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VAPOR EXPLOSION POTENTIALS UNDER LWR HYPOTHETICAL ACCIDENT CONDITIONS

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ABSTRACT

Analytical results predict that explosive vapor formation should cease at elevated ambient pressures typical of reactor operating conditions, and for contact interface temperatures greater than the thermodynamic critical temperature for non-chemically reacting systems. Simulant fluid experiments are reported which show that such termination of explosive events does occur.

INTRODUCTION

When considering hypothetical core meltdown conditions in light water reactor safety analyses, one physical phenomenon which must be considered when molten fuel, clad, or structure materials can come in direct contact with the water coolant is a vapor (steam) explosion. Extensive experimental investigations in this subject area¹⁻⁴ have shown that in order to have such an explosive event, two liquids at greatly different temperatures must be brought into intimate contact. An additional criterion that has been proposed^{5,6} is that the interface temperature upon contact, which is evaluated by

$$T_i = \frac{T_h + T_c \sqrt{\frac{k_c \rho_c C_c}{k_h \rho_h C_h}}}{1 + \sqrt{\frac{k_c \rho_c C_c}{k_h \rho_h C_h}}} \quad (1)$$

must be greater than the spontaneous nucleation temperature of the liquid pair before such explosive events occur. Definitive experiments^{4,7} have been conducted to determine the relationship of the contact interface temperature to the explosive potential of a system, and these experiments have found that this criterion does indeed describe the onset of explosive interactions for the liquid pairs investigated.

When considering the contact temperature between molten fuel and water, molten stainless steel and water, and molten zircaloy and water, the interface temperature upon contact is far greater than the thermodynamic critical point of the coolant. Detailed evaluations of the spontaneous

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nucleation density^{8,9} and the accompanying thermal boundary layer development and energy transfer processes in explosive interactions have shown that for contact temperatures in the vicinity of the thermodynamic critical point, the spontaneous nucleation rate is so large that all intimate contacts immediately result in vapor blanketing at the interface. Consequently, it is of direct interest for a light water reactor system to evaluate whether such termination of explosive interactions will actually occur at these elevated temperatures.

In addition, if core meltdown considerations are made at nominal operating pressures, the high level of reduced pressure used in light water reactors dictates that essentially all vapor growth would be controlled by the rate at which energy could be conducted from the liquid to the phase boundary or bubble interface. If an explosion is produced by releasing high pressure vapor, i.e., vapor pressure higher than the ambient pressure, then these elevated pressure environments may provide a limiting parameter for the onset of explosive events. Again, this aspect is of direct interest to light water reactor safety analyses.

The purpose of this study was to explore these high pressure, high temperature arguments with fluid pairs that have been demonstrated by different investigations^{4,7,10} at different laboratories, to be a reproducibly explosive system when the appropriate thermal conditions are established. This simulant fluid pair of Freon-22 and mineral oil, and in the case of the high pressure experiments Freon-22 and water, provides a nonchemically reacting system in which these individual effects can be isolated.

THEORETICAL CONSIDERATIONS

HIGH PRESSURE SYSTEMS

In order to have a true vapor explosion, a system must experience nucleation of the vapor from the liquid phase and growth of this vapor phase at pressures higher than the ambient. The details of bubble growth characteristics found in Ref. 11, show that there are many considerations which must be made in understanding the complete, integrated behavior relating to the formation and growth of the vapor space. However, from a first order point of view, there are two basic regimes of interest. These are the inertially dominated growth regime and the thermally dominated regime. In inertially dominated systems, the pressure inside the vapor space is higher than that of the surrounding liquid and the temperature within the bubbles essentially is equal to that of the surrounding liquid. The growth under these circumstances is limited by the rate at which the surrounding liquid can be pushed out of the way. In a thermally dominated system, the pressure inside the vapor space is essentially the same as the surrounding liquid and the temperature within the bubble is considerably different than the temperature far removed from the vapor-liquid interface. Under these conditions, the growth is determined by the rate at which thermal energy can be conducted from the liquid to the interface.

For inertially dominated growth, the bubble radius versus time can be calculated from Rayleigh's equation

$$r = \sqrt{\frac{2}{3} \frac{P_v - P_l}{\rho_l}} t \quad (2)$$

which predicts that the radius varies linearly in time with the driving potential being the vapor pressure within the bubble minus the liquid pressure far removed from the bubble boundary. The thermally dominated regime can be approximated by^{1,2}

$$r = 2 Ja \sqrt{\alpha_l t} \quad (3)$$

for spherical bubble growth. Under these conditions, the radius increases as the square root of time and the driving potential is the temperature difference between the liquid temperature far removed from the bubble and the local saturation value. Figure 1 is a simplified illustration of these growth behaviors in terms of the bubble radius versus time. Since the controlling behavior is the one with the slowest growth, the growth is inertially limited initially with the latter portions being dominated by the rate at which energy can be conducted to the liquid vapor interface.

Given the above arguments, it is seen that if a vapor explosion is to occur, it must take place under the conditions where the inertially dominated growth can exist to a significant radius. The magnitude of this radius will be discussed in the following paragraphs.

The thermal boundary layer, spontaneous nucleation model developed in Ref. 8 provides a prediction of the droplet size of cold liquid which can be captured by the hot liquid and thus initiate an explosion. This droplet size is strongly dependent upon the contact temperature, and small scale

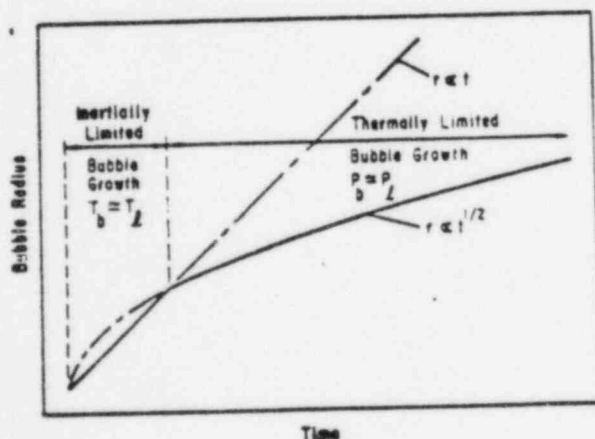


Fig. 1. Inertial and Thermal Dominated Bubble Growth Behaviors.

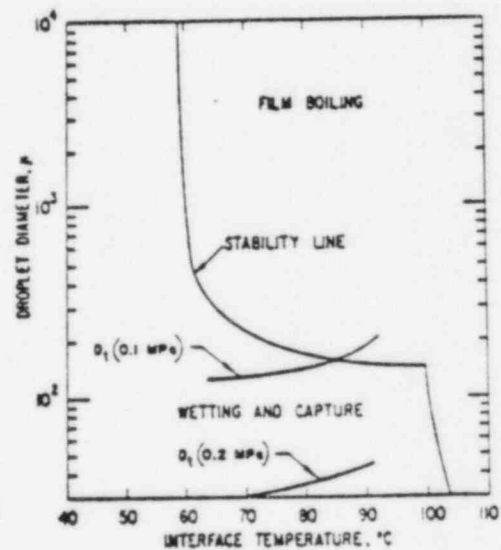


Fig. 2. Freon-22 Capture Diameters Versus Interface Temperature at a Pressure of 0.1 MPa.

experiments^{9,13} have confirmed that this dependency does indeed exist. When the interface temperature upon contact is above the homogeneous nucleation temperature but below the critical temperature, the capture diameter for Freon-22 is order of magnitude 100 μ as shown in Fig. 2. It is interesting to compare this stability curve, which is essentially invariant for pressures from 0.1 MPa to 0.5 MPa with the approximate radius where inertial to thermal growth transition occurs. This is obtained from Eqs. 2 and 3 and is given by

$$r_t = \frac{4 Ja^2 \alpha_l}{\sqrt{\frac{2}{3} \frac{P_v - P_l}{\rho_l}}} \quad (4)$$

The transition diameter for system pressure of 0.1 MPa is shown in Fig. 2 as is the corresponding diameter for a system pressure of 0.2 MPa. The transition dimension is essentially the same as the capture diameter for system pressure of 0.1 MPa, but for a pressure of 0.2 MPa it is an order of magnitude less than the capture size. Consequently, at atmospheric pressure the inertially dominated growth is capable of growing through the entire drop dimension with a vapor pressure higher than the surrounding liquid pressure. As illustrated in

Fig. 3, as a high pressure vapor source inside of a liquid droplet approaches the opposite surface, the droplet will burst open producing a fine liquid spray and releasing the stored high pressure vapor which is the incipient shockwave to start the interaction. However, the most important aspect is the highly fragmented liquid spray which would be produced as the droplet is ruptured. It is this very fine liquid spray, which is much smaller than the parent droplet and therefore certainly less than the capturable size, which can provide the highly fragmented cold liquid material necessary for sustained propagation. Since the dimensions of this liquid spray may be an order of magnitude of 1 to 10 μ , nucleations within these small liquid droplets, as they contact additional hot liquid interfaces, are capable of experiencing inertially dominated growth through their entire dimensions at liquid pressures considerably greater than 0.1 MPa. This process of nucleation, inertial growth, and

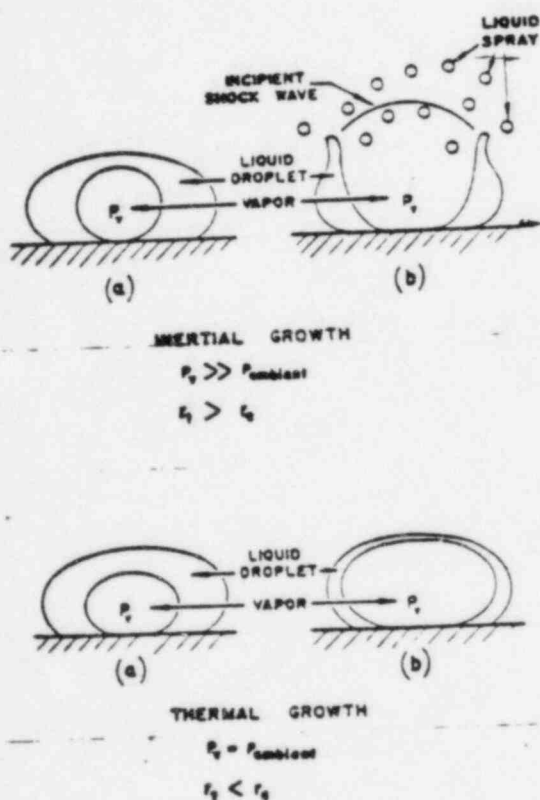


Fig. 3. Liquid Drop Behaviors for Inertially and Thermally Dominated Bubble Growth Behaviors.

liquid fragmentation continues until a pressure is achieved where the inertially dominated bubble growth is terminated at the critical size as discussed in Ref. 9. The other crucial aspect is the time required for this inertial growth, which is given by

$$t_t = \frac{6 \text{ Ja}^2 \alpha_2 \rho_2}{P_v - P_l} \quad (5)$$

The time required for initial growth of Freon-22 at an interface temperature of 60°C and a system pressure of 0.1 MPa is 5 μ sec. Consequently, this mechanism can occur on a time scale which is two orders of magnitude shorter than the overall pressure rise, and thus, this internal fragmentation mechanism can be extremely effective in providing propagation for the explosive event.

The above behavior should be contrasted to a system in which the growth becomes thermally dominated well inside the liquid droplet dimension as illustrated by the lower pair in Fig. 3. For such conditions, the vapor pressure inside the cavity is essentially the same as the surrounding system pressure which means that there is no high pressure source to rupture the surrounding liquid and no release of high pressure vapor to initiate the shockwave. Without the fragmentation and propagation mechanism, any initiating event is essentially incapable of escalating and propagating throughout the remainder of the system. Therefore, the liquid captured bubble growth arguments predict that there should be a significant difference between the behavior observed at a system pressure of 0.1 MPa and 0.22 MPa when Freon-22 is the cold liquid.

HIGH TEMPERATURE SYSTEMS

As shown by the homogeneous nucleation rates for Freon-22 given in Table I, when the interface temperature upon contact approaches the thermodynamic critical point, each molecule effectively becomes a nucleation site. As discussed in Ref. 8 this near-critical region is essentially a singularity in the standard representation of homogeneous nucleation because surface tension does not exist at the thermodynamic critical point. Consequently, the absolute values of these nucleation rates may be physically unrealistic, but they do demonstrate that homogeneous nucleation should be extremely dense at these elevated temperatures. This leads to the conclusion that at near critical and super-critical contact interface temperatures, the high density spontaneous nucleation should provide a rapid and stable vapor layer which terminates energy transfer between the two liquids on a very rapid time scale. The details of the thermal boundary layer development and nucleation rates are given in Ref. 8.

TABLE I.

Homogeneous Nucleation of Freon-22
 $J = \text{ND} \exp \left(- \frac{U}{RT} \right)$

Temp. °C	P_v bar	σ dyn/cm	T °K	Number of Molecules	J $\text{cm}^{-3} \text{sec}^{-1}$	Nucleation Time sec 10^7
50	25.4	4.71	323	28	1.8×10^{12}	2.5×10^{-12}
52	25.1	4.65	325	28	5.2×10^{11}	1×10^{-11}
54	24.7	4.58	327	28	1.5×10^{11}	1×10^{-11}
56	24.3	4.52	329	28	4.2×10^{10}	1×10^{-11}
58	23.9	4.45	331	28	1.2×10^{10}	1×10^{-11}
60	23.5	4.38	333	28	3.5×10^9	1×10^{-11}
62	23.1	4.31	335	28	1×10^9	1×10^{-11}
64	22.7	4.24	337	28	3×10^8	1×10^{-11}
66	22.3	4.17	339	28	8×10^7	1×10^{-11}
68	21.9	4.10	341	28	2×10^7	1×10^{-11}
70	21.5	4.03	343	28	5×10^6	1×10^{-11}
72	21.1	3.96	345	28	1×10^6	1×10^{-11}
74	20.7	3.89	347	28	2×10^5	1×10^{-11}
76	20.3	3.82	349	28	5×10^4	1×10^{-11}
78	19.9	3.75	351	28	1×10^4	1×10^{-11}
80	19.5	3.68	353	28	2×10^3	1×10^{-11}

EXPERIMENTAL APPARATUS

HIGH PRESSURE EXPERIMENTS

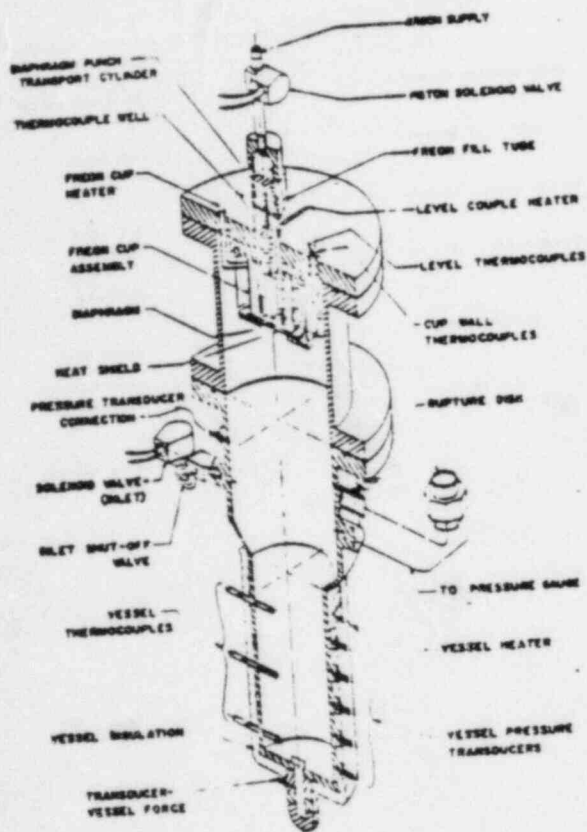


Fig. 4. Elevated Pressure Apparatus.

Separate systems were constructed to investigate Freon-mineral oil and Freon-water explosive interactions under elevated ambient pressures. This was done to simplify the progression of the experiments as well as to prevent system contamination that would result in switching between water and mineral oil as the hot liquids in the same system. Figure 4 illustrates the system configuration for the experiments. The vessels were fabricated from 10 and 15-cm pipes with electrically heated, insulated cups suspended from the upper flange. Thermocouples were provided within this cup to measure the liquid level prior to the test as well as the initial fluid temperature, and a solenoid-punch system was used to rupture the diaphragm at the base of the cup and initiate the test. The lower portion of the vessel in which the second fluid was maintained, was fitted with thermocouples to measure the temperature distribution within the bath and high response, piezoelectric pressure and force transducers to monitor the interaction behavior of the two liquids. This portion of the vessel was also outfitted with trace heaters and insulation for control of the liquid temperature.

In addition, the gas space above the interaction zone was equipped with a pressure transducer to monitor the overall system pressure and a rupture disc and vacuum line were mounted in this same locale. The vacuum line was outfitted with a solenoid valve which was energized simultaneously with the diaphragm-punch transport cylinder to isolate the vacuum system once the test was initiated.

HIGH TEMPERATURE EXPERIMENTS

The near and supercritical interface temperature experiments were conducted in the apparatus shown in Fig. 5. A large inerted volume surrounded this apparatus so that the change in system pressure due

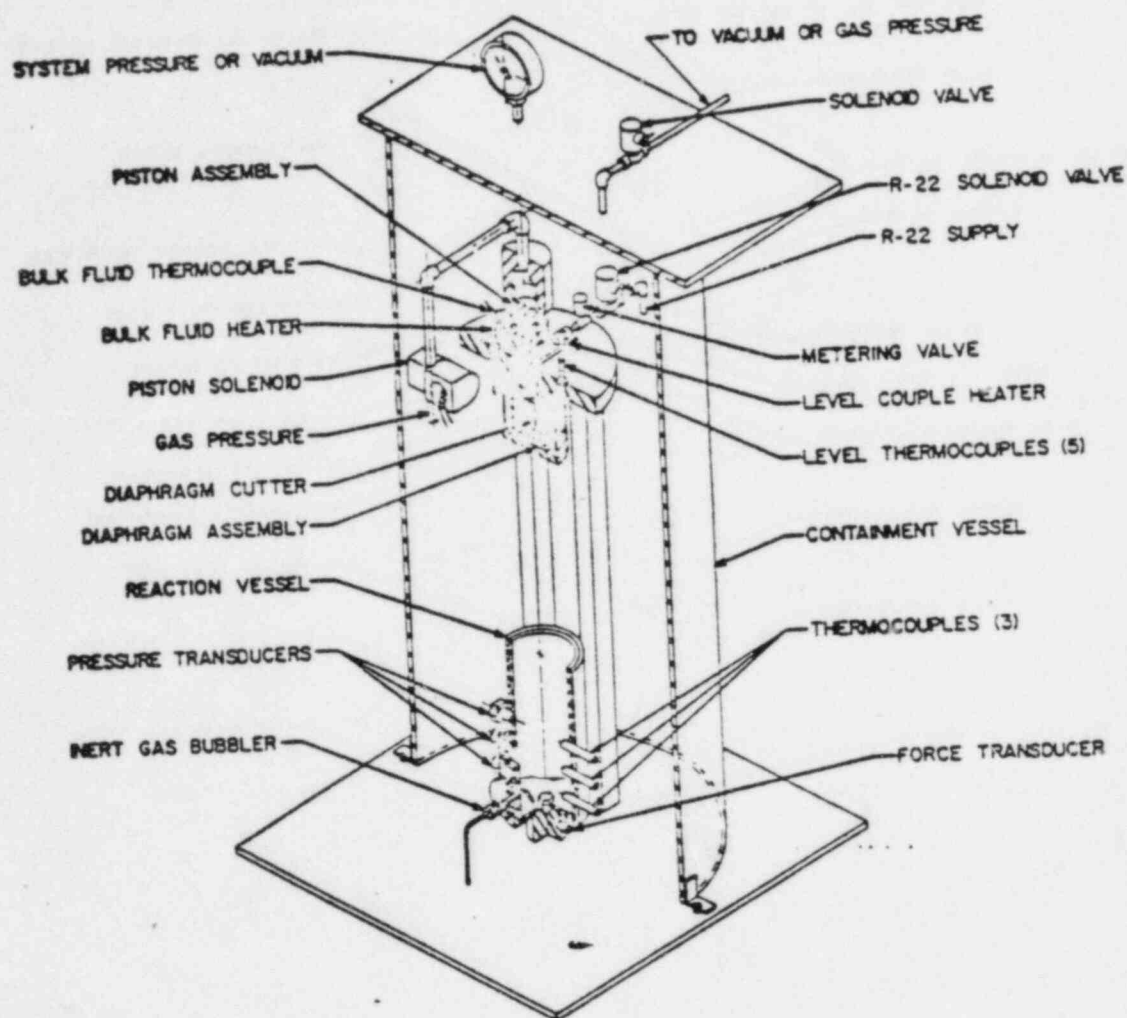


Fig. 5. Large Scale Vapor Explosions in a Contained Volume

to vaporization of the Freon in film boiling did not increase the system pressure to a level sufficient to terminate the explosion itself. This large volume ensured that the equilibrium pressure following complete vaporization of the Freon was less than 0.1 MPa for an initial system pressure of 0.5 MPa. Since prior experiments with this liquid pair^{4, 7, 10} have shown that explosions can be readily reproduced when the system pressure is 0.1 MPa, the slight pressurization of the test chamber is insufficient to preclude explosive interactions. This apparatus is much like the high pressure system described above in that the Freon is dropped into the hot mineral oil bath and the system instrumentation includes the initial temperature of the two liquids, piezoelectric pressure transducers to measure the interaction behavior and a force transducer to establish the experimental consistency of the pressure measurements. An argon gas bubbler is included in the mineral oil to insure that the hot liquid is well stirred.

EXPERIMENTAL RESULTS

HIGH PRESSURE EXPERIMENTS

Elevated pressure experiments were conducted with liquid pairs of Freon-22 and mineral oil and Freon-22 and water, both of which have been shown to be explosive at atmospheric pressures. In order to determine that the system configuration itself did not affect the characteristics of the given liquid pairs, the initial set of experiments in both systems were conducted at an ambient pressure of 0.1 MPa with argon as the cover gas. The results of these first experimental sequences are shown in Figs. 6 and 7 for the Freon-oil and Freon-water systems, respectively. The data are in excellent agreement with the previously published results of Refs. 4 and 8 for both the onset of explosive conditions and the maximum pressures measured. The maximum interaction pressure of 2.5 MPa is very close to the value deduced from bubble growth considerations⁹ where the vapor growth would be thermally dominated beginning at the critical size. These test sequences at a pressure of 0.1 MPa demonstrate that the apparatus did not affect the explosive behavior of the liquid pairs. Following the experimental sequences at atmospheric pressure, similar experiments were conducted at ambient pressures of 0.22 and 0.8 MPa, and the results of these experiments are also shown in Figs. 6 and 7. At both of these elevated system pressures and in both the Freon-mineral oil and Freon-water systems, no explosive interactions were observed throughout the entire temperature range for the hot liquid.

In these elevated pressure experiments, the system pressure was the only difference in the macroscopic details of the system configuration and the test procedures. Therefore, it is difficult to envision why the initiation of an explosive interaction would be so highly pressure dependent. On the other hand, if the propagation in an explosive event is due to the inertially dominated

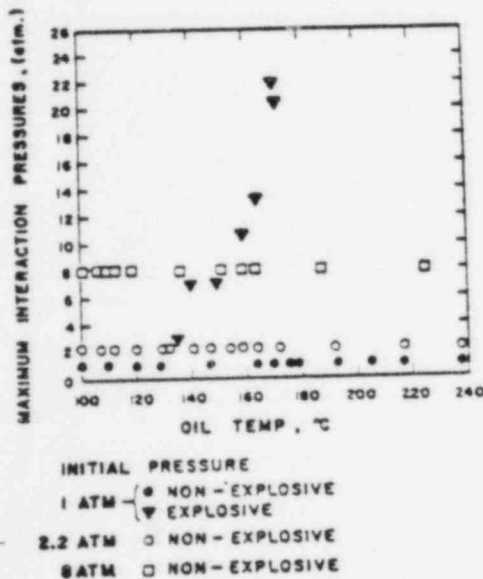


Fig. 6. Interaction Behavior of Freon-22 and Mineral Oil for Various System Pressures.

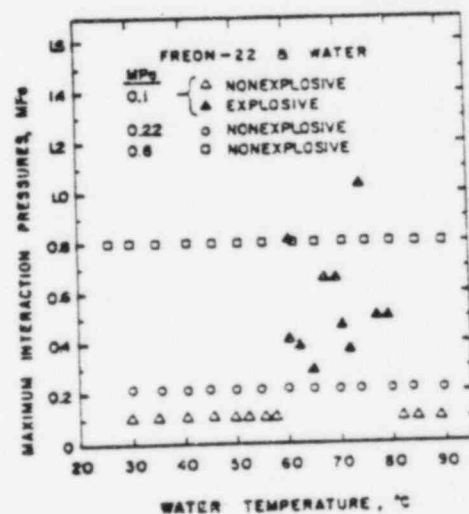


Fig. 7. Interaction Behavior of Freon-22 and Water for Various System Pressures.

bubble growth and the resulting internal fragmentation of the cold liquid phase, then such extreme pressure sensitivity is more apparent. As an example, the transition radius for Freon-22 with an interface temperature of 70°C decreases from 100 μ to 20 μ as the system pressure increases from 0.1 MPa to 0.22 MPa. Consequently, at the higher pressure levels the system must be fragmented to an extremely small size before the propagation can be sustained. This fragmentation size is an order of magnitude smaller than the capturable size during the normal film boiling and prefragmentation process. Thus, at elevated pressures, in these free contacting modes, the system is simply not able to fragment down to a size capable of sustaining an explosive behavior. While the prefragmentation process is definitely dependent upon contacting mode and the magnitude of any external trigger, if the system pressure is sufficiently high so that inertially dominated bubble growth is essentially terminated at the critical size, it is difficult to envision how any physical (vapor) explosion can be initiated or sustained.

When the above results are applied to postulated fuel failures in nominal reactor operating conditions, the 15.5 MPa operating pressure of a PWR dictates that the transition radius between inertially and thermally dominated bubble growth is essentially at the critical size of a vapor cavity. The argument is not quite so clearcut for the 7.0 MPa system pressure of a boiling water reactor, but the transition radius of 0.2 μ indicates that such extensive prefragmentation of the water is highly unlikely. However, this conclusion should be confirmed by water experiments.

HIGH TEMPERATURE EXPERIMENTS

As discussed above, the nucleation considerations indicate that when the interface temperature upon contact between hot and cold liquids approaches the critical temperature of the cold liquid, film boiling should be generated immediately after contact as a result of the tremendous nucleation rate. Since the critical temperature of Freon-22 is 96°C, a near-critical interface temperature could not be obtained with a Freon-water system at a pressure of 0.1 MPa or below. To achieve the near-critical regime upon contact in a Freon-22-mineral oil system, initial mineral oil temperatures of 200°C or higher are required, which means that the test must be performed in an inert environment so that oxidation of the oil does not occur. Such oxidation could significantly change the thermal physical properties of the mineral oil and thus, greatly confuse the calculation of interface temperature. The closed system is required to provide this inert environment and some pressurization of the system will result from normal film boiling of the Freon before the initiation of the explosive interaction. To insure that the system pressure effects discussed above were not important in this test, the initial pressure chosen was 0.05 MPa which resulted in a final equilibrium pressure of less than 0.1 MPa.

The results of these high temperature experiments are shown in Fig. 8 for maximum interaction pressures observed as a function of the initial mineral oil temperature. They clearly show that when the oil temperature reaches a sufficiently high level, all explosive interactions cease. For saturated Freon-22 at 0.05 MPa, which corresponds to an initial temperature of -54°C and an initial mineral oil temperature of 205°C, the interface temperature upon contact evaluated from Eq. 1 is 96°C which is exactly the critical temperature of Freon-22. While this close agreement may be somewhat fortuitous, these experiments certainly demonstrate that vapor explosions are eliminated at very high interface

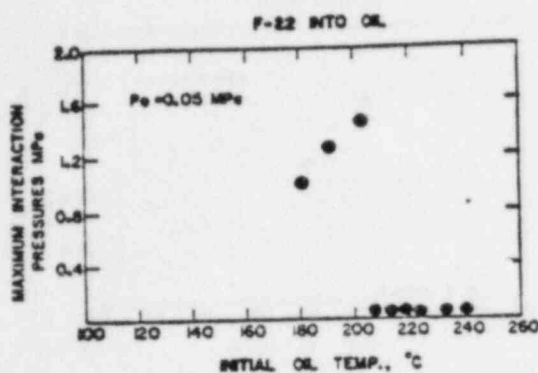


Fig. 8. Interaction Behavior for Freon-22 and Mineral Oil at High Oil Temperatures.

temperatures for nonchemically reacting systems. This is also in agreement with the recent interpretation of the tin-water experiments¹⁴ which are available in the literature.

For the hypothetical core meltdown conditions following a loss-of-coolant accident, one possible fluid pair of concern would be water and molten fuel. The interface temperature upon contact between water and fuel is more than three times the critical temperature of water. Therefore, these experiments would indicate that the explosive potential for such a system is negligible. This is in agreement with the large scale fuel-water experiments reported in Ref. 15.

CONCLUSIONS

These definitive simulant fluid experiments have demonstrated that elevated system pressures can result in the cessation of explosive interactions. This elimination of explosive events is related to the propagation mechanism required for sustaining an explosive event. In addition, high temperature interaction experiments with simulant fluids have demonstrated that for nonchemically reacting systems, a supercritical contact interface temperature also results in the elimination of vapor explosive events.

NOMENCLATURE

C	specific heat
h_{fg}	latent heat of vaporization
Ja	Jakob number - $\frac{(T_w - T_{sat})\rho_l C_l}{\rho_v h_{fg}}$
k	thermal conductivity
P	pressure
r	radius
T	temperature
t	time
α	thermal diffusivity
ρ	density

Subscripts

c	cold
h	hot
i	interface
l	liquid
sat	saturation
t	transition
v	vapor
w	wall

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