion phase, fuel sodium bubbles may be discharged into the sodium pool and sodium slug may impact on the vessel head or thermal shield plates. Depending on the magnitude of the pool impact, various head responses may result such as head lift and doming, rotating plug jump and failure of seals, control rod drive ejection, failure of instrument tubes or breach of in-vessel transfer machine ports. If leak paths should develop, the two-phase mixture may then be forced under the temporarily high pressure into the reactor containment building (RCB). Ejection could occur by: (1) discharge

of two-phase mixtures through vertical passageways, such as control rod housings, which could provide a direct and nonobstructed flow path to the RCB, and (2) ejection primarily in radial direction through a gap generated, for example, by rotating plug jump. In the first event, a sodium spray fire may ignite in the containment air atmosphere, whereas event (2) would very likely lead to a pool fire. In both events, sodium and core debris would collect on and interact with the concrete floor of the RCB.

Core Meltdown:

If a core meltdown event is postulated, core debris and sodium may penetrate through the bottom of the reactor vessel and guard tank following creep rupture failure or meltthrough, and drain into the reactor cell. Since the heat generation from radionuclide decay and sodium-concrete interactions exceeds the heat removed by conduction into the concrete walls, the sodium pool will reach the boiling point (on the order of 10 hours) and sodium vapor along with hydrogen from sodium-concrete reactions will be transported to the containment and heat transport system cells.

3. Hydrogen Generation and Effects on Containment

Hydrogen in the containment is generated by exothermic chemical reactions between metals such as sodium or molten steel and water. The dominant source for hydrogen is the sodium-water reaction, which, for the postulated scenario, may take place in the reactor cell or the containment building. The rate of reaction is mainly determined by the rate at which water is released from concrete. The water existing as moisture in the containment atmosphere is instantaneously available for reaction with sodium.

Concrete contains approximately 8 weight percent of water in the form of capillary water, adsorbed and interlayer water, and chemically bound water which are released at specific temperatures (see Table I). The rate of water release from concrete has been determined experimentally in small scale and intermediate scale tests[1,2] and with analytical models[3,4] that are based on diffusion and filtration mechanisms (mass flow under pressure gradient) for water vapor transport. In general, the diffusion caused by concentration gradients controls the water release below the boiling point of water, above this temperature, filtration of vapor and more significant releases can occur.

The sodium-water reactions, reaction products and energy releases are summarized in Table II. For the first reaction to proceed, it is required that the quantity of sodium is unlimited. For excess water, the second reaction dominates. Several computer codes, including the extended SOFIRE code[5] and CACECO[1] can be used to determine, among other things, hydrogen generation

Table I. Water Release from Concrete (Approximated)

Component	Amount, [%]	Release Temp, [°C]	Remarks
Capillary Water	0	<105	The capillary water to cement ratio increases approximately linearly with the water to cement ratio (W/C) for W/C > 0.4.
Adsorbed and Interlayer Water	50	<350	
Chemically Bound Water	50	<900	The gel water content is fairly independent of the water/cement ratio. The dehydration of calcium silicate hydroxides in β -dicalcium silicate (Ca_2SiO_4) occurs between 100 and 871°C. The dehydration of calcium hydroxide occurs at 480°C.

Table II. Sodium-Water and Sodium-Hydrogen Reactions

Chemical Reaction	Enthalpy Change ΔH^* (cal/mole of reaction), Temperature in °K	Remarks
$2\text{Na}(\text{l}) + \text{H}_2\text{O}(\text{g}) \rightarrow \text{Na}_2\text{O}(\text{l}) + \text{H}_2(\text{g})$	$-44750 + 7T$	Spontaneous reaction.
$\text{Na}(\text{l}) + \text{H}_2\text{O}(\text{g}) \rightarrow \text{NaOH}(\text{l}) + \frac{1}{2}\text{H}_2(\text{g})$	$-44030 + 6.5T$	Secondary reaction with excess H_2O .

*Reactions with $\Delta H < 0$ are exothermic.

rates for the scenarios described in Section 2. With the extended SOFIRE code, short-term sodium-water reactions can be calculated for a horizontal concrete and sodium pool interface. With the CACECO code, two possible processes for hydrogen generation following reactor vessel meltthrough can be evaluated (1) sodium-concrete interactions at horizontal and vertical surfaces with completely or partially failed liners, and (2) sodium vapor reactions with water vapor in the RCB or primary heat transport system (PHTS) cells. It should be noted that the existing analytical tools provide rough estimates only and should be used with caution when applied as a basis for containment or containment equipment design evaluations because of significant limitations in modeling of

sodium-concrete interactions, chemical reactions in cells, and water release from concrete.

The containment atmosphere contains oxygen that can autocatalytically combine with hydrogen should the hydrogen concentration reach 19%.* (See Fig. 1.)

Fig. 1. Flammability Limits for Hydrogen-Air-Moisture Mixtures.

To avoid hydrogen explosions, devices are usually included in the containment designs to purge the atmosphere so that the hydrogen concentration is below the flammability limit. This procedure, however, is highly undesirable because it could lead to significant radioactivity releases to the environment.

Flame propagation of lean mixtures of hydrogen and air when ignited can occur at low hydrogen concentrations. Burning proceeds generally in upward direction by the buoyancy of small individual flamelets. The minimum concentration of hydrogen to support such flamelets is 4% at room temperature, and this is the accepted lean limit of upward flame propagation.[6,7] Because the propagation of flame surfaces of small radius curvature in downward direction is opposed by buoyancy, the lean limit for downward propagation in hydrogen air is 8- to 9%. A steep pressure rise after ignition was observed for hydrogen-air mixtures with hydrogen concentration greater than 8%. For a H_2 concentration of 16% the pressure rise reached 5 atm.[6] Preheating of a mixture in general enhances its flammability. It can be demonstrated that the limit concentration

* All concentrations are expressed in volume percent.

becomes zero when the preheat temperature is equal to the limit flame temperature which is 828°K for hydrogen and a linear relationship appears to exist between flammability limit and preheat temperature.[8]

Under certain conditions, hydrogen transported from the reactor cell to the containment will recombine continuously with oxygen in the presence of sodium vapor so that the likelihood of hydrogen explosions is reduced. Data obtained from ignition experiments[9] indicated that a hydrogen jet discharged in air can be expected to ignite, if its temperature is above 260°C and if it contains more than 6 g of Na per cubic meter of hydrogen (see Fig. 2). After

Fig. 2. Ignition Characteristics of a Hydrogen-Sodium Vapor/Aerosol Jet.

ignition, the jet burned continuously and completely for sodium concentrations between 0 and 400 g Na/m³ H₂, atmospheric water vapor concentrations up to 25%, jet velocities from low laminar conditions to highly turbulent flow, jet temperatures from 20°C to 880°C (sodium boiling point) and jet nitrogen concentrations up to 25%.[9] The burning efficiency of hydrogen decreased exponentially with decreasing oxygen concentration and increased for higher jet sodium concentrations. Judging from the experimental data, it is expected that containment building flow paths (with rupture disk) from the reactor cell to containment building could be designed to promote hydrogen recombination.

For typical meltthrough scenarios, the oxygen concentration should decrease below the flammability limit of 10%, before a significant amount of hydrogen is generated, if hydrogen recombination takes place.[10] For energetic scenarios, oxygen depletion will occur by sodium spray or pool fires before the buildup of significant hydrogen concentrations (see Fig. 3), e.g., continuous hydrogen recombination to prevent hydrogen explosions is not required. A strategy for

Fig. 3. Mole Fractions of Hydrogen and Oxygen in Reactor Containment Building.

containment designs seems to emerge from this conclusion: an effort should be made for meltthrough events to deplete the oxygen concentration in the containment by sodium-hydrogen-oxygen reactions before a critical hydrogen concentration is reached. This would help to avoid containment purging with outside air at an early time (26 hours for CRBRP) and would, therefore, reduce the risk to the public significantly.

4. Sodium Interactions and Effects on Containment

4.1 Spray Fires

The ignition of sodium sprays depends on a number of variables such as sodium temperature, and oxygen and moisture content of the environment. Condensed sodium vapor mist can ignite even at room temperature, sprayed liquid droplets ignite at 120°C, but solid jets of liquid sodium or liquid pools may not ignite in air below 200 to 250°C. For the postulated accident conditions, sodium temperatures are expected to be significantly higher and instantaneous ignition should occur in air.

The oxidation of sodium spray in the RCB, as postulated in the energetic phase of a core-disruptive accident, involves droplet formation and motion, gas and droplet interaction, heat and mass transport at the droplet surface, and

combustion thermodynamics. The sodium-oxygen reaction rate is controlled by diffusion of oxygen to the combustion zone surrounding the droplet (see Fig. 4). The heat generated in the combustion zone is transmitted by convection, conduction and radiation to the droplet, to the surrounding atmosphere and to the containment surfaces as well.

Fig. 4. Reaction Model for Sodium Droplet.

Both the SPRAY[11] and SOMIX[12] codes can be used to evaluate the consequences of sodium spray releases into the containment building. The SPRAY code was designed to treat circulation within the containment atmosphere with the use of a quasi-two-dimensional model in cylindrical coordinates. A reaction rate constant was introduced to represent the burning efficiency assuming an instantaneous reaction. The water vapor present in the cell atmosphere was assumed to react with the sodium droplet. However the burning rate was underestimated substantially.[13]

In the SOMIX code an instantaneous reaction is assumed and the stream function-vorticity formulation is employed to determine fluid motion generated by buoyant forces which are caused by heat transfer from burning sodium and by viscous interaction between falling droplets and the surrounding gas. The SOMIX results for various amounts of sodium injection into the RCB are shown in Fig. 5.[14] The calculated maximum pressure rise was about 2.1 atm which exceeds the typical containment design pressure, but should not present a real threat to containment integrity.

Fig. 5. Containment Building Pressure Rise.

4.2 Pool Fires

For energetic events which may be postulated to result in significant vessel head or seal failures, sodium and debris material may be discharged into the containment building and a shallow sodium pool could form on the containment floor causing a pool fire and interactions with concrete. The pool temperature is expected to stabilize at a temperature of approximately 730°C independent of initial conditions.[15] In Fig. 6, the vapor and liquid phases existing during

Fig. 6. Sodium Pool Fire (Schematic).

during a sodium pool fire are illustrated. The following processes are expected to occur:[16] evaporation of sodium, diffusion of sodium vapor to the flame zone, formation of Na_2O in the flame zone, formation of Na_2O_2 in atmosphere outside the flame zone, diffusion of Na_2O and Na_2O_2 particles into the atmosphere and precipitation of reaction products and formation of a crust on the sodium pool surface. The burning of pool fire can be evaluated by the SOFIRE-II code.[5] The burning rate in that code is proportional to the oxygen concentration and is controlled by the diffusion of oxygen to the pool surface. The net energy added to the system by the burning pool fire is the heat of formation, which is the difference between the heat of combustion for each oxide and the heat of vaporization for the sodium. The energy released by radioactive decay and sodium concrete interactions, may contribute significantly to the thermal challenge of the containment (see Fig. 7). The heat generated by sodium-concrete

Fig. 7. Relative Importance of Various Heat Sources as Evaluated with the Modified SOFIRE Code.

interactions is mainly from the sodium-water reaction, because sodium penetration into the concrete was assumed to be self-limiting. Typical pressure and temperature histories for a large LMFBR calculated by the extended SOFIRE code for various postulated releases are shown in Figs. 8 and 9, respectively. As expected, cases with larger sodium and core material releases result in higher

Fig. 8. Containment Pressure Histories Following Energetic Disruption.

Fig. 9. Containment Temperature Histories Following Energetic Disruption.

gas pressure and temperatures. The maximum pressure and temperature are 1.2 atm and 530°C, respectively.

4.3 Sodium Vapor Reactions

Following meltthrough event, sodium vapor may be released from the reactor cavity and/or heat transfer cells as a result of sodium boiling. Although experimental data on sodium vapor reactions are insufficient, it is expected that

oxygen, water vapor and carbon dioxide, if present, all react with sodium vapor. Typical temperature and pressure histories for large LMFBRs were calculated with the CACECO and SPRAY codes.[17] The CACECO code evaluation is overly conservative in the model, it is assumed that the entire sodium spray is in the vapor phase and it reacts with the available oxygen with 100% efficiency. CACECO results and a comparison with SPRAY results are shown in Figs. 10 and 11. The peak pressure determined with CACECO was on the order of 2.1 atm.

Fig. 10. Comparison of Cell Temperatures Calculated by CACECO and SPRAY Codes.

Fig. 11. Comparison of Cell Pressures Calculated by CACECO and SPRAY Codes.

5.0 Concluding Remarks

Sodium is a very attractive fast reactor coolant, which allows reactor designs with a low system pressure and a high reliability, but on the other hand, any leakage of sodium into the containment can result in exothermic reactions with air and concrete that in turn can lead to pressure buildup, thermal loading and potentially hydrogen explosions in the containment. For the examined extremely low probability core melting and energetic core disruption events, peak pressure on the order of 2.1 atm and temperatures of approximately 530°C were calculated. It was shown that hydrogen explosions do not follow energetic events because of early oxygen depletion by sodium fires. For meltthrough events, hydrogen recombination in the containment following sodium jet ignition, may deplete the oxygen not rapidly enough, so that purging of the containment atmosphere may be required at an early time to avoid a hydrogen explosion.

Although temperature and pressure histories in the containment are very design and material dependent, some general guidelines could be followed in the LMFBFR containment design to minimize the risk to the public:

- (a) Minimize hydrogen generation rate by minimizing water release from concrete: This can be accomplished by insulating the concrete by a refractory ceramic such as MgO or Al_2O_3 . Cooling of concrete structures or sodium pool or preheating of concrete to remove capillary and adsorbed water would also reduce release rates. Hydrogen generation can be prevented if liquid metal-water reactions can be eliminated.
- (b) Design reactor cell-containment leak paths that promote sodium jet ignition and hydrogen recombination.
- (c) Design containment so that early venting (<24 hours) is not required, e.g., containment should accommodate peak pressures without venting. This can be accomplished by designing a containment for high pressure or large volume or by including a cooling system.
- (d) Include adequate filter systems that can accommodate sodium aerosols if venting is required to maintain containment integrity.
- (e) Purging to prevent excessive hydrogen concentrations at early times, should be avoided. In addition to the measures described under item (a), controlled injection of flammable materials into the containment could be considered to deplete oxygen early.

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