

ROLE OF FUEL BUBBLE PHENOMENOLOGY IN ASSESSMENT OF LMFBR SOURCE TERM

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ABSTRACT

Phenomenological aspects of a fuel vapor bubble formed in the sodium pool in a hypothetical severe accident are considered. The potential for fuel bubble collapse in the sodium pool is analyzed. It appears that for a wide range of hypothetical LMFBR accidents involving core vaporization, the fuel vapor bubble would likely be quenched and collapse prior to migration to the cover gas region. Such rapid quenching is due mainly to radiative heat transfer from the fuel bubble, coupled with the inherent capability of the sodium pool (large subcooling and high thermal conductivity) to dissipate thermal energy. Major uncertainty in the analysis concerns fuel vapor condensation phenomena at the sodium interface and its effect on the sodium surface radiation absorptivity. This is discussed in detail.

INTRODUCTION

When assessing the radiological source term for LMFBR site evaluation, a hypothetical accident involving vaporization of part of the core is often postulated. It is generally perceived that the potential for plutonium release in an LMFBR accident would significantly increase with fuel vaporization. However, no quantitative evaluation of the plutonium source term associated with fuel vaporization has yet been made, mainly because of a number of phenomenological uncertainties involved. Among the important uncertainties is the dynamic behavior of a fuel vapor bubble formed in the sodium pool. This paper represents an attempt to delineate some of the important aspects of the fuel bubble phenomena as they relate to the plutonium source term. First the key issues involved will be illustrated, based on a scoping analysis of the behavior of a single fuel vapor bubble formed in

the sodium pool. Then, the dominant mechanism for cooling of the fuel bubble, namely, radiative heat transfer will be discussed. A key parameter in evaluating the radiative heat flux is the absorptivity of the sodium surface, which is a strong function of the degree of contamination of the sodium surface by fuel vapor condensate. A contamination mechanism due to gravitational settling of fuel-condensate fog particles will be described in detail.

OVERALL SOURCE TERM PHENOMENOLOGY

For illustrative purposes, we consider a hypothetical accident involving fuel vaporization in a large LMFBR. The vaporized fuel expands out from the core, forming a single vapor bubble in the sodium pool, as shown in Fig. 1. The use of a single bubble concept, while not accounting for interference of the upper internal structure with the fuel vapor expansion, would be expected to be helpful in estimating the overall bubble phenomenology.

The bubble, while expanding, would tend to migrate to the cover gas region under the force of gravity. As will be shown later, this migration would take 2 ~ 3 seconds for a large LMFBR.

¹ In a typical accident scenario involving fuel vaporization, a high-pressure fuel vapor, probably mixed with liquid fuel droplets as well as other gaseous species such as sodium vapor, steel vapor, and fission product gases, would be ejected from the core into the sodium pool above. This gaseous mixture ejected into the sodium pool will be loosely termed "fuel bubble." In the discussions that follow, it is assumed that the initial content of the bubble is predominantly fuel vapor.

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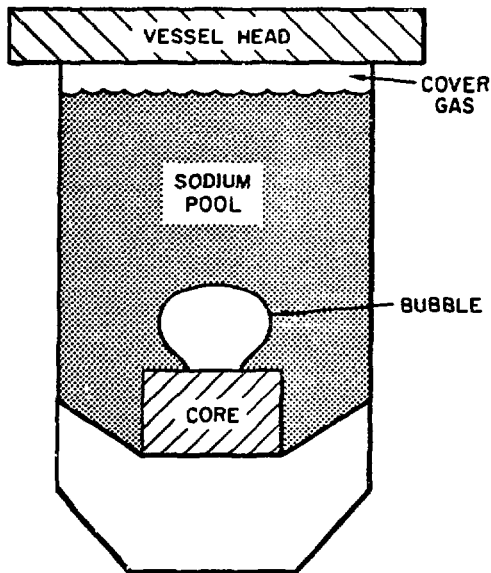


FIG. 1 FORMATION OF A FUEL VAPOR BUBBLE IN SODIUM POOL

Also, it is likely that the bubble would undergo a few oscillations before it reaches the cover gas region. If the bubble collapses due to condensation of the fuel vapor before it migrates to the cover gas region, all the fuel in the bubble would be harmlessly dispersed in the sodium pool.

If the bubble migrates from the core to the cover gas region, it would break through the sodium pool surface and mix with the cover gas. This gaseous mixture would cool here by thermal radiation and natural convection, thereby causing additional rapid condensation of the remaining fuel vapor into fine aerosols. Leaks in the reactor vessel cover (some of which may develop during the initial phase of the bubble expansion) would then provide the path for the gaseous mixture to carry the fuel aerosols into the containment. This aerosol transport into the containment would compete with natural aerosol removal processes occurring in the cover gas region.

There are three mechanisms that would reduce the fuel aerosol source term. First, heat loss from the bubble to the cold surroundings would cause fuel vapor condensation as well as a reduction in the bubble pressure. If the heat loss is sufficiently large during the expansion and migration of the bubble, the bubble would collapse in the sodium pool before it rises to the cover gas region. In this case, the fuel source term would be little different from that for benign core meltdown without fuel vaporization. Second, natural aerosol removal processes (e.g. thermal and kinetic agglomeration, gravitational settling, and plateout) would provide some reduction in fuel aerosol concentration in the cover gas region. Third, hydrodynamic impaction and/or diffusive deposition of fuel

particles in the various leak paths leading to the containment would reduce the leak rate and might possibly "clog" the leak path from further fuel particle release. A scoping analysis of the first mechanism is described below.

SCOPING ANALYSIS FOR BUBBLE RISE/COLLAPSE

To assess the potential for fuel bubble collapse in the sodium pool, a scoping analysis has been conducted considering a single fuel bubble formed in the sodium pool. Initially, the analysis used an assumption which had previously been found consistent with the results of the TREAT S-11 test (1), namely, that the sodium interface temperature was maintained close to the sodium boiling point at bubble pressure without sustained sodium vaporization. This assumption led to a simple expression for the bubble collapse time controlled by heat conduction into the sodium pool, viz.,

$$t_c = \frac{\pi}{4} \left(\frac{\rho_v L_v}{\rho c \Delta T} \right)^2 \frac{R_o^2}{\alpha} \quad (1)$$

where ρ_v is the fuel vapor density, L_v is the fuel heat of vaporization, ρ , c and α are the density, specific heat and thermal diffusivity of the sodium, respectively, R_o is the initial bubble radius, and ΔT is the temperature difference between the interface and the bulk of the sodium pool. The bubble collapse time can then be compared to the time for bubble migration to the cover gas region. If the bubble rise velocity is approximated by that of a "Taylor" bubble based on the initial radius, the bubble migration time is given by

$$t_m = \frac{H}{\sqrt{gR_o}} \quad (2)$$

where H is the sodium pool height and g is the gravitational constant. If the migration time is larger than the collapse time, the bubble would be quenched and collapse in the sodium pool before it migrates to the cover gas region. The plutonium source term would then be significantly reduced. This would happen if

$$R_o^{5/2} < \frac{H}{\sqrt{g}} \left(\frac{4}{\pi} \right) \left(\frac{\rho c \Delta T}{\rho_v L_v} \right)^2 \alpha \quad (3)$$

The fuel vapor density ρ_v depends on the energetics of core vaporization and is a function of the fuel vapor temperature (or pressure). Equation (3) is illustrated in Fig. 2 for $H = 10$ m. It is seen that a fuel vapor bubble initially at 10 atm (the corresponding temperature being 4200 K) would collapse in the sodium pool prior to migration to the cover gas region, if the initial bubble diameter is 1 m or less.

These calculations have two major uncertainties. The first concerns the application of planar (one dimensional) heat conduction transients to spherical conduction. Although this approximation becomes increasingly inappropriate during the second half of the bubble collapse

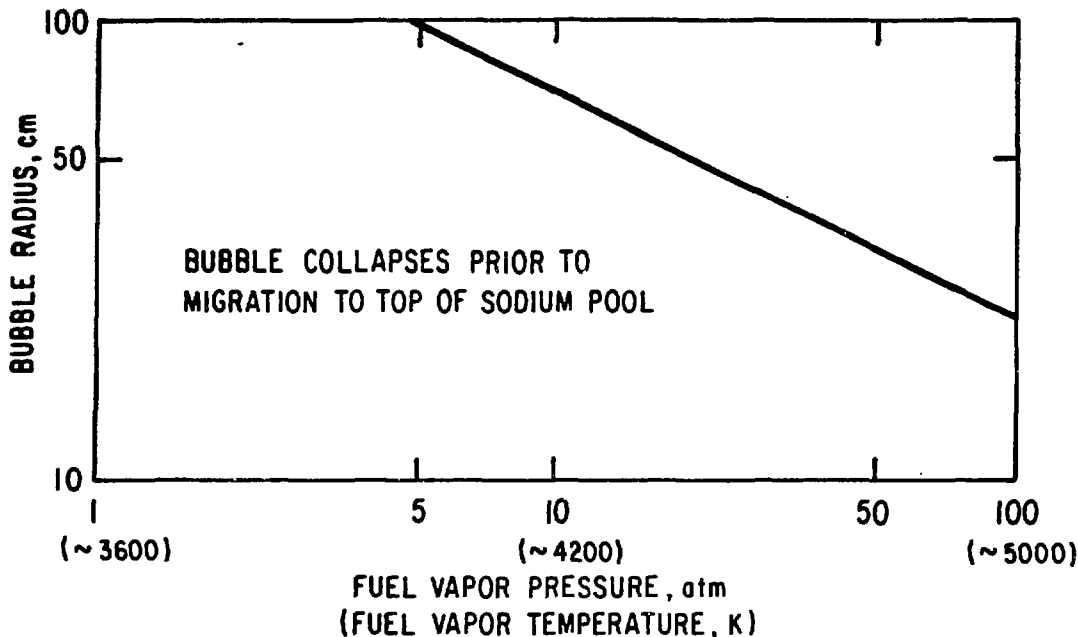


FIG. 2 FUEL VAPOR BUBBLE COLLAPSE BEHAVIOR CONTROLLED BY SODIUM-SIDE HEAT CONDUCTION (the interface temperature was assumed to be equal to the sodium boiling point at bubble pressure)

period, its use in Eq. (1) is considered to be adequate for purposes of the scoping analysis.

The second uncertainty is the above cited assumption — that heat flux from bubble to sodium is sufficient to maintain the interface temperature close to the sodium boiling point, but not so large as to cause significant sodium vaporization. The mechanism for this vapor side heat flux is not well understood but it is likely to involve radiation flux of the heat generated by bulk vapor phase condensation of fuel into small particles — fog or sleet — in the vicinity of the interface. The uncertainty is the resulting effect on the radiant heat absorption of the sodium interface. This absorptivity is of dominant significance. If the condensed fuel fog does not significantly contaminate the surface, then current estimates ($\sim .06$) of clean sodium absorptivity (2) are applicable. Alternatively, if the fuel fog does contaminate the surface, the absorptivity would rise much closer to unity, as will be discussed later. In this case the radiative heat flux would be an order of magnitude higher and the time for the surface to reach sodium boiling point would be decreased by a factor $(.06)^2$, or two orders of magnitude smaller.

In the former case, the potential for sodium vaporization is negligible and the bubble collapse time is best estimated via radiation flux calculations rather than sodium side heat conduction. In the latter case, the radiative heat transfer would raise the sodium surface temperature to saturation in tens of milliseconds and momentarily promote intense sodium

vaporization. If this vaporization blows off the fuel contaminant sufficiently quickly, the sodium surface would become clean again, reducing the heat flux absorbed into the sodium by an order of magnitude. The sodium vaporization would then cease and the surface temperature would tend to drop below saturation. As a consequence, the fuel vapor would rush in toward the sodium surface and the process of fuel vapor condensation and surface contamination would repeat itself. This mechanism suggests that the sodium surface temperature would fluctuate somewhat, but remain close to saturation. It supports the approximations of the above calculations if the time scale for contamination is small enough to prevent much lowering of surface temperature, and also if the time scale for decontamination of surface is small enough to disallow much sodium vaporization.

If, however, the time scale for contamination is longer, then the decontamination time need not play a role, and clean surface absorptivity may be used. At this absorptivity level ($\sim .06$) the principal heat transfer mechanism is still radiation.

Parametric calculations assuming a constant heat flux due to thermal radiation were performed for two time scales; the time for fuel condense-out (and hence bubble collapse in the absence of significant inert or sodium vapor) and the time for bubble surface to reach sodium boiling temperature and initiate sodium vaporization into the bubble. The collapse time is given, in the absence of inerts or sodium vapor, by

$$t_c = \frac{\rho L R}{v v_o} \quad (4)$$

and the time to sodium boiling is estimated by

$$t_s = \frac{\pi}{\alpha} \left[\frac{k(T_s - T_o)}{2q} \right]^2 \quad (5)$$

where q is the radiative heat flux and T_s and T_o are the sodium boiling point at bubble pressure and the sodium pool bulk temperature, respectively. Figure 3 shows the results of illustrative calculations for a fuel vapor bubble at 3600 K (UO_2 boiling point at one atmosphere) and sodium pool subcooling ($T_s - T_o$) = 400 K. Well into Region I the fuel bubble collapses without sodium vaporization whereas well into Region II, sodium vaporizes into the bubble prior to fuel vapor condense-out. Uncertainty occurs only near the boundary between the two regions given by the equation

$$R_o = \frac{\pi}{4} \frac{[k(T_s - T_o)]^2}{\alpha \rho L v q} \quad (6)$$

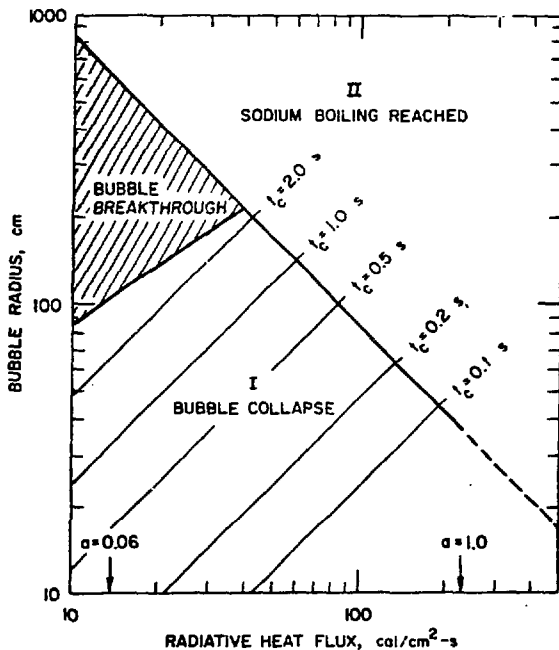


FIG. 3 FUEL VAPOR BUBBLE COLLAPSE BEHAVIOR DUE TO CONSTANT RADIATIVE HEAT FLUX (pressure = 1 atm, bubble temperature = 3600 K, bulk sodium subcooling = 400 K; in Region I, bubble collapses without sodium vaporization whereas in Region II, sodium vaporizes into the bubble prior to fuel vapor condense-out)

The shaded area of Region I depicts conditions where the bubble is able to reach the cover gas region prior to complete collapse. This area is based on Eq. (2) with $H = 10$ m, yielding a lower boundary defined by

$$R = (Hq/\rho_v L \sqrt{g})^{2/3} \quad (7)$$

Figure 3 shows, for example, that for pure fuel vapor and bubble radii less than .86 m, a bubble cannot reach the cover gas region if the sodium interface is clean or is partially contaminated up to an absorptivity $\alpha = 0.43$; and that such a fuel vapor bubble of radius up to 0.36 m does not reach the cover gas even with a fully contaminated ($\alpha = 1.0$) interface. It is also seen that for the wide range of conditions depicted as Region I, the radiative heat flux from condensing fuel vapor is removable by sodium long before its surface temperature can approach the boiling point. The radiative heat flux depends on the sodium surface absorptivity as well as on the emissivity of the fuel bubble, as is further discussed in the next section. Assuming the bubble emissivity is unity, the radiative heat fluxes translate into the values of the sodium surface absorptivity indicated on the abscissa of Fig. 3. Clearly, the propensity for fuel bubble collapse is dependent upon large subcooling of the sodium pool coupled with the high thermal conductivity of sodium. In addition, the low values of thermal radiation absorptivity of a clean sodium surface minimize the potential for sodium vaporization. This is especially true for high pressure fuel bubbles at vapor temperatures much above the normal boiling point of 3600 K.

RADIATIVE HEAT TRANSFER

In this section we provide a simplified formulation of the radiative heat flux to the boundary of a high temperature HCDA bubble submerged in liquid sodium. The bubble contains fuel vapor and possibly liquid fuel droplets with a noncondensable gas as minor constituents. The liquid sodium surface may be contaminated by a fuel vapor-condensate layer causing an effective reflectivity smaller than that of pure sodium.

Assuming that the sodium surface is uniform and its radiation properties are constant, then the net radiative flux to the surface can be written as

$$q = \epsilon_{eq} E_{bg} - \alpha_{eq} B_e \quad (8)$$

where $E_{bg} = \sigma T_g^4$ is the emissive power of a black body at the mixture temperature T_g and B_e is the surface radiosity. ϵ_{eq} and α_{eq} are the equivalent emissivity and absorptivity of the two-phase mixture of the bubble, respectively. Assuming the radiation properties of the bubble mixture are constant and gray, then

$$\alpha_{eq} = \epsilon_{eq} \quad (9)$$

Also, the surface radiosity can be expressed as

$$B_e = \epsilon_e E_{be} + \rho_e H_e \quad (10)$$

where ϵ_e and ρ_e are the effective emissivity and reflectivity of the sodium surface and E_{be} is the black-body emissive power at the surface temperature. H_e is the incident radiation on the surface given by

$$H_e = B_e \tau_{eq} + E_{bg} \epsilon_{eq} \quad (11)$$

with $\tau_{eq} = 1 - \alpha_{eq}$ the equivalent transmittance. Combining all equations above and after some algebraic manipulations, we obtain

$$q = \frac{E_{bg} - E_{be}}{\frac{1 - \alpha_e}{\alpha_e} + \frac{1}{\epsilon_{eq}}} \quad (12)$$

where the gray surface assumption that $\epsilon_e = \alpha_e = 1 - \rho_e$ has been made. This equation is an explicit expression for the radiative heat flux to the surface as a function of the surface absorptivity and the equivalent emissivity of the bubble mixture.

The equivalent emissivity of the bubble depends on the constituents of the bubble. For a large bubble containing a significant amount of UO_2 droplets and UO_2 vapor, the equivalent emissivity of the bubble is close to unity, i.e.

$$\epsilon_{eq} = 1$$

On the other hand, when the bubble contains just the UO_2 vapor without droplets, the emissivity will be less than unity and can be estimated from the mean beam length theory,

$$\epsilon_{eq} = \alpha_{eq} = 1 - e^{-KL}$$

K is the extinction coefficient (here reduced to the absorption coefficient) and L_e is the mean beam length given by

$$L_e = 3.5 (\text{volume/surface area}) = 0.583 D$$

in which D is the diameter of the bubble. Using the pressure base absorption coefficient $K_\alpha = K/P$ (P = pressure), the emissivity becomes

$$\epsilon_{eq} = 1 - e^{-0.583 K_\alpha PD}$$

In the absence of any measurements on UO_2 vapor spectra, we approximate the absorption coefficient of UO_2 vapor by that of CO_2 . The latter can be extrapolated from the CO_2 emissivity chart of Hottel (3). For example, $\epsilon = 0.026$ at $4444^\circ K$ and $P L = 5$ atm-ft in which P_c is the pressure of CO_2 and ϵ and L are equivalent to the present ϵ_{eq} and L_e respectively. Since $K_\alpha = -\ln(1-\epsilon)/PL$, we find $K_\alpha = 0.00526 \text{ ft}^{-1} \text{ atm}^{-1}$. If

$P = 19$ atm and $D = 10$ ft, then $\epsilon_{eq} = 0.442$ for the pure vapor case.

To determine the effective absorptivity of the sodium surface contaminated by a deposit layer of fuel vapor condensate, reference should be made to a recent study (4) where an analytical expression for the effective reflectivity ($\rho_e = 1 - \alpha_e$ under gray assumption) of the contaminated surface has been obtained. There the deposit layer is considered as an absorbing, emitting and scattering layer on an opaque liquid substrate. Numerical results are presented in Fig. 4, in which the effective reflectivity of such a sodium surface is plotted as a function of the optical thickness of the condensate layer and the albedo. (The optical thickness is a product of the extinction coefficient of the condensate deposit and the physical thickness of the deposit, while the albedo is a ratio of the scattering and extinction coefficients of the deposit.) It is seen that for most values of the scattering albedo, the reflectivity is reduced by an order of magnitude as the optical thickness increases from zero to unity.

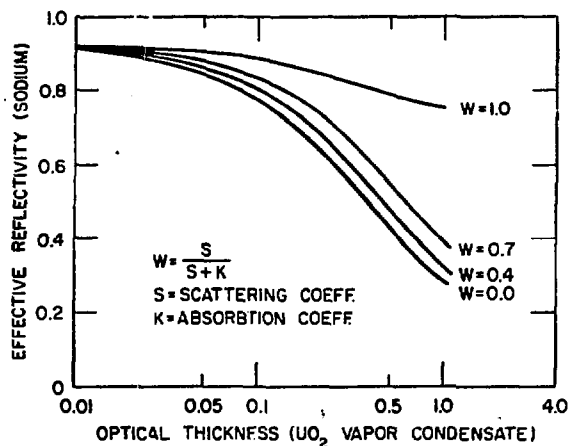


FIG. 4 EFFECTIVE REFLECTIVITY OF A SODIUM SURFACE CONTAMINATED BY UO_2 VAPOR CONDENSATE

CONTAMINATION OF SODIUM SURFACE BY FUEL

As discussed above, a key parameter in assessing fuel vapor condensate-out potential during bubble rise through liquid sodium pool is the absorptivity of the sodium surface. Available experimental data indicates that the absorptivity of a clean sodium surface is about 0.06. The absorptivity, however, could approach unity if the sodium surface is contaminated by fuel vapor condensate. Since these larger values of absorptivity can potentially affect the source term by altering the ability of the sodium pool to digest all fuel vapor as condensate-out, it is imperative to understand the rate at which the surface contaminates in relation to rise/collapse times.

As a first step, an analysis of transient condensation of UO_2 vapor on a sodium surface was conducted. It was assumed that the UO_2 vapor diffuses through a layer of noncondensable gases and condenses on the sodium surface without forming fog. It was found that in typical hypothetical accident situations, the condensate layer would grow to about 10 μ m in one second after the contact between the fuel vapor and sodium surface. This analysis also indicates that the temperature drop in the gas/fuel vapor layer would be as large as 3000 K. Under these conditions, UO_2 fog formation will inevitably occur and quickly eliminate fuel vapor from the immediate vicinity of the sodium surface. This vapor depletion plus radiative cooling of the fuel vapor, not accounted for in the diffusion analysis, suggests that fog formation is the major mode of fuel vapor condensation. The contamination of the sodium surface via direct surface condensation of fuel vapor would be several orders of magnitude less than indicated by vapor diffusion analysis; it is most likely due to deposition of fuel fog particles (aerosols).

There are several well-known aerosol deposition mechanisms that would promote the sodium surface contamination, including gravitational settling, diffusional deposition, thermophoresis, and diffusiophoresis. In addition, the dynamic behavior of the fuel bubble could contribute to the surface contamination, e.g. interface motion during collapse. For a complete assessment of the contamination, these aerosol deposition mechanisms must be evaluated with inclusion of the process of simultaneous aerosol generation due to fuel vapor condensation and radiative cooling as the dominant heat transfer mechanism. Thermophoresis and diffusiophoresis, i.e. thermal or Brownian diffusion, are expected to contribute negligibly in the absence of electrostatic effects (electrophoresis). A detailed theoretical study of gravitational sedimentation of radiation generated fuel fog-aerosol has been made elsewhere (4). Results of calculations based on this formulation are summarized below.

SEDIMENTATION OF FOG CONDENSATE

The formulations of Ref. (4) provide detailed methodology, and prescription for calculations, for determining the growth on time of a fuel deposit layer on an initially clean horizontal sodium surface. The deposit layer is formed from bulk gas phase fog condensation controlled by radiation heat transfer to the sodium surface. The rate of layer growth is determined by a kinematic wave analysis of sedimentation used in conjunction with the rate of fog formation in an initially fog-free vapor space. As the layer grows in thickness its effective surface absorptivity grows from .06 to values approaching unity, causing substantial changes in the bulk rate of fog formation. The result is a feedback mechanism wherein the deposit layer and associated surface absorptivity grows at increasing rates until the absorptivity reaches near unity. It is necessary that this feedback be accounted for in determining the

time for transition from low to high surface absorptivity. Because of the importance of the surface absorptivity values to bubble collapse times, calculations of this transition time are presented here for comparison.

The primary uncertainty in specific calculations is the fog droplet radius R , which is treated parametrically over an entire range of reasonable values from .001 to .1 mm. The transition or deposit layer formation time, t_s , is defined as the time for growth of the deposit layer to value $Z_s = 10 \delta = .03$ mm where effective surface absorptivity reaches $\sim .75$. Table 1 shows results of calculations for two cases. Column 2 shows t_s values for a gas phase of pure fuel vapor at temperature $T_\infty = 3600$ K. Column 3 shows corresponding calculations using $T_\infty = 2000$ K, as well as gas phase density and viscosities close to those of air at this temperature. Both cases use a sodium interface temperature of 1000 K, though results are insensitive to this choice. In these calculations the upper extreme droplet sizes correspond to the beginning of breakdown of assumed Stokes Law settling as well as where transient acceleration of droplet velocities becomes significant. At the lower extreme, Brownian motion, i.e. thermal velocity dispersions, become significant and prevent settling.

TABLE 1 FUEL SEDIMENT LAYER - ABSORPTIVITY GROWTH TIMES IN SECONDS

R (mm)	$T_\infty = 3600$ K	$T_\infty = 2000$ K
.1	1.44	2.65
.05	2.94	5.38
.02	7.81	14.2
.01	17.5	31.2
.005	45.3	77.1
.002	215	345

A comparison of the time scales in Table 1 with bubble collapse times on Fig. 3 shows that the sediment-absorptivity growth times are much longer than bubble collapse times unless fuel fog droplet diameters are 0.2 mm or larger. This large droplet size is deemed highly unlikely.

CONCLUDING REMARKS

It appears that the radiological source term associated with hypothetical fuel vaporization in LMFBRs could be significantly reduced by rapid quenching and condense-out of the vaporized fuel in the sodium pool. Such rapid quenching is due mainly to radiative heat transfer from fuel vapor, coupled with the inherent capability of the sodium pool (large subcooling and high thermal conductivity) to dissipate thermal energy. The low values of thermal radiation absorptivity of a clean sodium surface

would minimize the potential for sodium vaporization. The conclusions drawn here seem to be borne out by the recent results of the FAST experiments at ORNL (5,6). In these experiments, fuel vapor bubbles, produced by a capacitor discharge technique, were found to collapse rapidly in sodium pools of varying depths. A key parameter in assessing the potential for fuel vapor condense-out is the thermal radiation absorptivity of the sodium surface. It could range from 0.06 for a clean surface to unity for a surface fully contaminated by fuel vapor condensate. For realistic assessment of the source term, therefore, it is important to understand fuel vapor condensation phenomena at the sodium surface including mechanisms of fuel fog/aerosol deposition on the surface.

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