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# Melt Progression in Severely Damaged Reactor Cores

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Sandia National Laboratories  
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## ABSTRACT

A model of melt formation and relocation in a two-dimensional core rubble bed is developed in this report. The analysis includes mass conservation equations for the species of interest ( $\text{UO}_2$  and  $\text{ZrO}_2$ ); a liquid phase momentum equation ( $z,r$ ) that incorporates the effects of drag, gravity and capillary forces; and an energy equation that includes internal heat generation by decay heating, convection by the liquid and the solid (as it collapses) as well as conduction and radiation through the bed. An equilibrium  $\text{UO}_2$ - $\text{ZrO}_2$  phase diagram is prescribed and radiative heat transfer through the bed is incorporated utilizing a temperature-dependent thermal conductivity.

Models developed in this work will be implemented in the MELPROG computer code that is being developed by Sandia and Los Alamos National Laboratories. The modified version of MELPROG will then be used to calculate melt progression, crust growth, pool formation, crust failure and the relocation of debris material into the lower plenum during the Three Mile Island accident and other nuclear reactor accidents.

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## NOMENCLATURE

$c_p$	specific heat at constant pressure [J/kg•K]
$d^p$	diameter of the bed [m]
$d_p$	particle diameter [mm]
$D^p$	mass diffusivity [ $m^2/s$ ]
$g$	gravitational acceleration [ $m/s^2$ ]
$h$	enthalpy [J/kg]
$h_f$	heat of fusion [J/kg]
$J$	Leverett function
$k$	thermal conductivity [W/m•K]
$L$	bed height [m]
$P_c$	capillary pressure [ $N/m^2$ ]
$P_g$	gas pressure [ $N/m^2$ ]
$P_l$	liquid pressure [ $N/m^2$ ]
$Q$	decay heat [W/kg of $UO_2$ ]
$r$	radial distance [m]
$S$	saturation
$S_e$	effective saturation, $S_e = (S - S_r) / (1 - S_r)$
$S_r$	residual saturation
$t$	time [s]
$T$	temperature [K]
$u$	liquid volumetric flux in the z direction [m/s]
$U$	velocity at which the solid collapses [m/s]
$v$	liquid volumetric flux in the r direction [m/s]
$Y_{ij}$	volume fraction of species j in phase i
$z$	distance from bottom [m]

### Greek

$\alpha$	volume fraction
$\alpha_t$	thermal diffusivity [ $m^2/s$ ]
$\gamma$	surface tension [N/m]
$\epsilon$	emissivity
$\kappa$	permeability [ $m^2$ ]
$\mu$	viscosity [Pa•s]
$\rho$	theoretical density [ $kg/m^3$ ]
$\rho$	density [ $kg/m^3$ of total volume]

### Subscripts

$g$	gas
$l$	liquid
$s$	solid

## 1. INTRODUCTION

During severe Light Water Reactor (LWR) accidents involving the loss of coolant, the reactor core can sustain considerable damage. Water present in the reactor vessel at the onset of the accident can be boiled away as energy is released by fission product decay [1]. As temperatures in the reactor core increase, steam from the boiling process oxidizes the zircaloy cladding on the fuel rods, releasing additional energy and forming a  $ZrO_2$  layer on the outside of the cladding [2]. Materials with relatively low melting points, such as silver, liquefy and flow downward as core temperatures increase. When the temperature reaches 2200 K, zirconium melts and starts dissolving  $UO_2$  and  $ZrO_2$  (both of which have melting temperatures above 2800 K) [3]. This U-Zr-O liquid solution flows downward if there is a gap between the cladding and the fuel or if holes form in the  $ZrO_2$  crusts overlying the unoxidized cladding [2]. Zircaloy oxidation ceases if all of the Zr is consumed or if a blockage forms as molten materials flow into lower, colder portions of the core and freeze. During this time, the fuel rods can fragment, converting the core into a large rubble bed [4-7]. If the rods are quenched and subsequently shatter, the particles will be small, with diameters on the order of 1 mm or smaller. A rubble bed can also form in the absence of such an event if the damaged fuel rods collapse; in this case the particles will be approximately the same size as a fuel pellet ( $\approx 1$  cm in diameter).

Postaccident analysis of the TMI-2 reactor revealed that the severely damaged core was primarily composed of  $UO_2$  and  $ZrO_2$  particles [4-6]. Of interest to this study is melt progression in such debris beds. Although the energy released by fission product decay falls to less than one-percent of normal operating power one hour after reactor shutdown, temperatures in a dry debris bed can surpass the  $UO_2$  melting point (3100 K) due to the low  $UO_2$  and  $ZrO_2$  thermal conductivities [8,9]. Because dry debris represents a significant risk, the dryout [9-27], quenching and reflooding [28-32] of internally heated particle beds has received considerable attention in the literature. Squarer et al. [10] and Lipinski [11] have presented reviews of recent debris coolability research. Lipinski [12] elucidated much of the physics involved in dryout and developed a steady, one-dimensional model that successfully predicted dryout limits.

Time-dependent models were subsequently developed by E. Gorham-Bergeron [13] and Turland/Moore [14,15]. In-pile D series [16-22] and Degraded Core Coolability [23-24] experiments at Sandia National Laboratories investigated debris dryout in Liquid Metal Fast Breeder Reactors and Light Water Reactors, respectively. Related out-of-pile experiments were conducted by Dhir and Catton [25] and Squarer and Peoples [26].

Relatively little attention has been given to the postdryout meltdown of debris beds that form during LWR accidents. Several Molten Pool [33-37] and Dry Capsule (DC) [38-39] experiments conducted at Sandia National Laboratories investigated incipient debris melting within the context of Liquid Metal Fast Breeder Reactor (LMFBR) accidents. A series of complementary Melt Progression (MP) experiments are being planned at Sandia to study the transition of a dry LWR debris bed to a final molten pool state. The importance of U-Zr-O chemistry will be examined in the MP experiments whereas the earlier LMFBR work stressed  $UO_2$ -stainless steel interactions. Larger particles will be used in the MP study (average diameters near 2 mm versus 0.2 mm in the DC experiments) and the temperature gradients will be smaller. Due to large thermal gradients in the DC experiments, evaporation and recondensation was a dominant crust formation process [39].

Natural convection effects in the final molten pool state have been investigated in many LMFBR studies [40-47]. Emara and Kulacki [40], Suo-Anttila and Catton [41], Cheung [42] and Tveitereid [43] modeled natural convection flows in horizontal fluid layers with internal energy generation. Kulacki and Goldstein [44], Baker et al. [45] and Faw et al. [46] present experimental correlations for upward and downward heat partitioning in such fluid layers.

In the present study a two-dimensional model is presented for the postdryout meltdown of a  $UO_2$ - $ZrO_2$  bed to a final state consisting of a molten pool supported by dense solid crusted regions. This model is an extension of earlier work by Kelly [48], who developed a simplified one-dimensional debris meltdown model for use in the MELPROG computer code that is being developed at Sandia and Los Alamos National Laboratories [48,49]. Uncertainties in the MELPROG debris model can affect the manner in which molten material is released into the lower plenum

and fission product release in degraded reactor cores. Conclusions in a recent review [49] of the current MELPROG model follow: a momentum equation should be included in the analysis; the energy balance should be modified so that more realistic phase diagrams can be employed; and a multidimensional model is needed to analyze accidents, such as Three Mile Island, during which radial crust failure is possible. It is anticipated that models developed in the present study will be implemented into MELPROG in the near future.

The equations governing melt relocation are similar to the undersaturated flow equations discussed in Refs. [51-55]. The primary differences are that in the present study source terms are needed in the mass conservation equations to account for phase changes and an energy equation (with an associated phase diagram) must be solved to determine the temperature increase of the bed and the rate of melting and freezing. The model consists of mass conservation equations for the species of interest ( $UO_2$  and  $ZrO_2$ ); a liquid phase momentum equation that incorporates the effects of drag, gravity and capillary forces; an energy equation that includes internal heat generation by fission product decay, convection by the melt, as well as conduction and radiation through the bed; and an equilibrium  $UO_2$ - $ZrO_2$  phase diagram. Natural convection effects in the molten pool will eventually be included in this model by using the work of Kulacki [44].

A discussion of the model follows. Solutions are presented in Chapter 3 for two particle diameters, 1  $\mu$ m and 5 mm. Important uncertainties in the model are discussed in Chapter 4. In-pile Melt Progression and out-of-pile MELPROG Validation experiments designed to evaluate the present work and to help guide future work are described.

## 2. ANALYSIS

Typical characteristics of a  $UO_2$ - $ZrO_2$  core rubble bed similar to the one found in TMI [4] are given in Table I. One hour after reactor shutdown, power generation falls to approximately one percent of peak power [8]. A TMI-2 type plant with an operating power of  $2.8 \times 10^8$  MW and 93,000 kg of  $UO_2$  in the core [5] has a decay heat  $Q$  on the order of 300 W/kg of  $UO_2$ . Because it takes several days for the decay heat to decrease significantly below the one hour value [8], it is assumed that  $Q$  is constant in the following analysis. A lower limit for the  $UO_2$  volume fraction  $Y_{s1}$  is estimated by assuming that all of the roughly 23,000 kg of Zr in a typical core [5] are oxidized, resulting in the formation of 30,000 kg of  $ZrO_2$ . Neglecting materials other than  $UO_2$  and  $ZrO_2$  in the core gives a maximum  $ZrO_2$  mass fraction of 0.25. Setting the solid densities of  $UO_2$  and  $ZrO_2$  equal to 10000 and 5700 kg/m<sup>3</sup> [5], respectively, gives a maximum  $ZrO_2$  volume fraction  $Y_{s2}$  of 0.36.

TABLE I

Typical initial properties of a core rubble bed.

Quantity of Interest	Typical Value
decay heat, $Q$ [W/kg of $UO_2$ ]	300.0
height of bed, $L$ [m]	1.0 <sup>a</sup>
diameter of bed, $d$ [m]	1.0 <sup>a</sup>
particle diameter, $d_p$ [mm]	0.1-10.0 <sup>b</sup>
initial solid volume fraction, $a_{s1}$	0.6 <sup>c</sup>
$UO_2$ solid volume fraction, $Y_{s1}$	$\geq 0.64$
$ZrO_2$ solid volume fraction, $Y_{s2}$	$\leq 0.36$

a. Order of magnitude estimate from Ref. [4].  
b. From Ref. [6].  
c. Porosity used in bed dryout studies [9].

## 2.1 Mathematical Formulation

The distance characteristic of changes in the pore structure of the debris bed,  $d_p$ , is defined as six times the volume of the solid phase divided by its surface area. The equations governing heat transfer and fluid flow within the bed are amenable to analysis for a particular range of  $d_p$ . When  $d_p$  is on the order of the mean free path of the gas molecules,  $\lambda$ , corresponding to the Knudsen limit, the diffusion models employed in this study must be modified. On the other hand, when  $d_p$  is large, of the same order as  $x_c$ , the distance characteristic of changes in temperature and species concentrations, the particles must be considered individually. The following analysis is restricted to debris beds in which  $d_p$  satisfies  $\lambda \ll d_p \ll x_c$ .

### 2.1.1 Conservation of Mass

Species diffusion is neglected in the following analysis. Typical mass diffusivities for liquids near their melting points are on the order of  $10^{-9}$  m<sup>2</sup>/s [56]. For time scales on the order of  $10^3$  s, the distance characteristic of diffusion,  $[Dt]^{1/2}$ , is approximately  $10^{-8}$  m. That is, liquid phase diffusion is only important over length scales comparable to the average particle diameter. Balancing the mass stored in a differential control volume, convection by the liquid and the solid (as it collapses) and production (or depletion) by melting gives for species  $j$  ( $j=1$  and  $j=2$  correspond to  $UO_2$  and  $ZrO_2$ , respectively)

$$\begin{aligned} \frac{\partial}{\partial t} [Y_{1j}\rho_{1j}a_1] &= - \frac{\partial}{\partial z} [Y_{1j}\rho_{1j}u] - \frac{1}{r} \frac{\partial}{\partial r} [rY_{1j}\rho_{1j}v] \\ &\quad - \frac{\partial}{\partial z} [Y_{sj}\rho_{sj}a_s U] - \frac{\partial}{\partial t} [Y_{sj}\rho_{sj}a_s] \end{aligned} \quad (1)$$

where the subscripts  $l$  and  $s$  refer to liquid and solid, respectively. The quantities  $a_s$  and  $a_l$  are solid and liquid volume fractions, respectively,  $\underline{u}=(u,v)$  is the liquid volumetric flux,  $U$  is the velocity at which the solid collapses downward,  $\rho_{ij}$  is the theoretical density of species  $j$  in phase  $i$  and  $Y_{ij}$  is the portion of phase  $i$  that is occupied by species  $j$ . The  $Y_{ij}$ 's are related to each other by  $Y_{11}+Y_{12}=1$  and  $Y_{s1}+Y_{s2}=1$ .

### 2.1.2 Liquid Phase Velocities

Assuming that the liquids are miscible, Darcy's Law gives for the momentum equations,

$$\rho_1 \frac{\partial u}{\partial t} + \frac{\mu}{\kappa_1} u = - \frac{\partial}{\partial z} P_1 - g\rho_1 \quad , \quad (2a)$$

and

$$\rho_1 \frac{\partial v}{\partial t} + \frac{\mu}{\kappa_1} v = - \frac{\partial}{\partial r} P_1 \quad , \quad (2b)$$

where  $\mu$  is the dynamic viscosity of the liquid,  $g$  is the gravitational acceleration and  $\kappa_1$  is the relative permeability. Equations (2a,2b) take into account viscous drag, which is assumed to vary linearly with velocity, gravity and motion due to changes in pressure. Capillary forces enter Eqs.(2a,2b) through the terms involving the liquid pressure,  $P_1$ . The capillary pressure is defined as the difference between  $P_1$  and the gas pressure,  $P_g$  (that is,  $P_c = P_1 - P_g$ ) [51-55]. Taking the gas flow to be isobaric gives  $\nabla P_1 = -\nabla P_c$ . From Eqs.(2a,2b) it is therefore evident that capillary forces move liquid into regions of high  $P_c$ .

Leverett [55] derived the following relation for  $P_c$  using dimensional analysis,

$$P_c = J\gamma \left[ \frac{1-\alpha_s}{\kappa} \right]^{1/2} \quad , \quad (3)$$

where  $\gamma$  is the surface tension,  $\kappa$  is the permeability and  $J$  is the Leverett function. He further noted that  $J$  is only a function of the saturation

$$S \equiv \frac{\alpha_l}{\alpha_l + \alpha_g} \quad , \quad (4)$$

and the residual saturation  $S_r$  (which is defined as the threshold value of  $S$  below which bulk liquid motion ceases). Physically,  $S$



represents the fraction of the porosity that is occupied by liquid. Bird et al. [57] derive a relation for  $\kappa$  by modelling the porous solid as a bundle of capillary tubes,

$$\kappa = \frac{d_p^2 (1-\alpha_s)^3}{150 \alpha_s^2} , \quad (5)$$

where the factor of 150 is determined empirically. Combining Eq.(3,5) gives

$$P_c = J\gamma \frac{\sqrt{150} \alpha_s}{d_p (1-\alpha_s)} , \quad (6)$$

Note that  $P_c$  decreases as the particle diameter  $d_p$ , increases. For  $\gamma \approx 0.5$  N/m, capillary forces are only important when  $d_p$  is on the order of 1 mm (or smaller) [50]. Increasing  $\alpha_s$  increases  $P_c$  and consequently, capillary forces tend to move liquid into regions of higher solid fraction (lower porosity).

Empirical correlations are needed for  $J$ , the relative permeability  $\kappa_1$ , and the residual saturation  $S_r$ . Hofmann and Barleon [58] give for  $J$

$$J = a (S_e + b)^{-c} , \quad (7)$$

where  $S_e [ \equiv (S - S_r) / (1 - S_r) ]$  is an effective saturation,  $a=0.38$ ,  $b=0.014$  and  $c=0.27$ . Equation (7) agrees with the results of Reed et al [23] except near  $S_e=0$ . The primary advantage of Eq.(7) is the absence of a singularity at  $S_e=0$ . Note that  $J$  and  $P_c$  reach their maximum value in regions of low saturation. Therefore, capillary forces tend to move liquid into regions of low saturation [51-55].

In fully saturated flow ( $S=S_e=1$ ),  $\kappa_1$  equals the permeability,  $\kappa$ , while in undersaturated flow, only a fraction of the solid is wetted and  $\kappa_1$  is proportional to  $\kappa$  (with the proportionality being a function of  $S$ ). Reed et al. [23] give for  $\kappa_1$

$$\kappa_1 = \begin{cases} \kappa S_e^3 & , \text{ for } S > S_r \\ 0 & , \text{ for } S \leq S_r \end{cases} \quad (8)$$

where  $S_e = (S - S_r) / (1 - S_r)$ . As discussed earlier, liquid does not begin to flow until the saturation reaches a threshold value,  $S_r$ . For  $S \leq S_r$ ,  $\kappa_1 = 0$  and Eq. (2) requires that  $q = 0$ . When  $S$  is less than  $S_r$ , the liquid consists of unconnected pendular rings. Liquid starts to flow when  $S$  is increased to the point that these rings touch and coalesce [55].

Brown et al. [59] give for the residual saturation  $S_r$ ,

$$S_r = \frac{1}{86.3} \left[ \frac{\gamma}{\kappa \rho_1 g} \right]^{0.263} \quad (9)$$

Thus,  $S_r$  varies with the ratio of surface tension to gravity. The dependence of  $S_r$  on the porous matrix is contained in the permeability  $\kappa$ . Increasing the particle diameter or the porosity raises  $\kappa$  and leads to lower values of  $S_r$ . For packed beds consisting of small, tightly packed particles, the saturation must be increased to a high value before bulk liquid motion is observed.

### 2.1.3 Solid Collapse

As solid melts in the center of the bed and flows outward under the action of gravity and capillary forces, the porosity in the melt zone increases. In-pile experiments have indicated that the melt zone can be self-supporting at high porosities (up to 0.7) [33-37]. In this study it is assumed that debris starts collapsing when the solid fraction decreases to a critical value  $\alpha_{s,\min}$  and that  $\alpha_s = \alpha_{s,\min}$  in regions into which solid is settling. Experiments are needed to determine  $\alpha_{s,\min}$  as a function of particle diameter and initial porosity. Uncertainties in  $\alpha_{s,\min}$  will affect the configuration of the melt zone and the growth of the molten pool. In the calculations presented in Chapter 3  $\alpha_{s,\min}$  is set equal to 0.3 and most of the discussion is focused on pre-collapse phenomenon.

#### 2.1.4 Conservation of Energy

Assuming that all the materials present are in local thermal equilibrium, only one temperature field needs to be determined. Radiation heat transfer in the packed bed is incorporated utilizing a diffusion model with a temperature dependent conductivity. Balancing the energy stored in the solid and the liquid, convection by the liquid and the solid (as it collapses), diffusion and internal heat generation,

$$\begin{aligned} \frac{\partial}{\partial t} \sum_j [\alpha_s Y_{sj} \rho_{sj} h_{sj} + \alpha_l Y_{lj} \rho_{lj} h_{lj}] &= - \frac{\partial}{\partial z} [U \sum_j Y_{sj} \alpha_s \rho_{sj} h_{sj}] \\ &- \frac{\partial}{\partial z} [u \sum_j Y_{lj} \rho_{lj} h_{lj}] - \frac{1}{r} \frac{\partial}{\partial r} [rv \sum_j Y_{lj} \rho_{lj} h_{lj}] \\ &+ \frac{\partial}{\partial z} k_{\text{eff}} \frac{\partial T}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} rk_{\text{eff}} \frac{\partial T}{\partial r} \\ &+ [\alpha_s \rho_{s1} Y_{s1} + \alpha_l \rho_{l1} Y_{l1}] Q, \end{aligned} \quad (10)$$

where  $h_{ij}$  is the enthalpy of species  $j$  in phase  $i$ ,  $Q$  is the decay heat expressed as energy release per mass of  $\text{UO}_2$  and  $k_{\text{eff}}$  is an effective thermal conductivity which accounts for both conductive and radiative heat transfer in the porous solid. Note that energy stored in the gas phase is neglected in Eq. (10). This is a reasonable approximation because the densities of materials of interest are very large (on the order of  $10^4 \text{ kg/m}^3$ ) compared with the gas density.

Radiation heat transfer in the packed bed is incorporated using a modified gas conductivity,  $k_g^* = k_g + k_{\text{rad}}$  [60,61]. That is, it is assumed that gas conduction and radiation act in parallel. Several researchers [60-64] have proposed that  $k_{\text{rad}} = 4\epsilon\sigma d T^3$ , where  $\epsilon$  is the emissivity of the solid and  $\sigma$  is the Stefan-Boltzmann constant. Empirical correlations for  $k_{\text{eff}}$  in solid-gas systems are available in the literature [60,61,64]. However, in the current problem, three phases (solid, liquid and gas) are present. In calculating  $k_{\text{eff}}$ , the solid and the liquid are treated as a single component with a volume averaged thermal conductivity,

$$k_{\sigma} = \frac{1}{\alpha_s + \alpha_1} \left[ \alpha_s \sum_j Y_{sj} k_{sj} + \alpha_1 \sum_j Y_{1j} k_{1j} \right] \quad , \quad (11)$$

where  $k_{ij}$  is the conductivity of species  $j$  in phase  $i$ . For a single phase  $i$ ,  $k_{\sigma}$  depends only on  $Y_{ij}$  and  $k_{ij}$ . That is,  $k_{\sigma} = Y_{s1}k_{s1} + Y_{s2}k_{s2}$  for  $\alpha_1=0$  (solid only) and  $k_{\sigma} = Y_{11}k_{11} + Y_{12}k_{12}$  for  $\alpha_s=0$  (liquid only). The following correlation is used to calculate  $k_{eff}$  [60,64],

$$k_{eff} = \psi k_g^* + \frac{1-\psi}{k_{\sigma}w + k_g^*(1-w)} k_{\sigma}k_g^* \quad , \quad (12)$$

where

$$w = 0.3 \alpha_g^{1.6} (k_{\sigma}/k_g^*)^{-0.044} \quad , \quad (13)$$

$$\psi = \frac{\alpha_g^{-w}}{1-w} \quad , \quad (14)$$

and  $\alpha_g$  is the volume fraction occupied by gas. Note that as  $\alpha_g \rightarrow 0$ ,  $w \rightarrow 0$ ,  $\psi \rightarrow 0$  and consequently,  $k_{eff} \rightarrow k_{\sigma}$ . As  $\alpha_g \rightarrow 1$ ,  $\psi \rightarrow 1$ , giving  $k_{eff} \rightarrow k_g^*$ .

### 2.1.5 Closure

In order to complete Eqs. (1-14) a phase diagram, initial conditions, boundary conditions and properties must be specified. Shown in Fig.1 is an equilibrium phase diagram for a homogeneous  $UO_2$ - $ZrO_2$  mixture. Figure 1 was constructed from the solidus and liquidus temperatures ( $T_s$  and  $T_l$ , respectively) given in Ref. [65]. Experiments needed to check the validity of the equilibrium assumption are discussed in Chapter 4. A discussion of the effect of diffusion at the particulate level is given by Dosanjh [50].

An initial uniform temperature  $T_i$  is prescribed. Boundaries continue to radiate to environments at this temperature as the bed temperature increases. The bed is assumed to have an initial porosity of 0.4 and the composition of the bed is taken to be uniform over distances characteristic of changes in  $T$ ,  $S$  and  $\alpha$ .

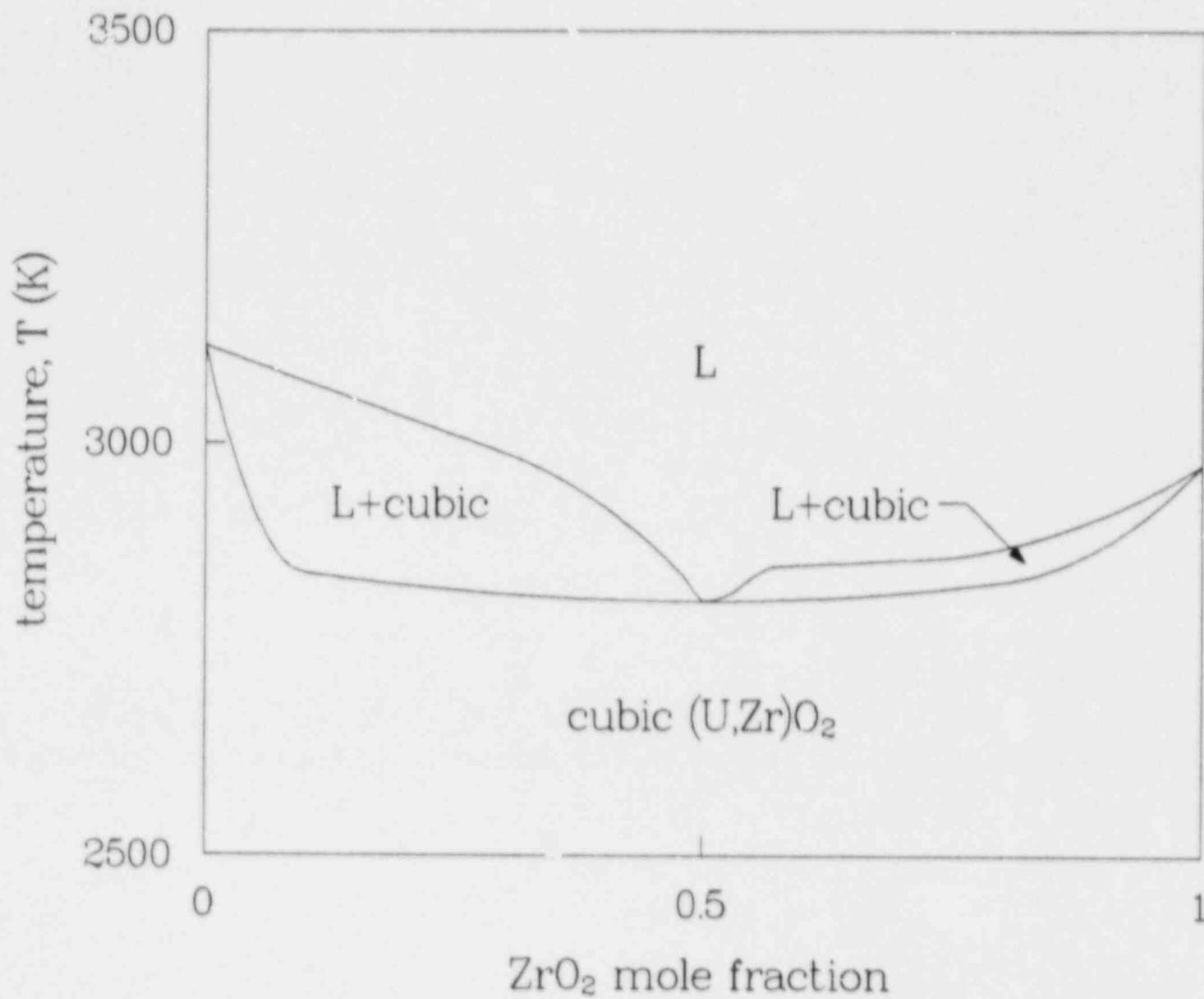


FIG. 1: Phase diagram for  $\text{UO}_2\text{-ZrO}_2$ .

Typical values of densities  $\rho_{ij}$ , specific heats  $c_{pij}$ , heats of fusion  $h_{fj}$ , thermal conductivities  $k_{ij}$ , viscosities  $\mu_j$  and surface tensions  $\gamma_j$  for  $UO_2$  and  $ZrO_2$  are given in Table II. Initial measurements of the liquid  $UO_2$  thermal conductivity ranged from 2.0 W/(mK) to 11.0 W/(mK) [66,67]. Upon subsequent analysis of these experiments, Fink and Leibowitz [71] suggested a value of 5.5 W/(mK). The gas is taken to be argon, for which  $k_g = 2.986 \times 10^{-4} T^{0.7224}$ , where  $k_g$  is measured in W/m $\cdot$ K and T is in Kelvins [3]. The viscosity of the melt is calculated by volume averaging. For simplicity, properties are assumed to remain constant in the calculations presented here.

TABLE II

Typical  $UO_2$  and  $ZrO_2$  properties given in Refs. [3,65-72].

Property	$UO_2$ (j=1)	$ZrO_2$ (j=2)
theoretical solid density, $\rho_{sj}$ [kg/m <sup>3</sup> ]	9,650	5,700
liquid density, $\rho_{lj}$ [kg/m <sup>3</sup> ]	8,700	5,700
solid specific heat, $c_{psj}$ [J/kg $\cdot$ K]	630	700
liquid specific heat, $c_{plj}$ [J/kg $\cdot$ K]	490	815
solid thermal conductivity, $k_{sj}$ [W/m $\cdot$ K]	3.0	2.0
liquid thermal conductivity, $k_{lj}$ [W/m $\cdot$ K]	5.5	2.7
heat of fusion, $h_{fj}$ [J/kg]	274,000	706,000
viscosity, $\mu_j$ [Pa $\cdot$ s]	0.0058	0.0035
surface tension, $\gamma_j$ [N/m]	0.45	0.45

## 2.2 Order of Magnitude Analysis

Consider a debris bed with an initial temperature profile,  $T_i(z,r)=T(z,r,0)$ . If the distance characteristic of changes in  $T_i$  is on the order of L, the height of the bed, conduction and radiation in the bed become important over the entire domain at time  $t=L^2/\alpha_t$ , where  $\alpha_t$  is an average solid thermal diffusivity. Using the  $UO_2$  properties in Table II, setting  $\epsilon=0.4$  and evaluating  $k_{eff}$  at 2000 K, gives  $\alpha_t \approx k_{eff}/[(1-\epsilon)\rho_s c_{ps}] \approx 10^{-6}$  m<sup>2</sup>/s. Rubble beds with heights on the order of 1 m heat up adiabatically until time  $L^2/\alpha_t \approx 10^6$  s. For the times of interest,  $T(z,r,t)$  is highly dependent on initial conditions and

conduction and radiation are only important within boundary layers of thickness  $\delta \approx [a_t t]^{1/2}$ . In the region  $\delta \ll x \ll L - \delta$ ,

$$T(z, r, t) - T(x, r, 0) = \frac{Y_{s1} \rho_{s1} Q t}{2 \sum_{j=1} Y_{sj} \rho_{sj} c_{psj}} \quad (15)$$

which is valid until melt forms. Setting  $Q = 300$  W/kg of  $UO_2$ ,  $Y_{s1} = 0.64$  and utilizing the properties in Table II gives a 0.35 K/s as an approximate lower bound for the heating rate. For beds composed of only  $UO_2$  particles ( $Y_{s1} = 1$ ), Eq. (15) gives a heating rate of 0.5 K/s. Therefore, it takes between 2000 s and 2860 s for the temperature of the bed to increase by 1000 K.

### 2.3 Solution Algorithm

Equations (1.2,10) along with auxiliary relations (3-9,11-14) are solved using an explicit finite difference scheme. Solution of these equations is somewhat complicated by the fact that time derivatives appearing in Eqs. (1,10) involve products of the primitive variables,  $T$ ,  $a_s$ ,  $u_1$ ,  $Y_{s1}$ ,  $Y_{s2}$ ,  $Y_{11}$  and  $Y_{12}$ . Given the values of all variables at a time step  $n$ , the solution algorithm is as follows. The velocity field is calculated at cell boundaries utilizing Eq. (2) and information at step  $n$ . Next, the bulk densities (which are based on total volume) of  $UO_2$  and  $ZrO_2$ ,

$$\tilde{\rho}_{UO_2} = a_s Y_{s1} \rho_{s1} + a_1 Y_{11} \rho_{11} \quad (16)$$

and

$$\tilde{\rho}_{ZrO_2} = a_s Y_{s2} \rho_{s2} + a_1 Y_{12} \rho_{12} \quad (17)$$

respectively, and the enthalpy function,

$$H = \sum_j \left[ a_s Y_{sj} \rho_{sj} h_{sj} + a_1 Y_{1j} \rho_{1j} h_{1j} \right] \quad (18)$$

are calculated at time step  $n+1$  at each calculation node from Eqs. (1,10). When only one phase is present, solution of

Eqs.(1,10) is straightforward. In two phase regions, phase diagram provides additional relations of the form

$$\frac{Y_{s2}\rho_{s2}/M_2}{\sum_{j=1}^2 Y_{sj}\rho_{sj}/M_j} = f(T) \quad (19)$$

and

$$\frac{Y_{l2}\rho_{l2}/M_2}{\sum_{j=1}^2 Y_{lj}\rho_{lj}/M_j} = g(T) \quad (20)$$

where  $M_j$  is the molecular weight of species  $j$  and  $f$  and  $g$  are specified functions of temperature. Terms on the left hand side of Eqs.(19,20) represent mole fractions of  $ZrO_2$  in the solid and liquid phases, respectively. Equations (16-20) along with the relations,  $Y_{s2}=1-Y_{s1}$  and  $Y_{l2}=1-Y_{l1}$ , are then solved for the primitive variables,  $T$ ,  $a_s$ ,  $a_l$ ,  $Y_{s1}$ ,  $Y_{s2}$ ,  $Y_{l1}$  and  $Y_{l2}$ .



### 3. RESULTS AND DISCUSSION

Solutions are presented in the following sections for a 0.5 m high  $\text{UO}_2\text{-ZrO}_2$  porous bed with a diameter of 0.5 m, an average particle diameter of 1 mm, an initial uniform porosity of 0.4 and a  $\text{ZrO}_2$  to  $\text{UO}_2$  mass ratio of 0.1. The bed is initially at a uniform temperature of 1500 K and the boundaries continue to radiate to an environment at 1500 K as the temperature of the bed increases due to decay heating. It is assumed in these calculations that the solid begins collapsing when  $\alpha_s=0.3$ . The effect of varying the particle diameter will be discussed in section 3.2.

#### 3.1 Base Case

Temperature contours in the bed at  $t=3200$  s are shown in Fig. 2. Because of the low thermal conductivities involved, the temperature in the bed is fairly uniform except in narrow thermal boundary layers. For a  $\text{ZrO}_2$  to  $\text{UO}_2$  mass ratio of 0.1 (which corresponds to a  $\text{ZrO}_2$  mole fraction of 0.18), Fig. 1 gives a solidus temperature of 2830 K. Therefore, the 2800 K contour approximately encloses the melt zone. A plot of the solid fraction  $\alpha_s$  at this time is shown in Fig. 3. Note that  $\alpha_s$  has decreased in the center of the bed (there is a corresponding increase in  $\alpha_l$ ). No liquid motion is evident because the saturation is below the critical residual saturation  $S_r$ . That is, liquid is held in place by surface tension effects.

Liquid motion is evident at  $t=3400$  s (see Fig. 4). Melt flowing downward and radially outward has refrozen, forming a crust as evidenced by the appearance of the cross hashed region. Radial relocation is driven by capillary forces, which tend to move liquid into regions of higher solid fraction. Downward relocation is due to both gravity and capillary forces. A crust has not formed above the melt zone at this time because gravity opposes upward relocation. As shown by the contours in Fig. 5, temperatures in regions adjacent to the melt zone increase as liquid freezes, giving up its heat of fusion.

Because the beds of interest are rich in  $\text{UO}_2$ , the  $\text{ZrO}_2$  mole fraction is initially larger in the liquid than in the solid (see Fig. 1). As liquid relocates the local  $\text{ZrO}_2$  mass fraction

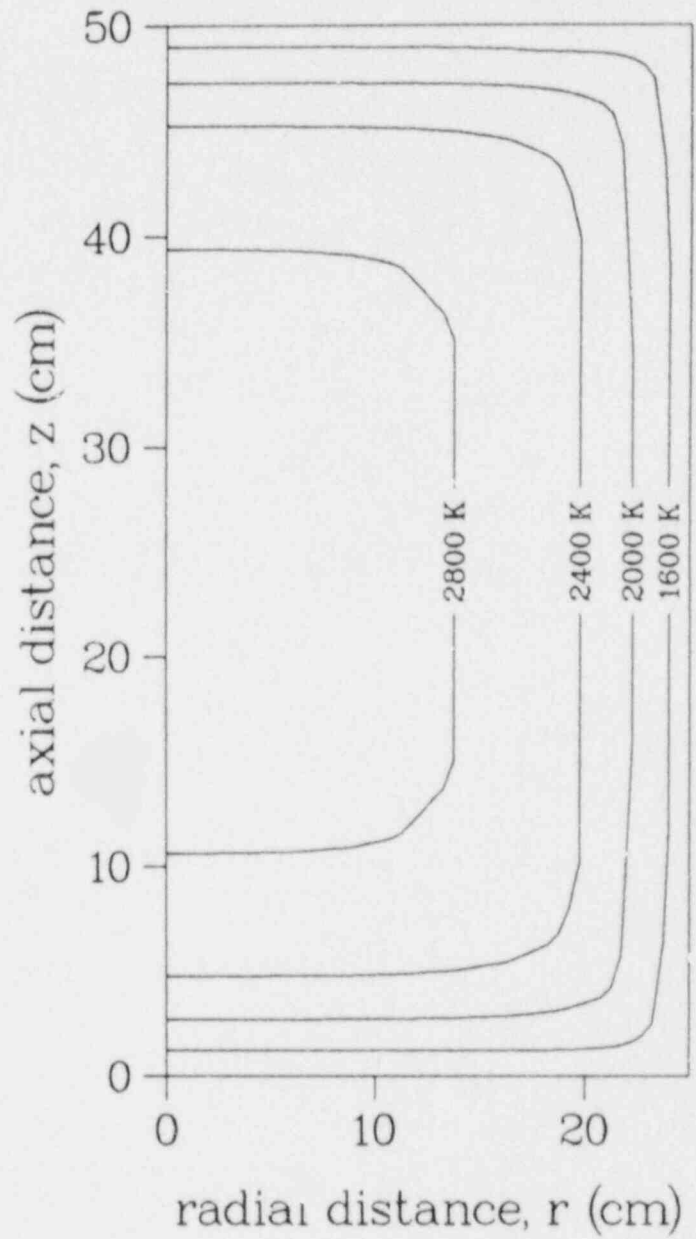


FIG. 2: Temperature contours in rubble bed at  $t=3200$  s for the base case. Contours are shown every 400 K.

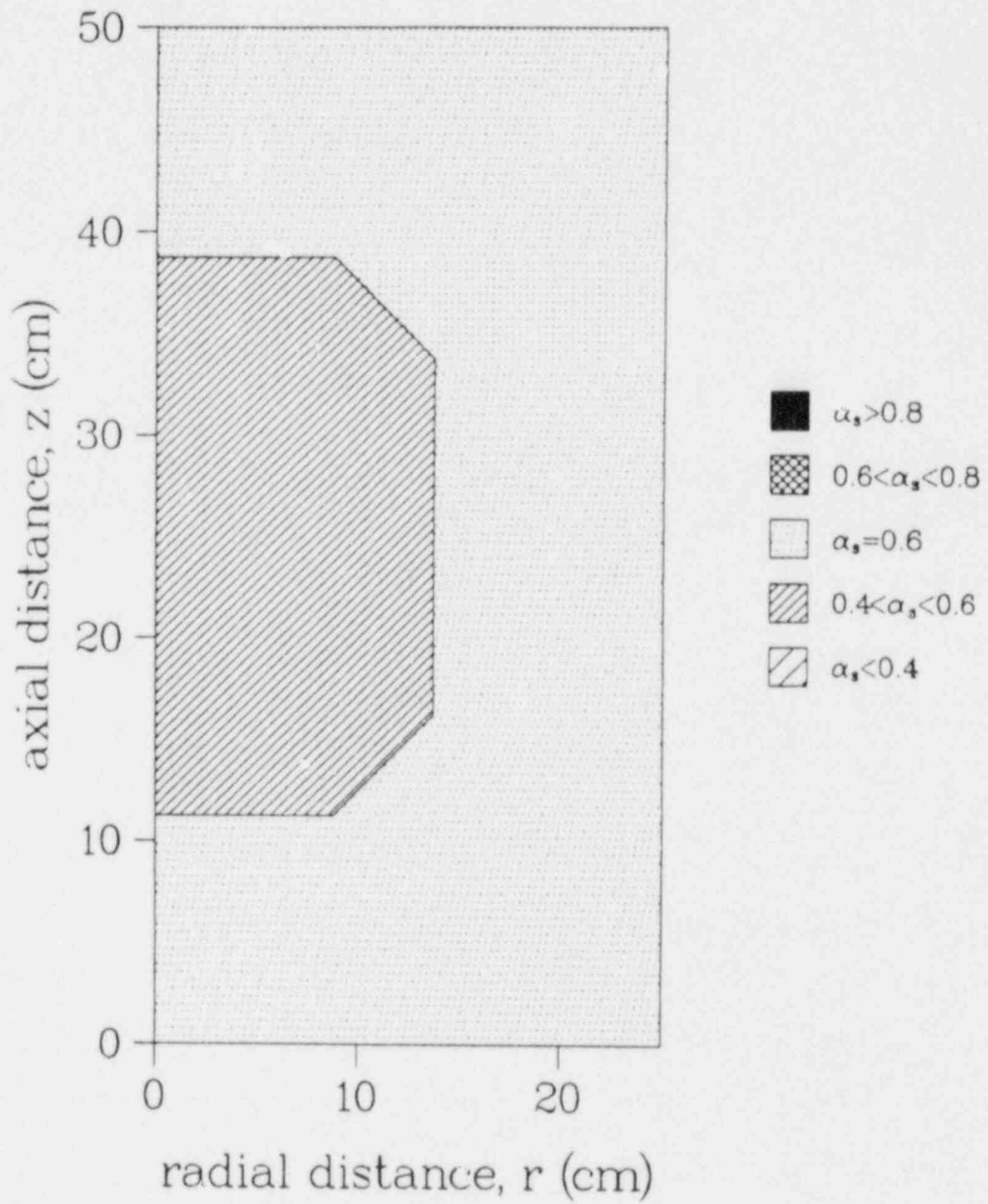


FIG. 3: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3200$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

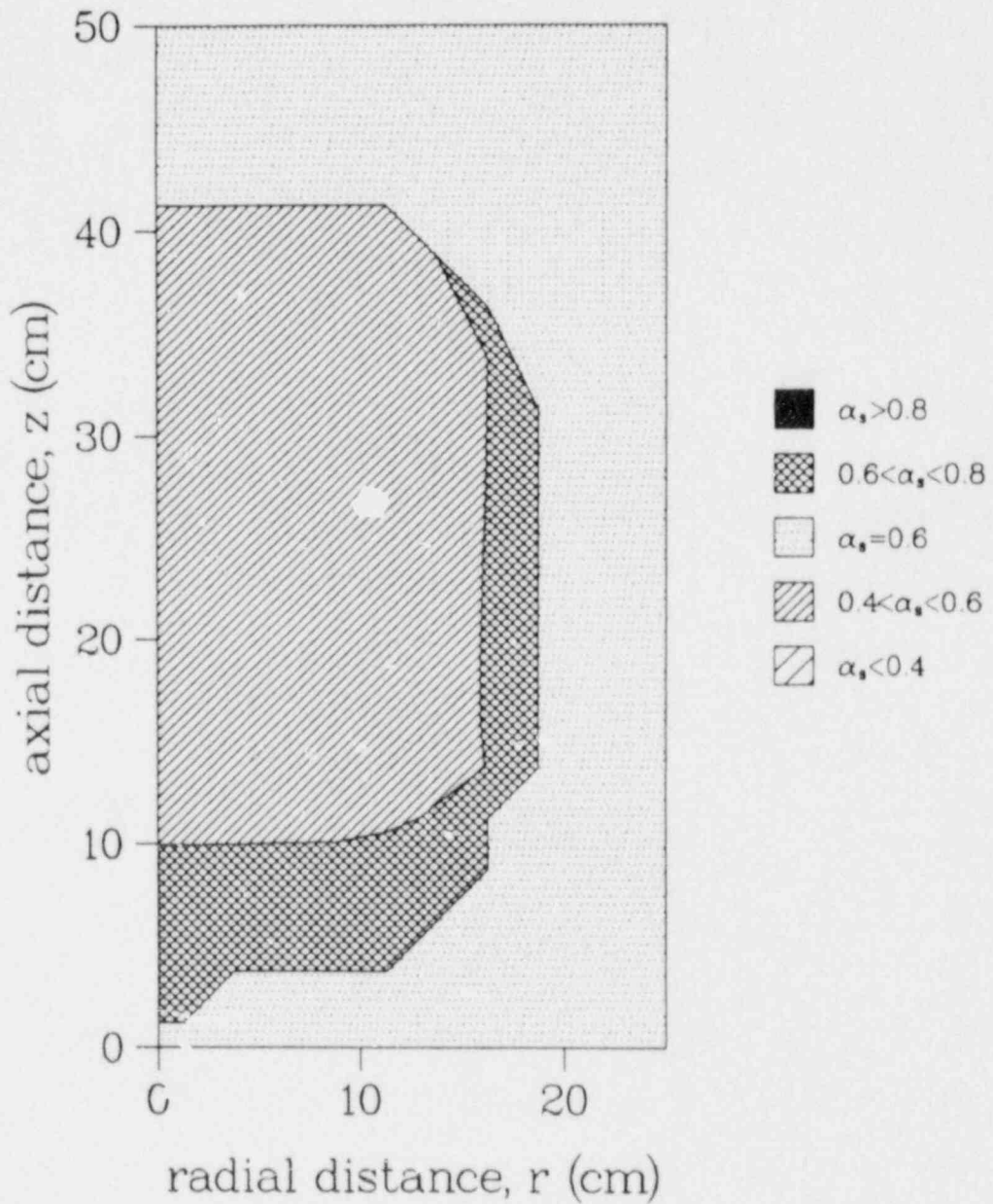


FIG. 4: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3400$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

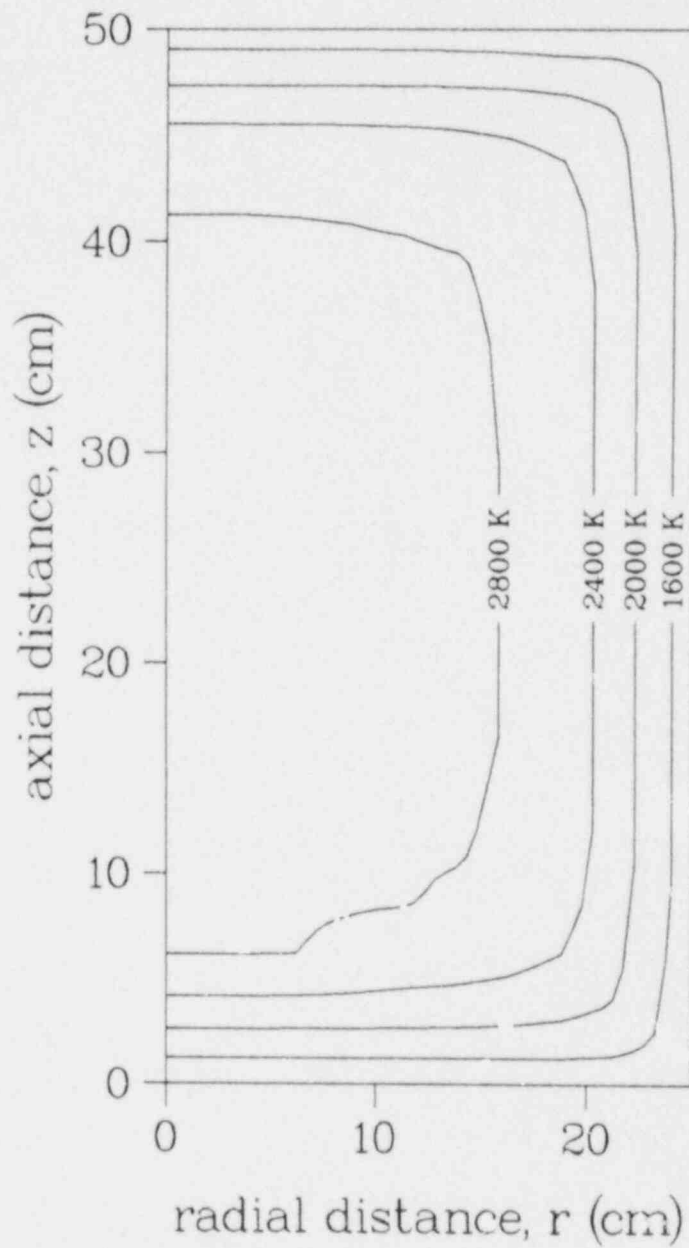


FIG. 5: Temperature contours in rubble bed at  $t=3400$  s for the base case. Contours are shown every 400 K.

changes. A normalized  $UO_2$  to  $ZrO_2$  mass ratio  $\eta$  is defined such that  $\eta=1$  initially. In a bed with an initial  $ZrO_2$  mole fraction of 0.5, the solidus and liquidus temperatures both equal 2810 K and consequently,  $\eta$  remains uniform as melt forms and relocates. Contours of  $\eta$  at 3400 s and 3600 s for the base case are shown in Figs. 6 and 7, respectively. Note that  $ZrO_2$  is depleted in the center of the bed and accumulates in the crust regions. At  $t=3600$  s,  $\eta$  varies from 0.5 in the center to 2.0 in the crust and the  $ZrO_2$  mole fraction changes from 0.1 to 0.3.

A blockage has formed below the melt zone before 3600 s elapse (see Fig. 8). Up to this time most of the melt has flowed downward under the action of gravity. As the solid fraction in the center of the bed continues to decrease, gradients of  $\alpha_s$  and  $P_c$  (which is related to  $\alpha_s$  by Eq.6) increase. That is, the driving force for capillary forces increases with time. As evidenced by Fig. 9, the solid fraction in the center of the bed is below 0.4 at  $t=3800$  s and a crust has begun to form above the melt zone.

The low solid fraction region ( $\alpha_s < 0.4$ ) in Fig. 9 continues to grow during the next 400 s (see Figs. 10 and 11). Liquid volume fractions at  $t=4200$  s are shown in Fig. 12. A two phase (liquid-solid) molten pool forms above the blockage as liquid accumulates. A small amount of liquid ( $\alpha_l < 0.1$ ) is suspended in the melt zone above this region by surface tension effects. Note that the liquid level increases towards the edges of the pool. This wicking is due to the variation of capillary rise  $x_c$  with solid fraction. An approximate relation for  $x_c$  can be obtained by balancing gravity and capillary forces in Eq.(2a), giving

$$x_c \approx \frac{\sqrt{150} a \gamma \alpha_s}{d_p \rho_g (1 - \alpha_s)} \quad (21)$$

In the region to the left of the blockage in Fig. 10,  $\alpha_s$  increases with  $r$ . Consequently,  $x_c$  increases near the edges of the pool. In the next section it will be shown that this effect is negligible for 5 mm diameter particles. The gas volume fraction,  $\alpha_g = 1 - \alpha_s - \alpha_l$ , at 4200 s is plotted in Fig.13. In the molten pool, the gas fraction is zero and the flow is fully

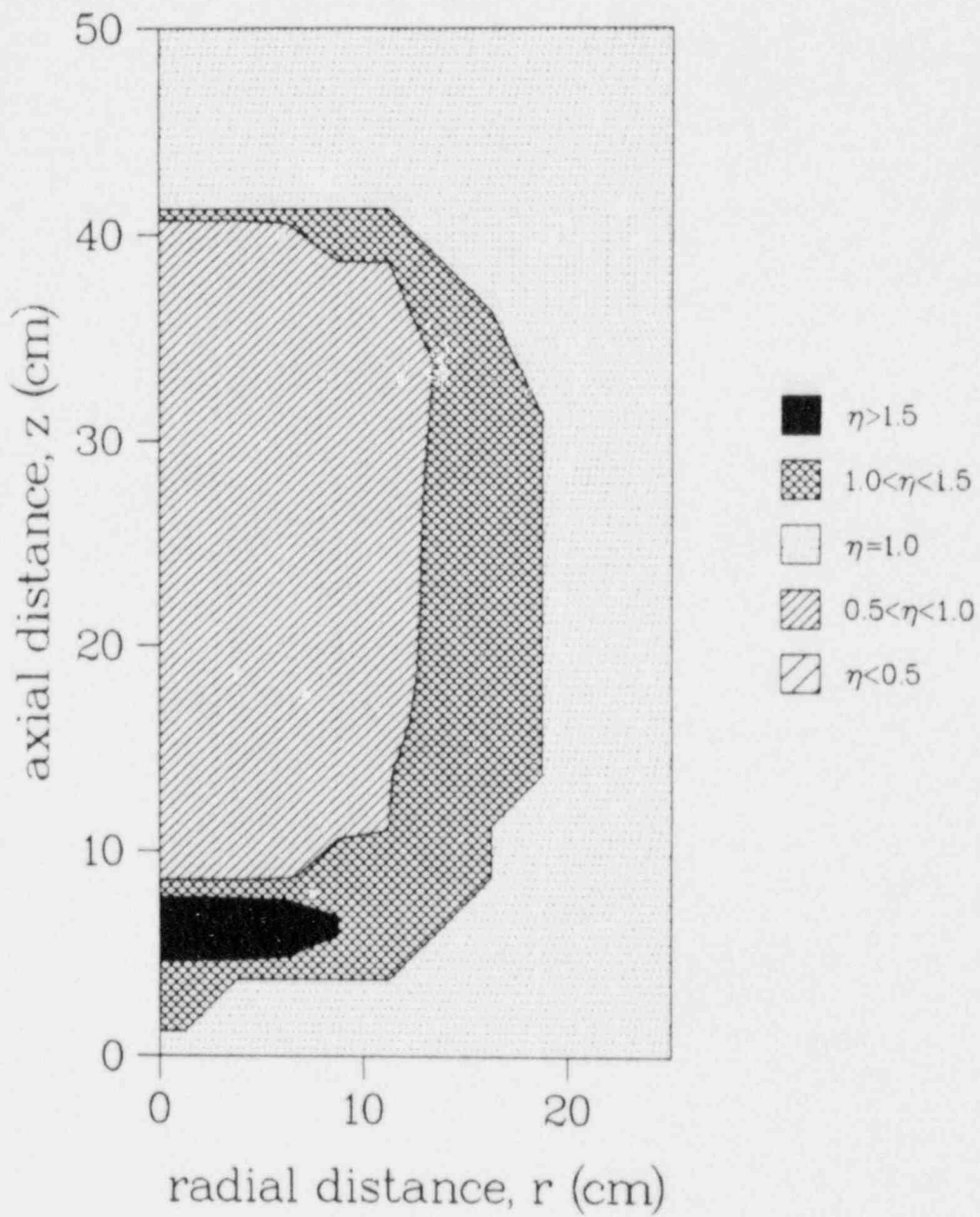


FIG. 6: Normalized  $\text{ZrO}_2$  to  $\text{UO}_2$  mass ratio  $\eta$  at  $t=3400$  s for the base case.

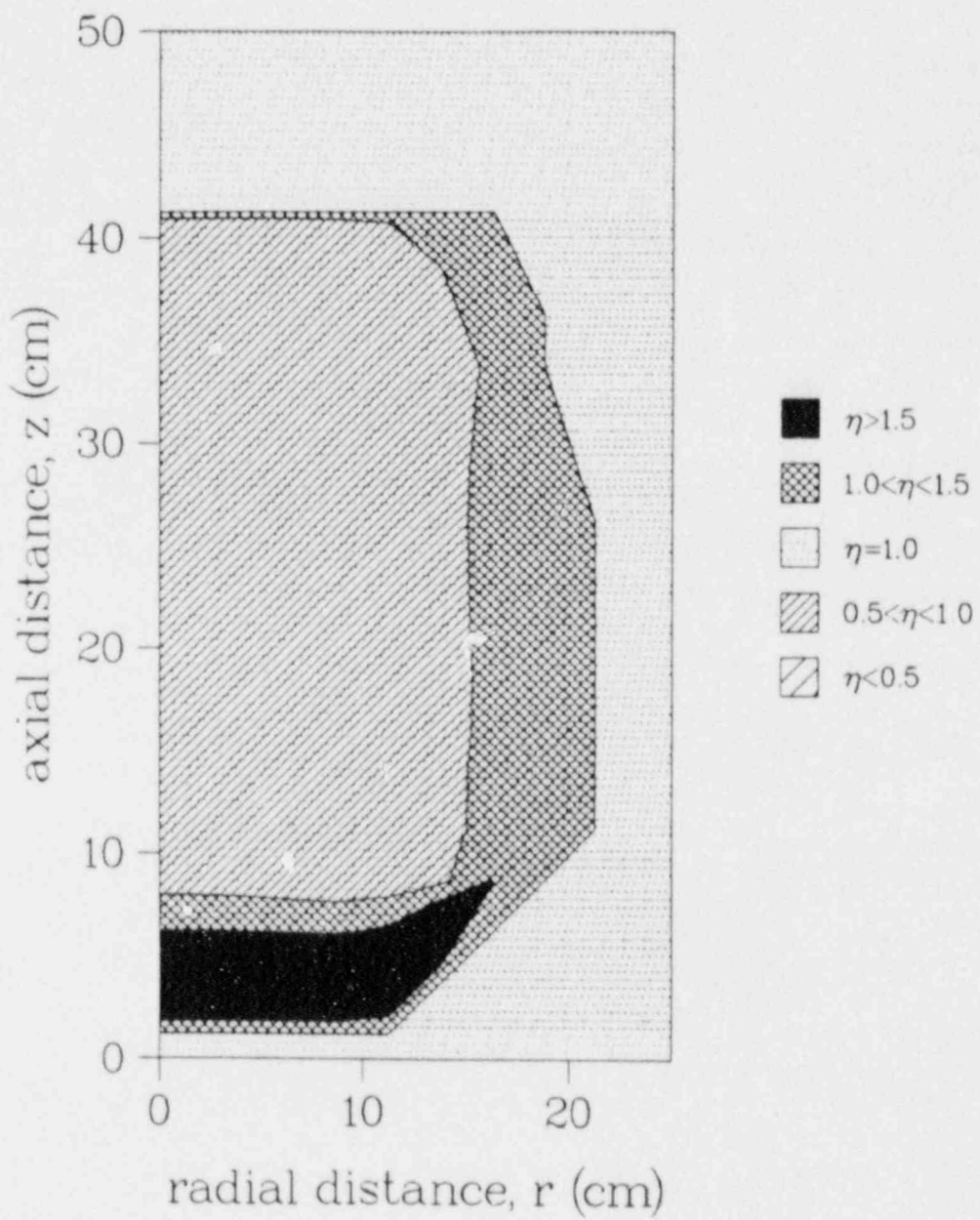


FIG. 7: Normalized  $\text{ZrO}_2$  to  $\text{UO}_2$  mass ratio  $\eta$  at  $t=3600$  s for the base case.



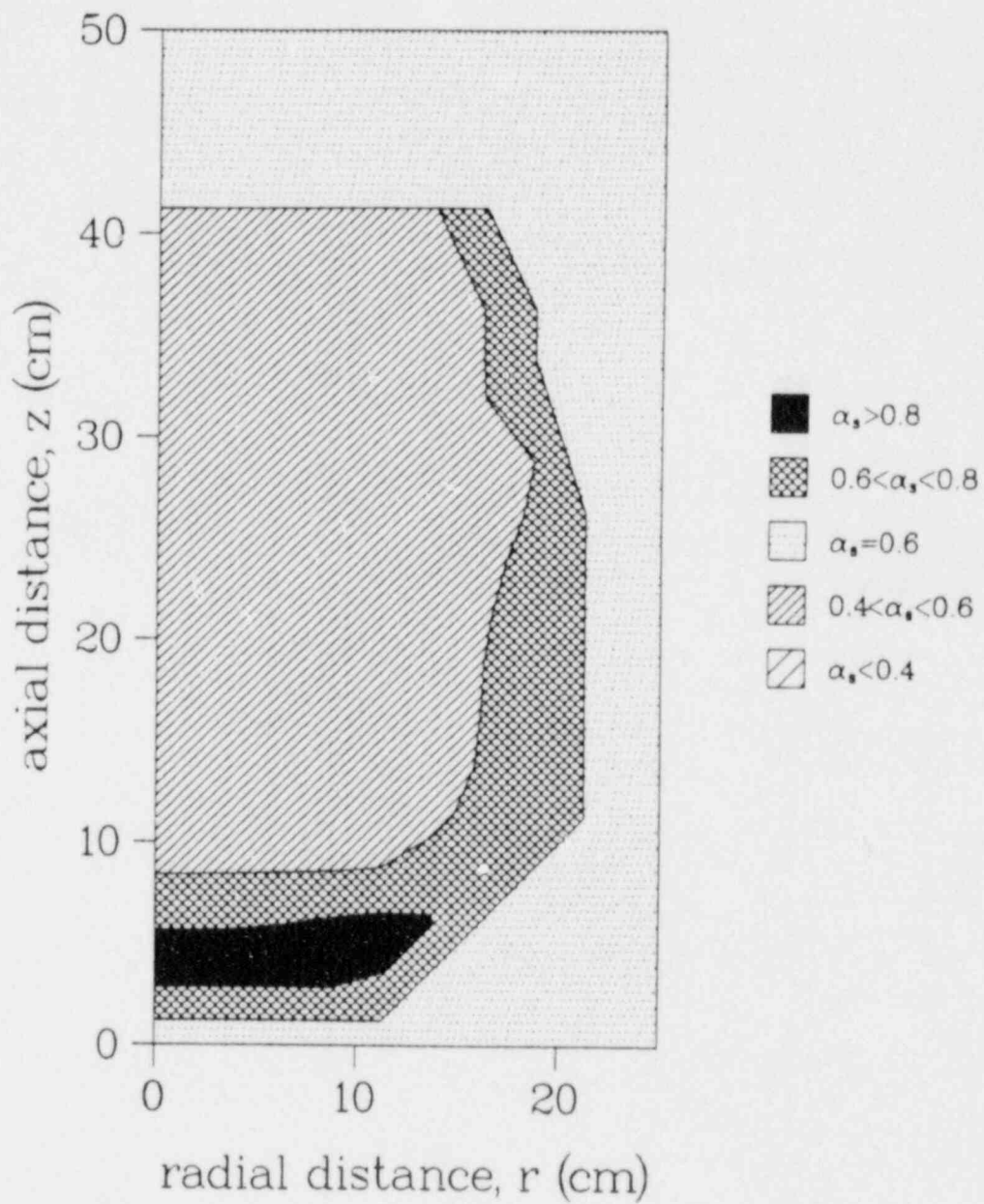


FIG. 8: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3600$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

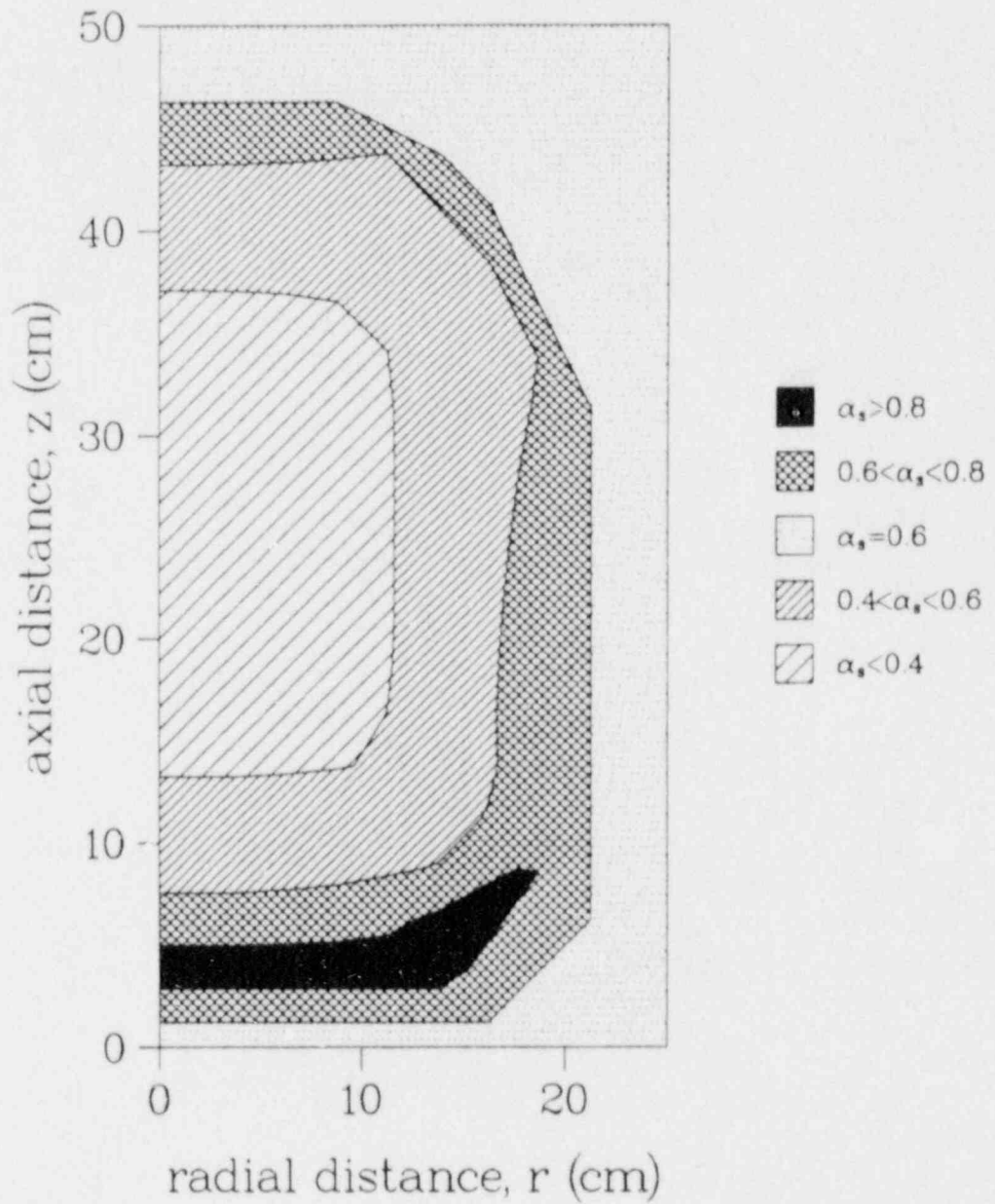


FIG. 9: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3800$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

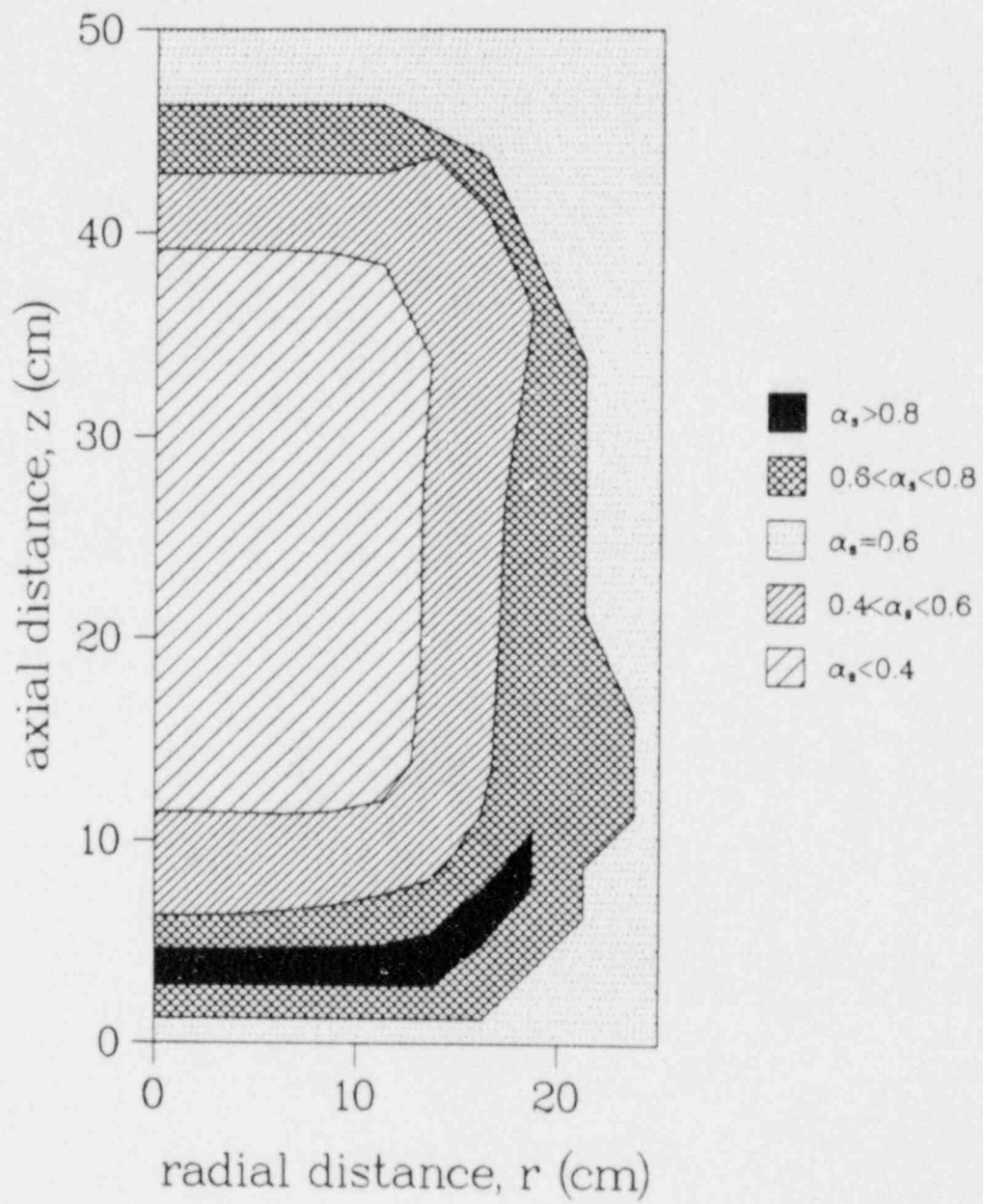


FIG. 10: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4000$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

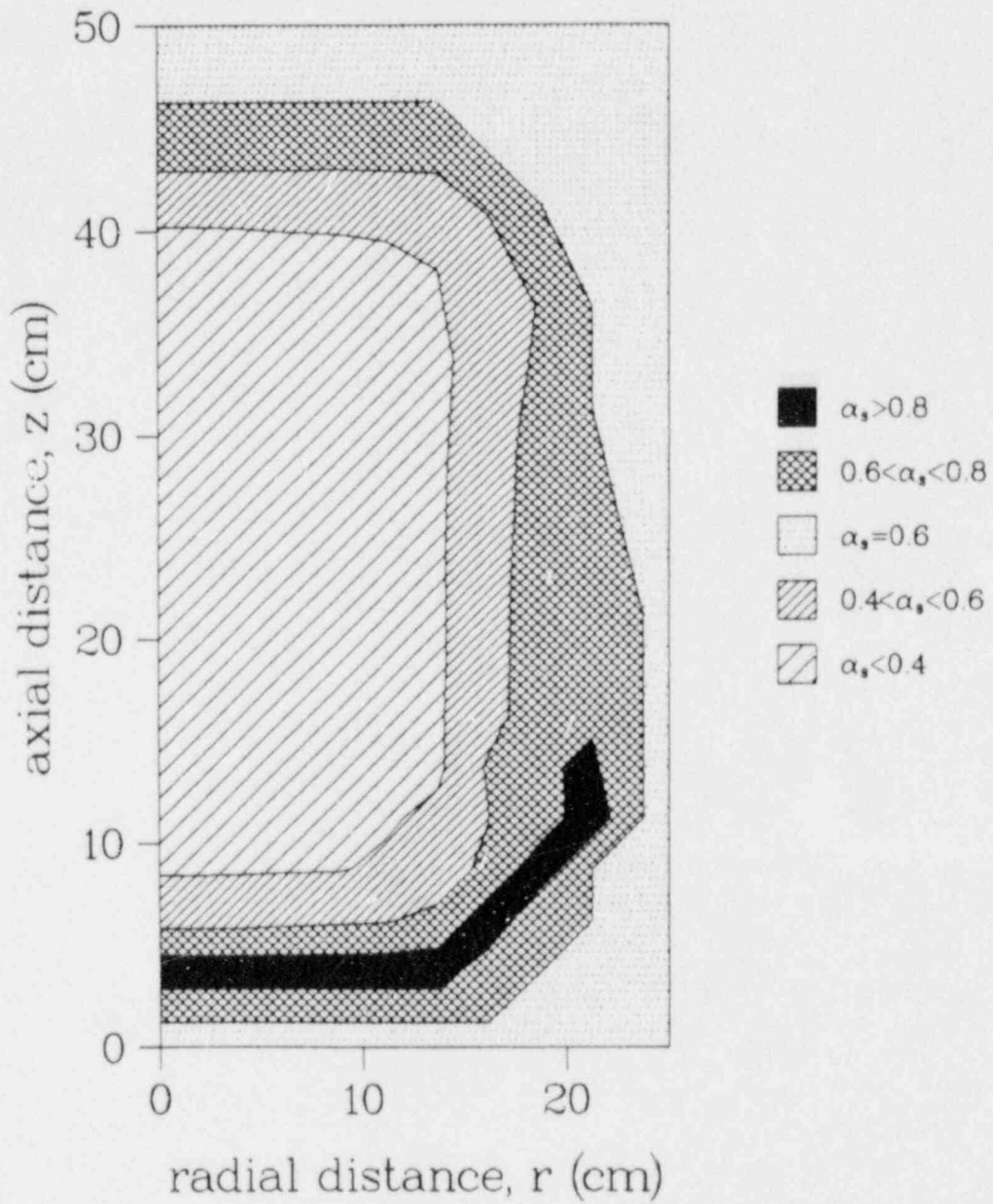


FIG. 11: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4200$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

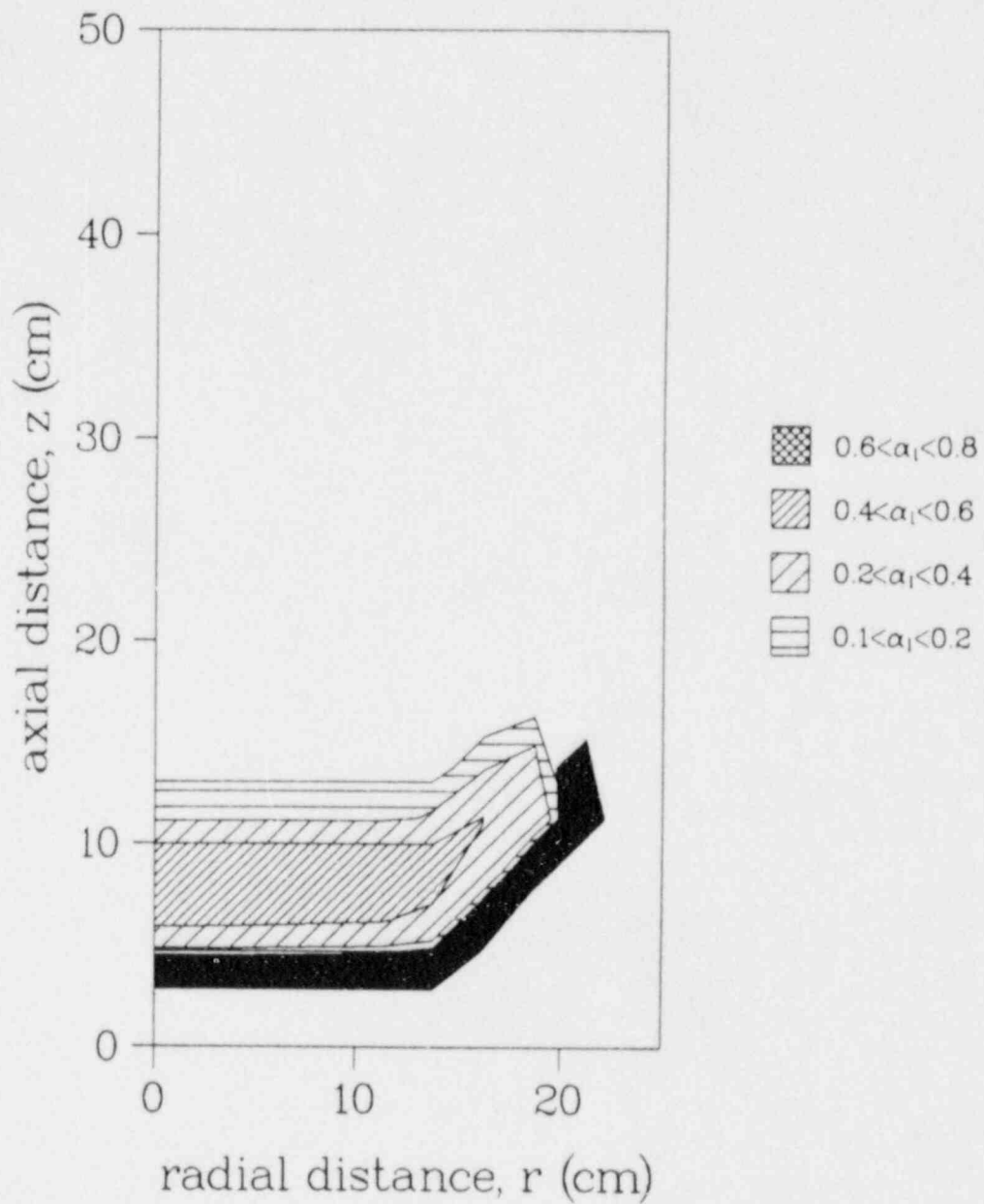


FIG. 12: Liquid volume fraction  $\alpha_1$  and the blockage region (in black) at  $t=4200$  s for the base case.

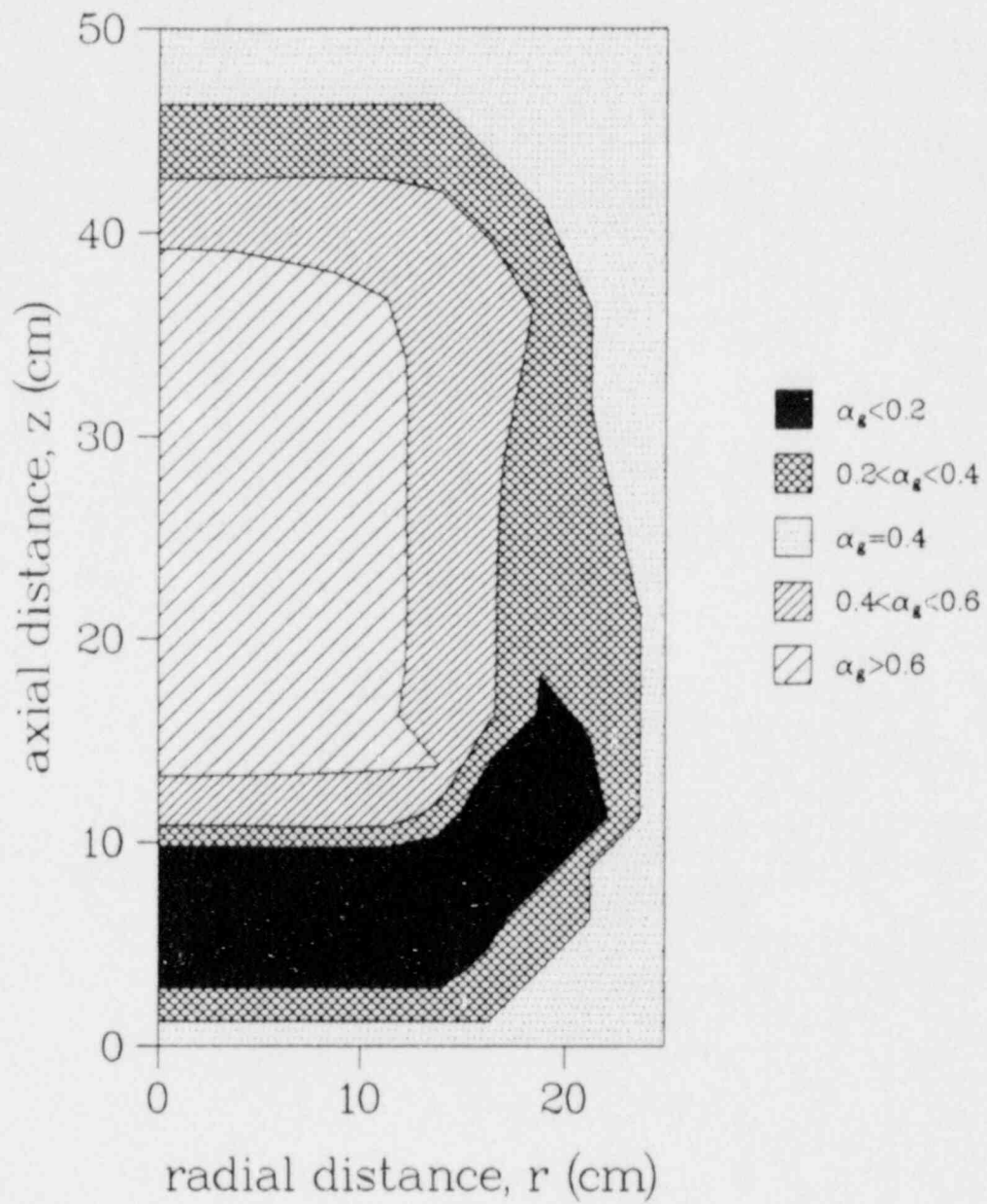


FIG. 13: Gas volume fraction  $\alpha_g$  in rubble bed at  $t=4200$  s for the base case. The rubble has an initial uniform porosity of 0.4.

saturated ( $S=1$ ). Because  $\alpha_s + \alpha_l = 1$  in the pool, variations of  $\alpha_l$  in Fig. 12 are attributable to changes in  $\alpha_s$ .

The porosity in the center of the bed reaches 0.7 at  $t=4200$  s and the solid begins settling downward. As is seen from Fig. 14, the temperature of the upper crust at 4200 s is below the solidus temperature. In the current calculation it is assumed that this crust can support the small amount of debris resting above it. Solid, liquid and gas volume fraction profiles at 4400 s are shown in Figs. 15, 16 and 17, respectively. A voided region has formed below the upper crust. In portions of the molten pool the solid volume fraction has decreased below 0.4 and the liquid volume fraction has increased above 0.6 (recall that  $\alpha_s + \alpha_l = 1$  in the pool).

Solid, liquid and gas volume fraction profiles at 4600 s are shown in Figs. 18, 19 and 20, respectively. As the high porosity solid in the center continues to settle downward, the size of the voided region increases. At  $t=4600$  s, this zone is 10 cm high and 11 cm in radius. Note that changes in the critical solid fraction  $\alpha_{s,min}$  will affect the growth of the voided region. Experiments are needed to evaluate the current bed collapse model and suggest possible improvements.

Zirconia and Urania melt fractions are plotted as functions of time in Figs. 21 and 22, respectively. Also shown are the fractions that have refrozen to form the crust. Solid begins melting at 3080 s and liquid starts refreezing soon after motion is first observed ( $\approx 3300$  s). At  $t=4500$  s, a third of the  $ZrO_2$  has undergone a phase change and almost sixty percent of the liquid  $ZrO_2$  has refrozen. Only twelve percent of the  $UO_2$  has melted, of which a small fraction ( $\approx 18\%$ ) has refrozen. A greater portion of the  $ZrO_2$  has melted because the concentration of  $ZrO_2$  in the liquid is initially larger than in the solid. This effect becomes less pronounced as time passes. As the  $ZrO_2$  is depleted, the rate at which  $UO_2$  melts increases rapidly while the rate of  $ZrO_2$  melt formation levels off. Because the bulk of the crust is formed early in the meltdown sequence, a large fraction of the  $ZrO_2$  refreezes.

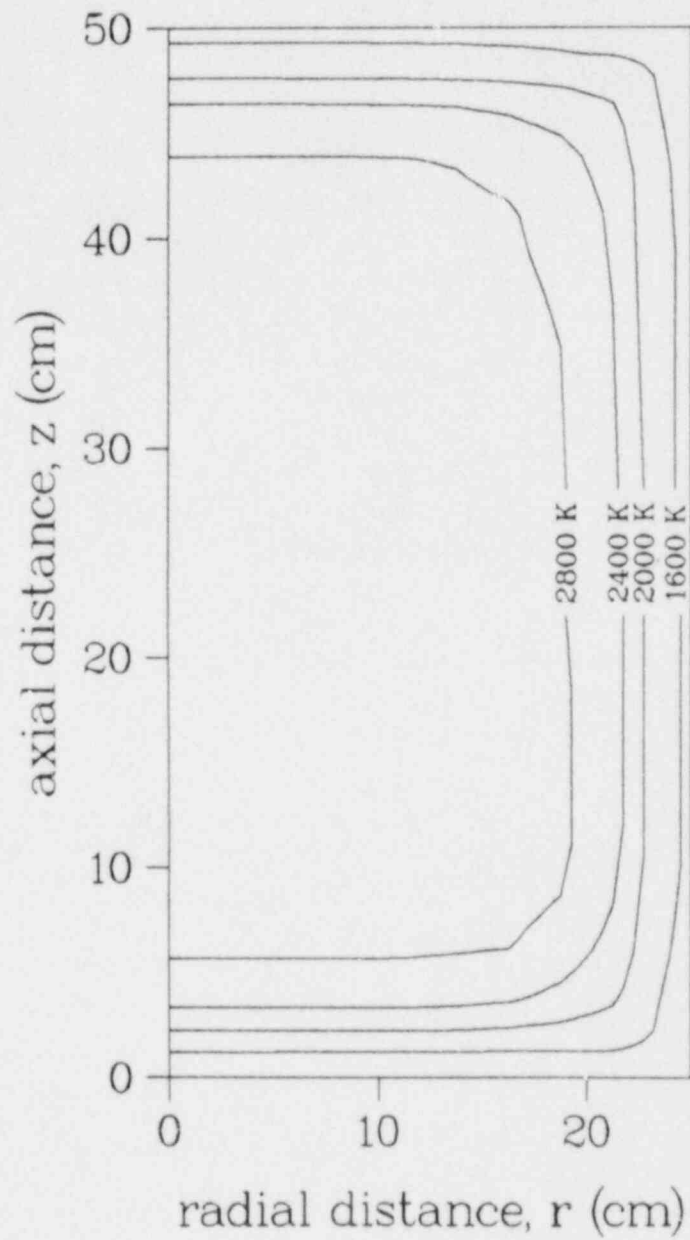


FIG. 14: Temperature contours in rubble bed at  $t=4200$  s for the base case. Contours are shown every 400 K.



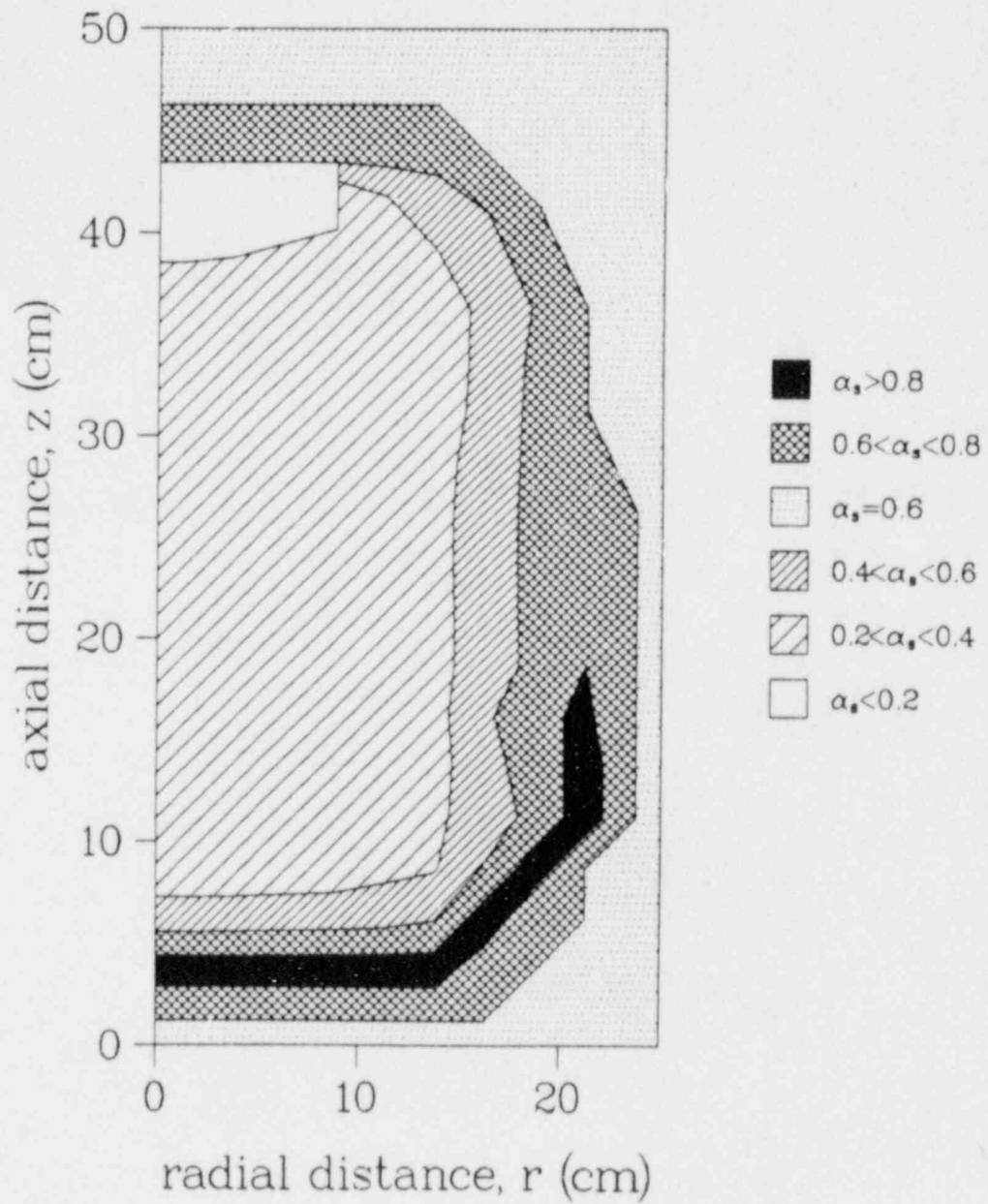


FIG. 15: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4400$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

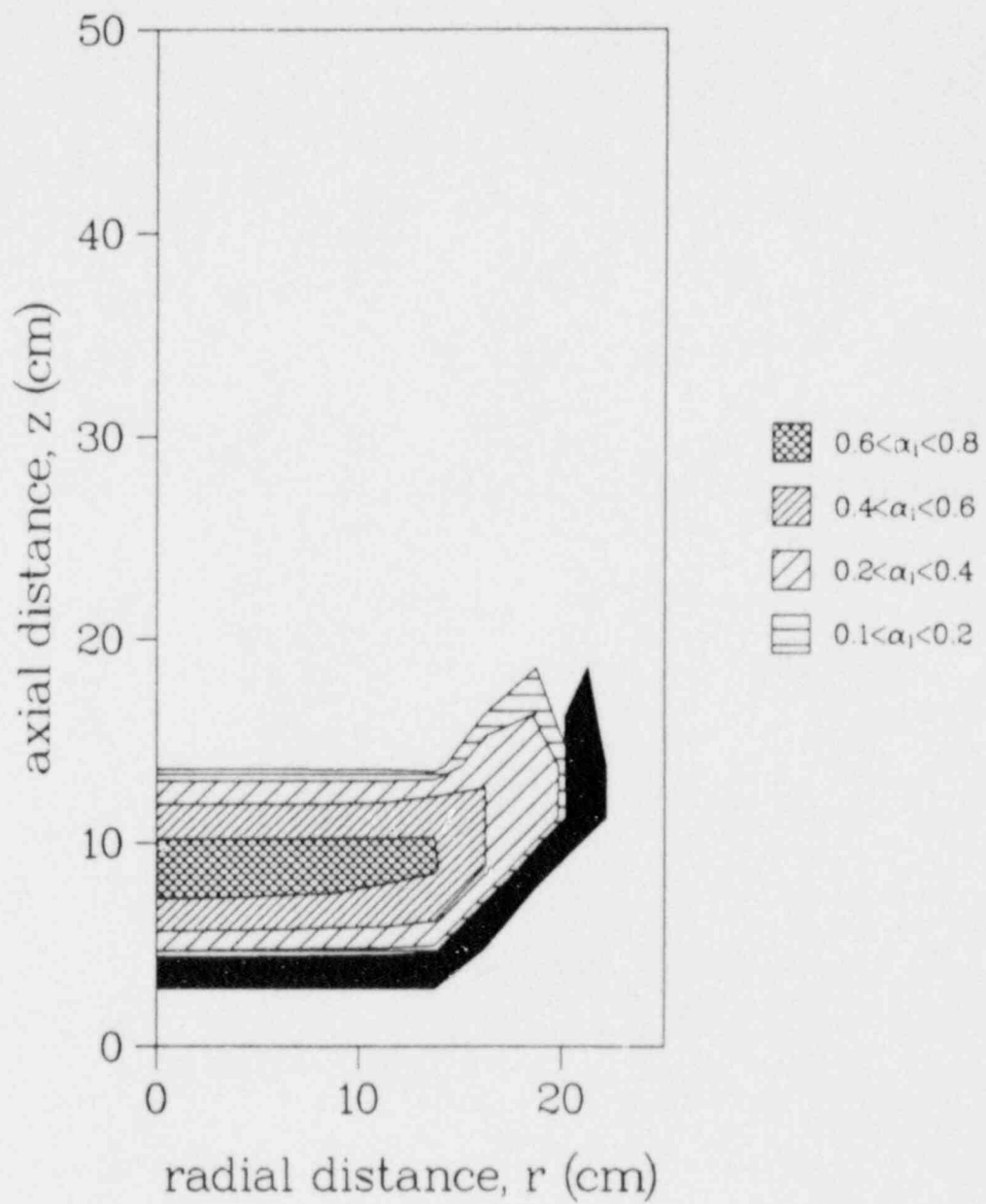


FIG. 16: Liquid volume fraction  $\alpha_1$  and the blockage region (in black) at  $t=4400$  s for the base case.

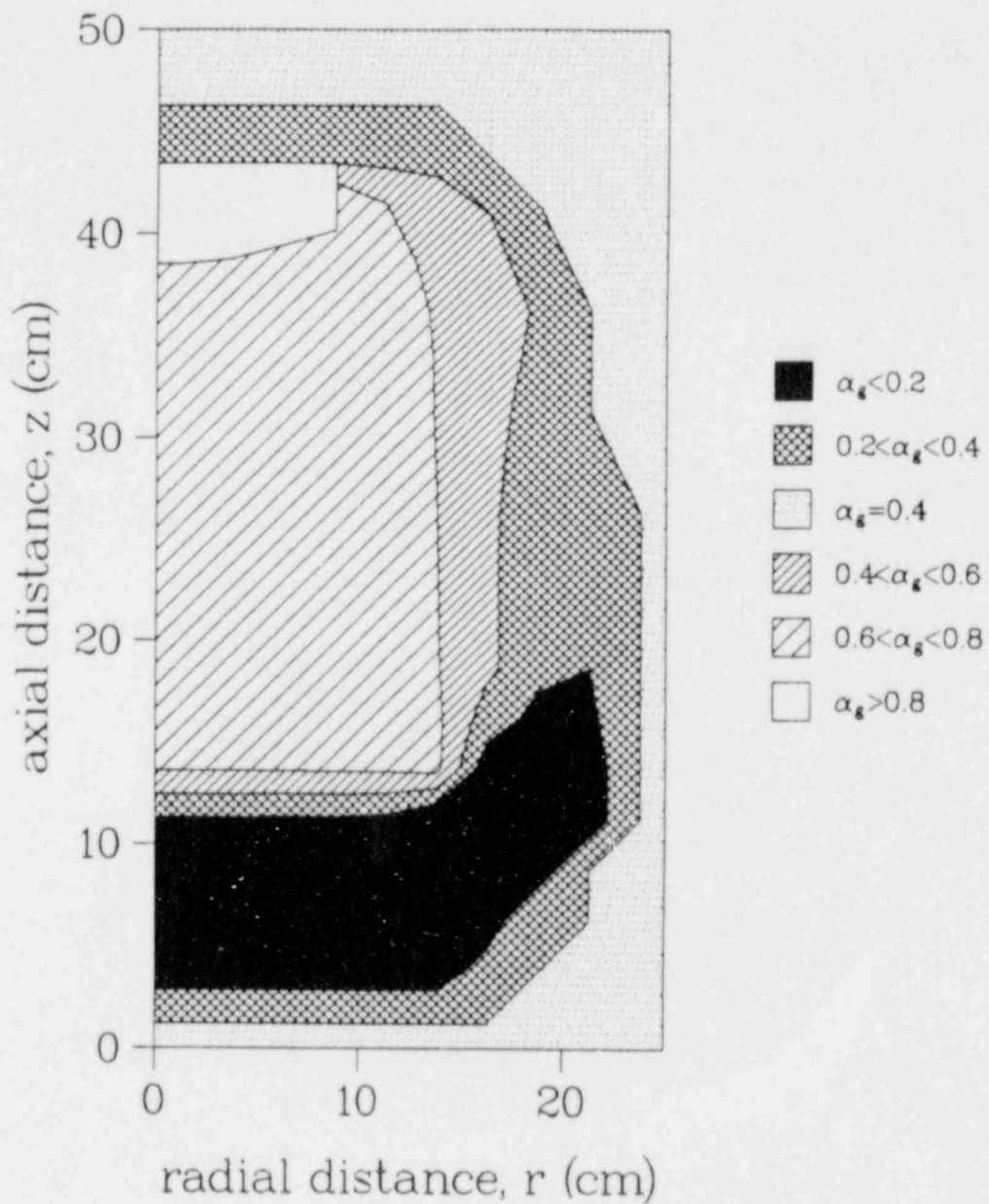


FIG. 17: Gas volume fraction  $\alpha_g$  in rubble bed at  $t=4400$  s for the base case. The rubble has an initial uniform porosity of 0.4.

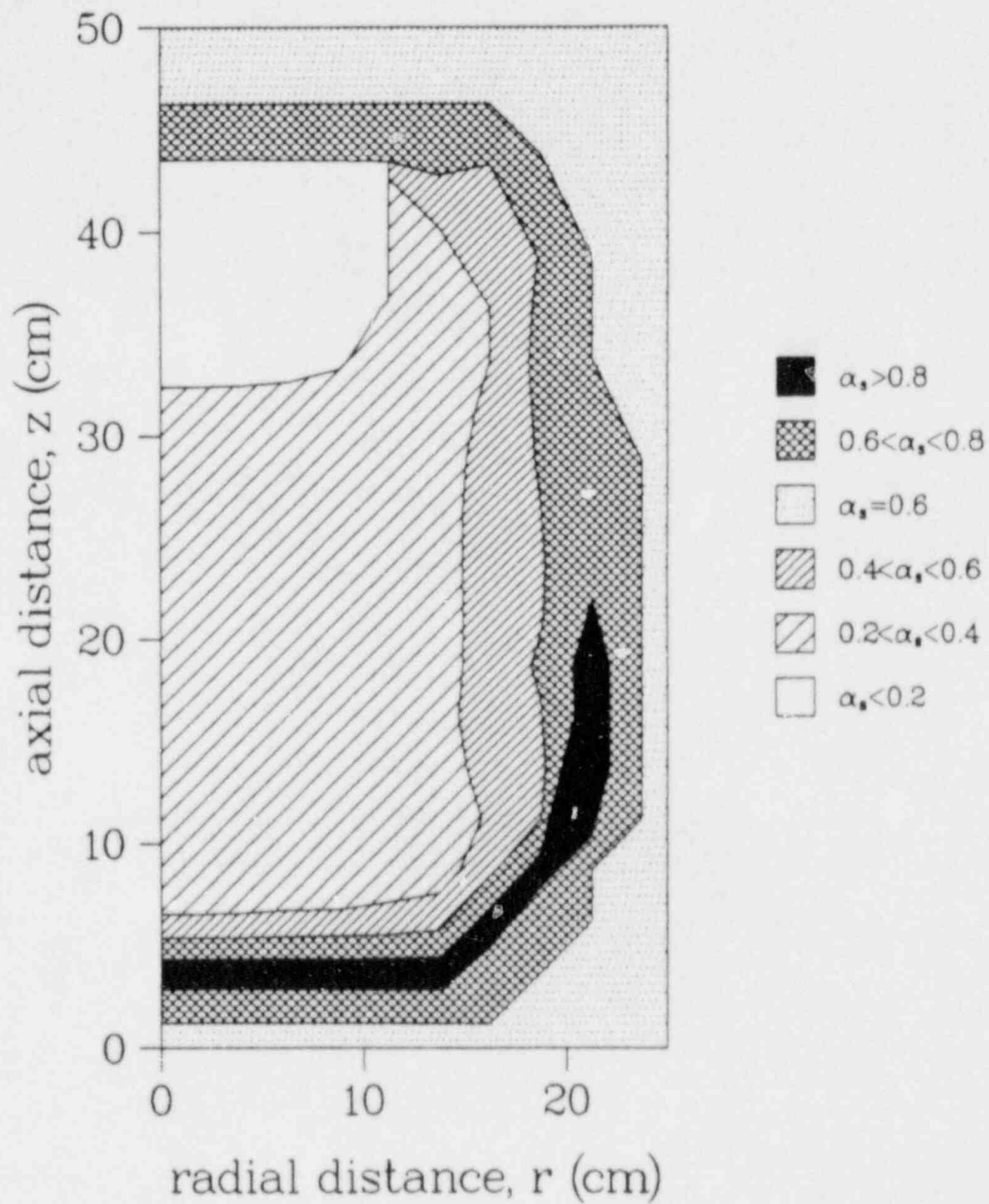


FIG. 18: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4600$  s for the base case. The rubble has an initial uniform solid volume fraction of 0.6.

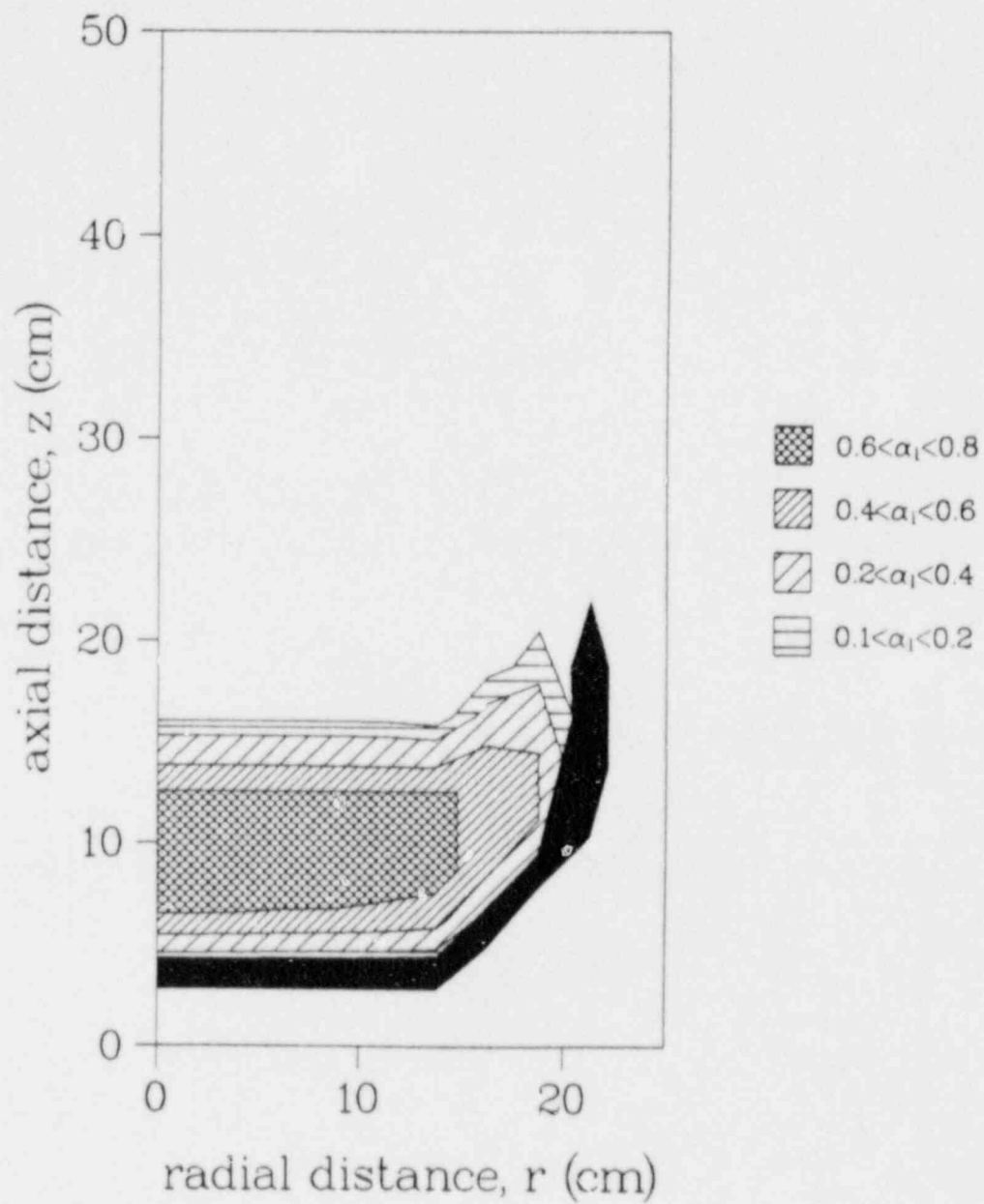


FIG. 19: Liquid volume fraction  $\alpha_1$  and the blockage region (in black) at  $t=4600$  s for the base case.

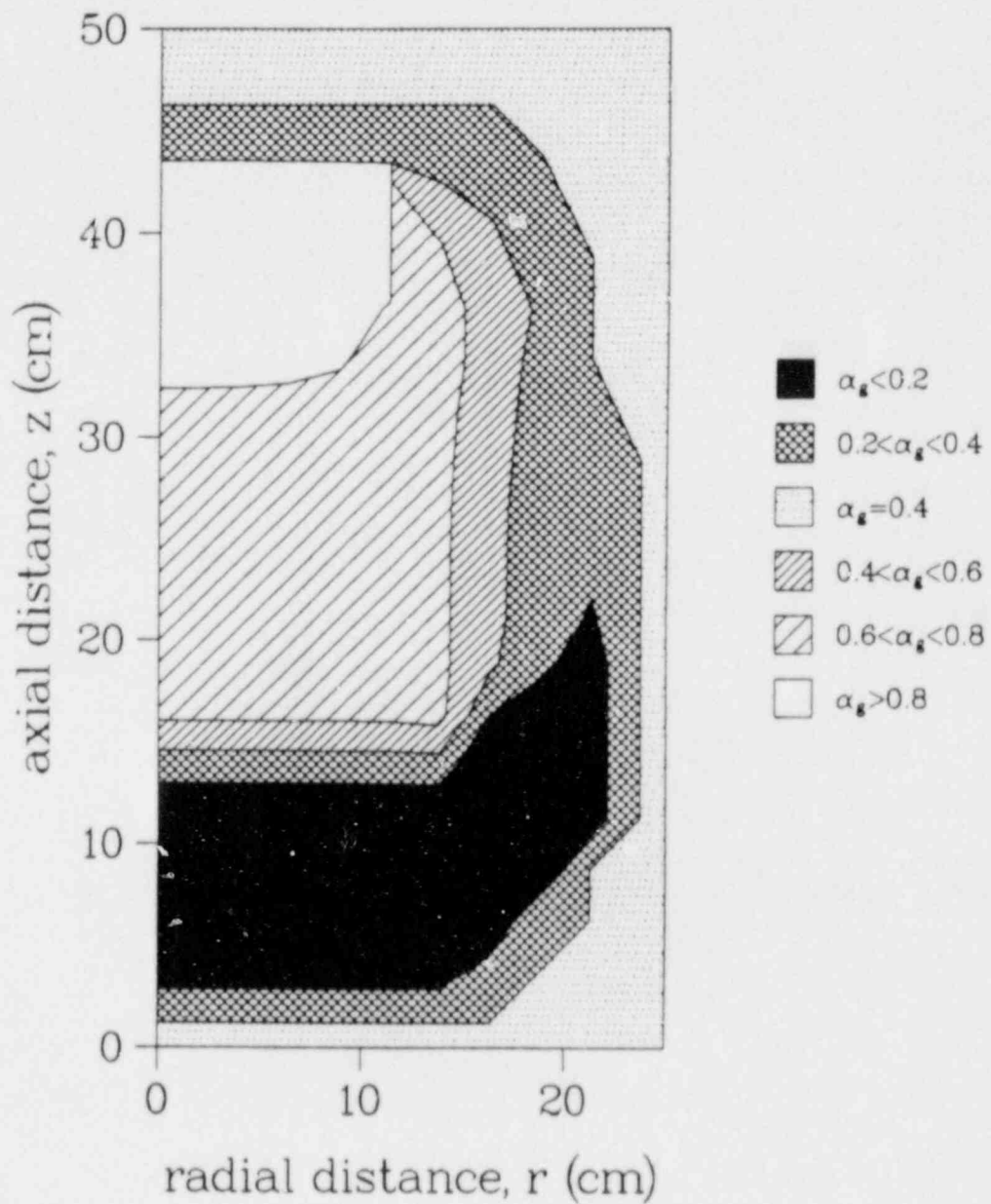


FIG. 20: Gas volume fraction  $\alpha_g$  in rubble bed at  $t=4600$  s for the base case. The rubble has an initial uniform porosity of 0.4.

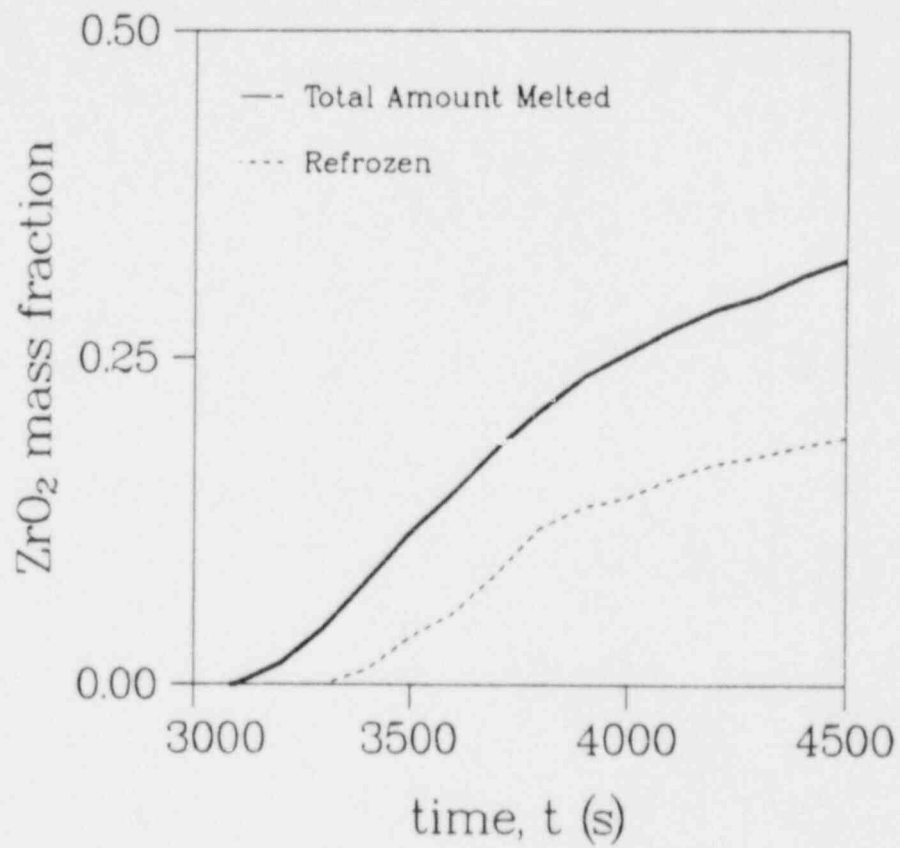


FIG. 21: Fraction of the ZrO<sub>2</sub> that has melted (solid line) and the fraction that has melted and refrozen (dashed line).

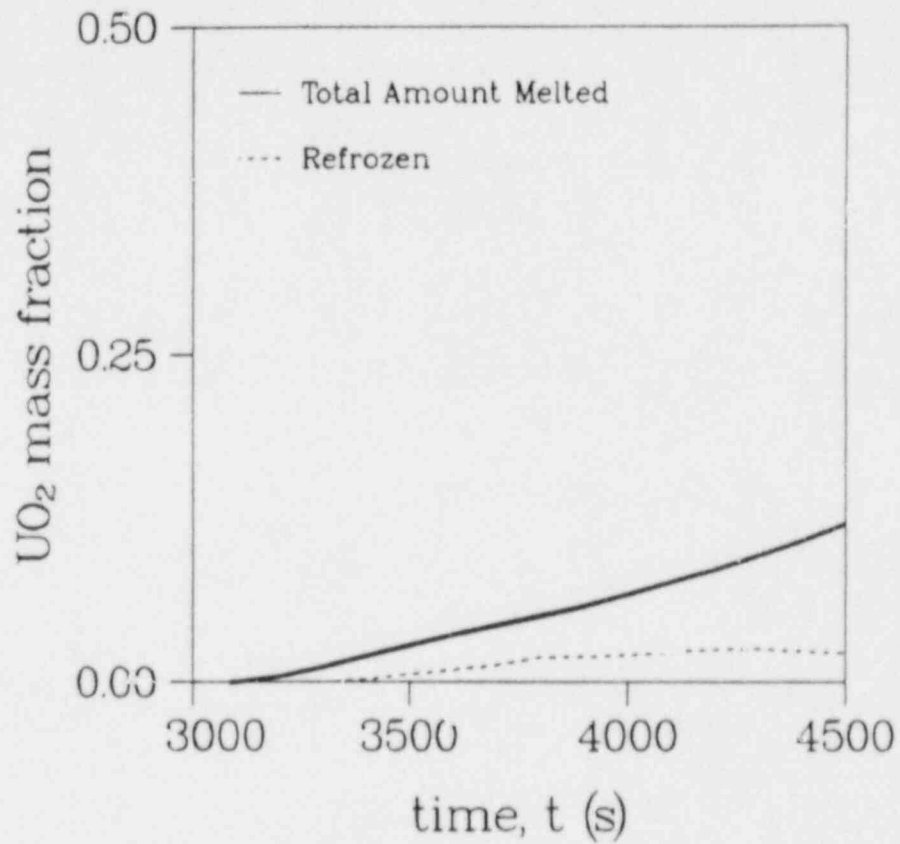


FIG. 22: Fraction of the UO<sub>2</sub> that has melted (solid line) and the fraction that has melted and refrozen (dashed line).



### 3.2 Effect of Varying the Particle Diameter

Solutions are presented in this section for a bed composed of 5 mm diameter particles. Parameters such as the initial and boundary conditions, the size of the bed and the composition, are kept fixed at their base case values. Solid volume fractions are shown every two hundred seconds from 3200 s to 4200 s in Figs. 23 through 28. Increasing  $d_p$  enhances radiation transfer through the bed, raising both the effective thermal conductivity and the thermal boundary layer thickness. Consequently, in beds composed of large particles, blockages form further from the boundaries, the solid starts melting later and the melt zone is smaller. These effects may have a bearing on the decision to add water to the reactor vessel during an accident if it is suspected that the rods might fragment into fine particulate when quenched.

Comparing Figs. 4 and 24 it is evident that the lower crust is initially thicker for a bed composed of 5 mm particles. In beds with large particles, melt relocation is faster (the permeability is proportional to  $d_p^2$ ) and liquid starts moving earlier because the critical residual saturation is lower ( $S_r \sim 1/d_p$ ). Liquid quickly flows out of the melt zone as it forms and refreezes in colder portions of the bed. Consequently, refreezing is much more vigorous in beds with large  $d_p$  and the initial crusts are larger.

Note that the capillary rise discussed in the previous section is less evident in Fig. 29 than in Figs. 12, 16 and 19. This is due to the fact that capillary forces are less important in beds with large particles (recall that the capillary pressure is inversely proportional to particle diameter).

Phase volume fraction profiles at 4400 s and at 4600 s are shown in Figs. 30-32 and Figs. 33-35, respectively. Debris starts settling before 4300 s elapse - see Figs. 36 and 37. In the bed with 1 mm diameter particles, a crust was created above the melt zone early in the meltdown sequence and a void formed above the melt zone as solid collapsed downward (see Fig. 11). Such an upper crust does not form in the bed with  $d_p=5$  mm because capillary forces are weaker.

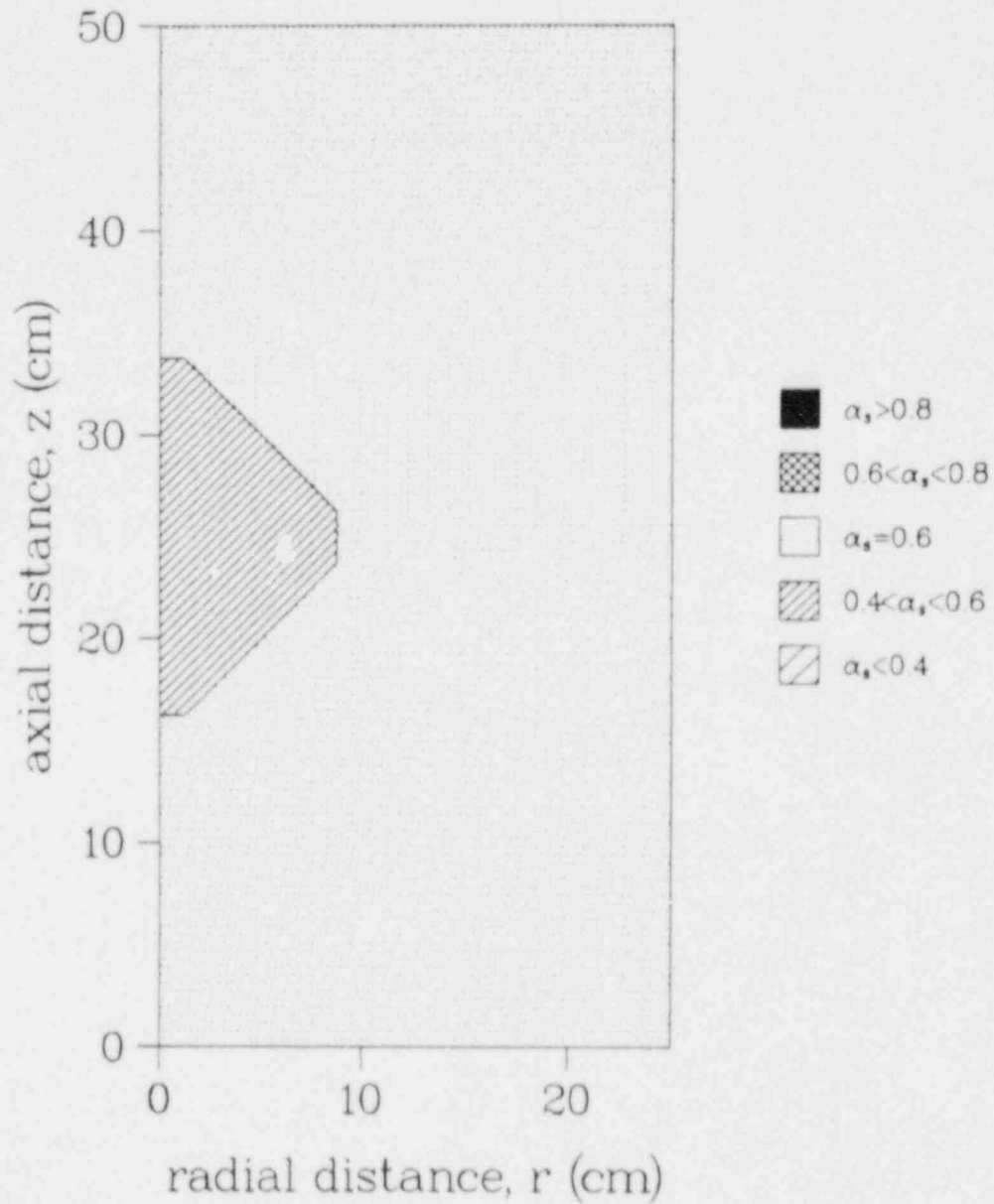


FIG. 23. Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3200$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

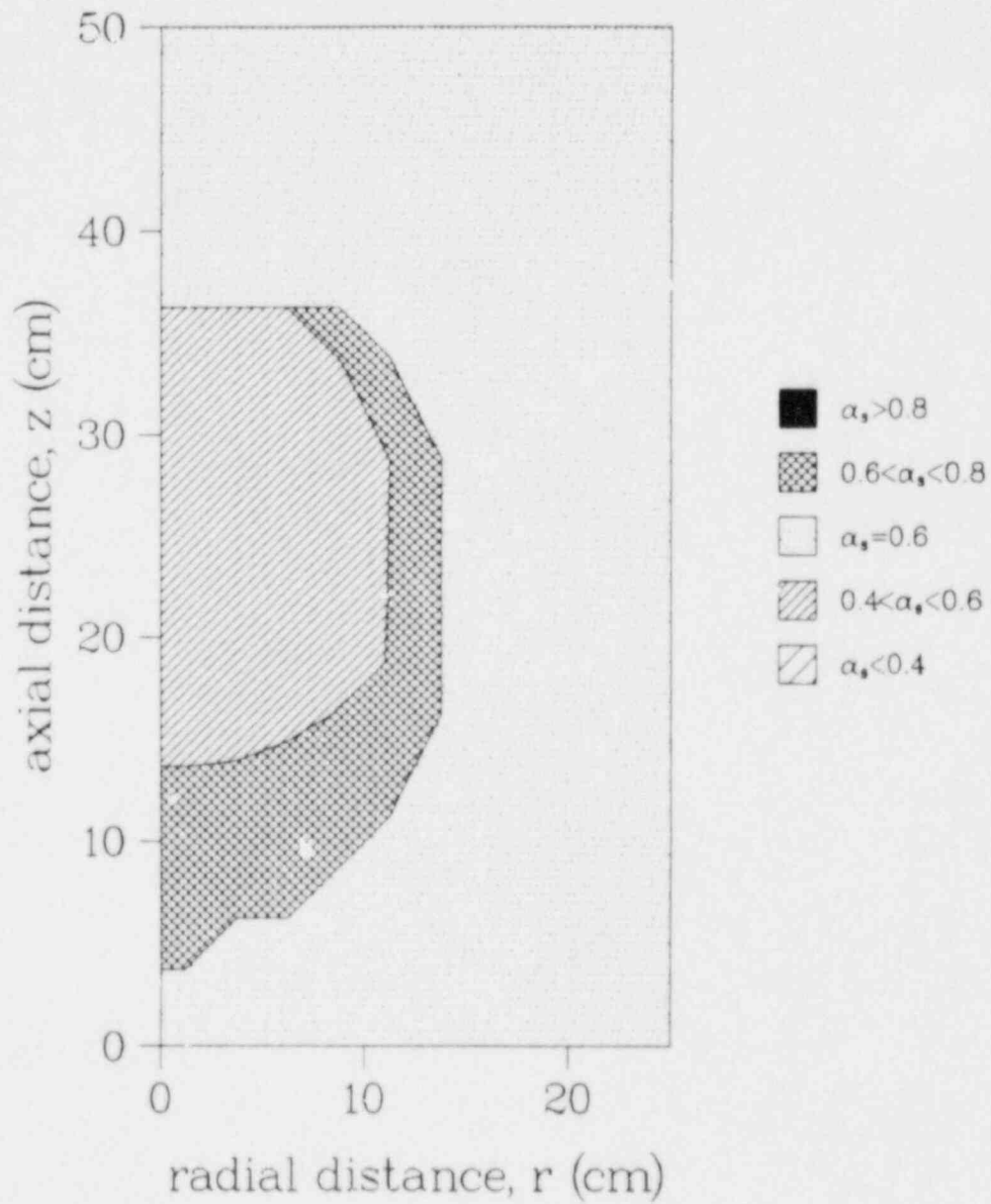


FIG 24: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3400$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

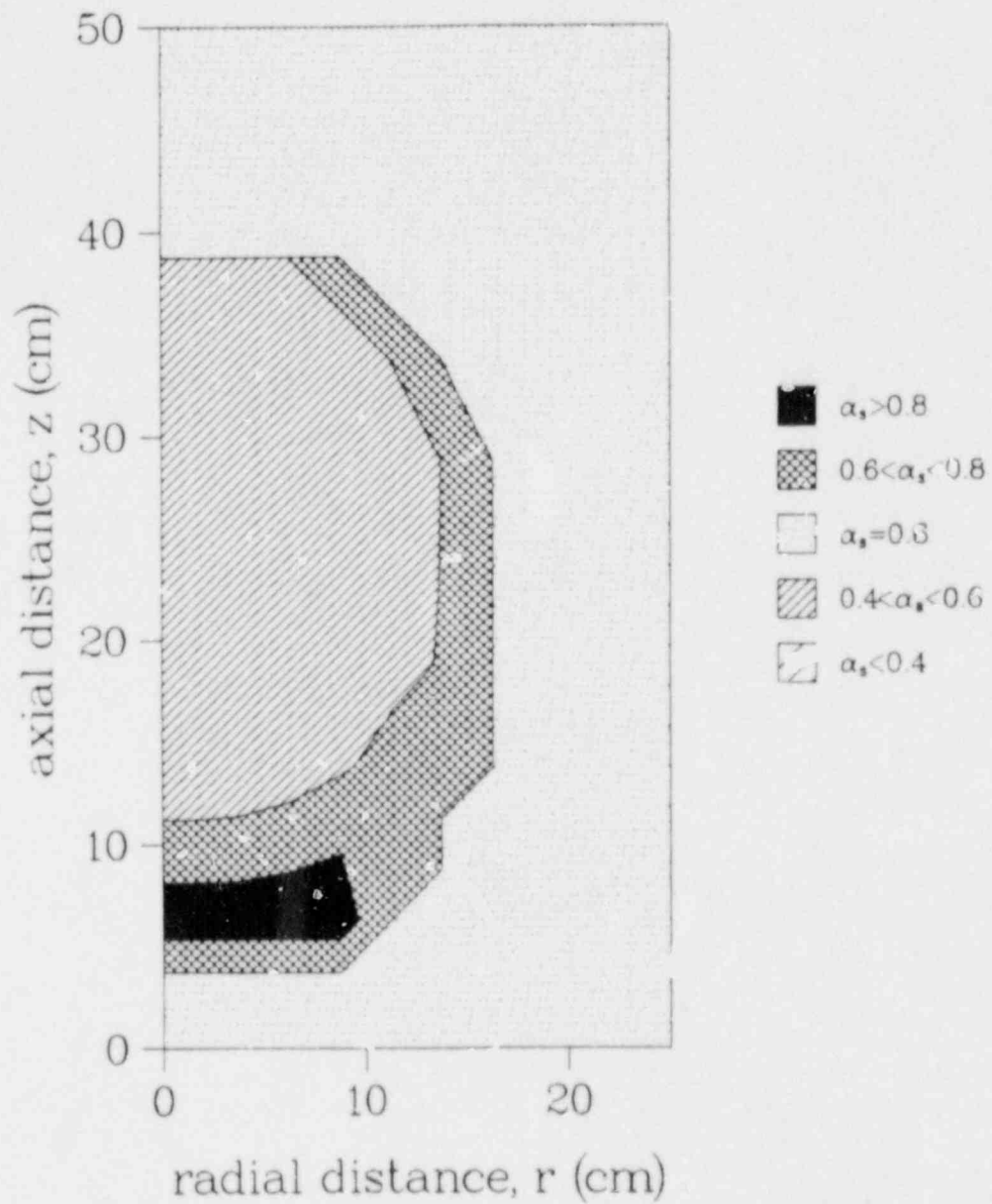


FIG. 25: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3600$  s for a bed with an average particle diameter of 5.1 mm. The rubble has an initial uniform solid volume fraction of 0.6.

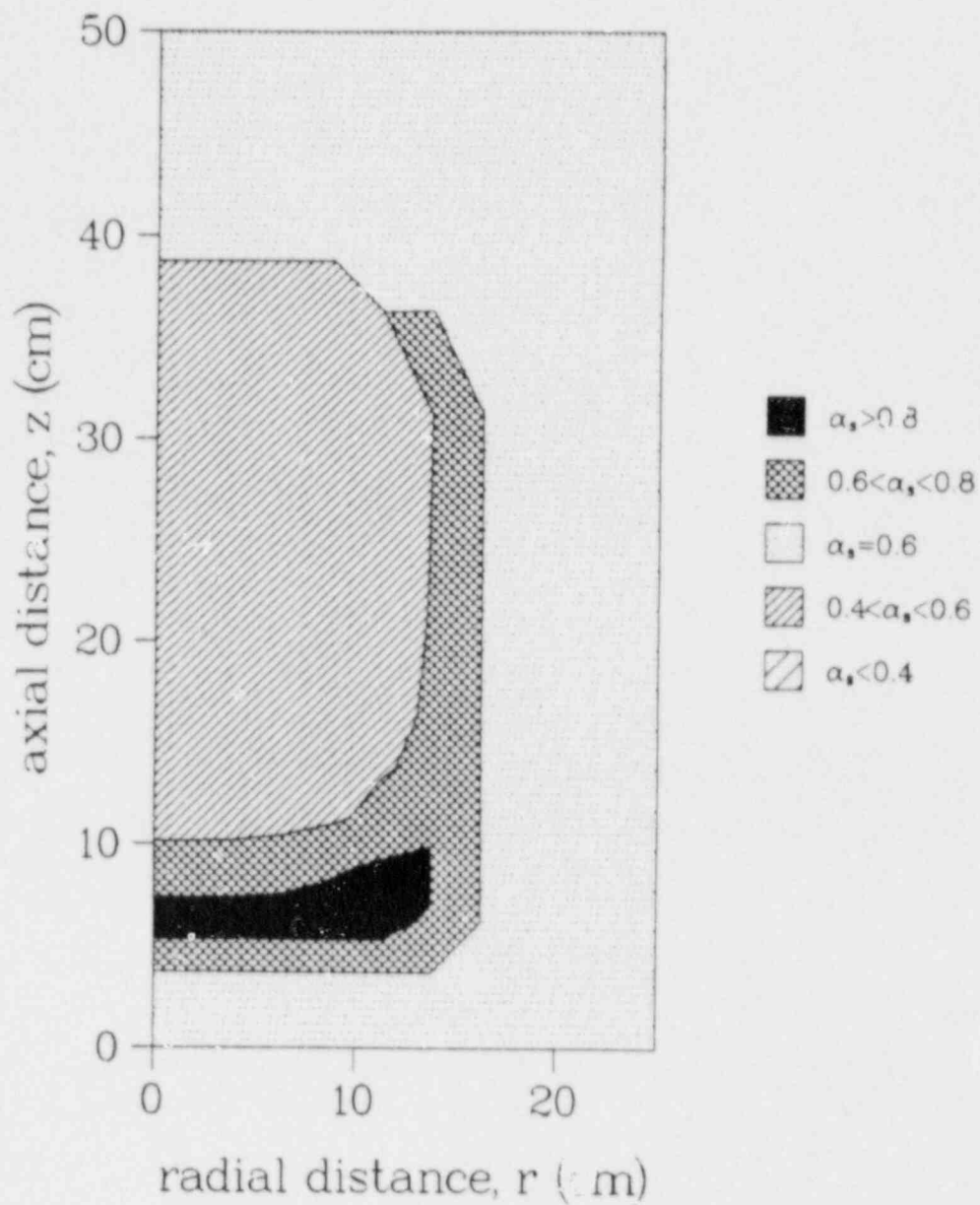


FIG. 26: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=3800$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

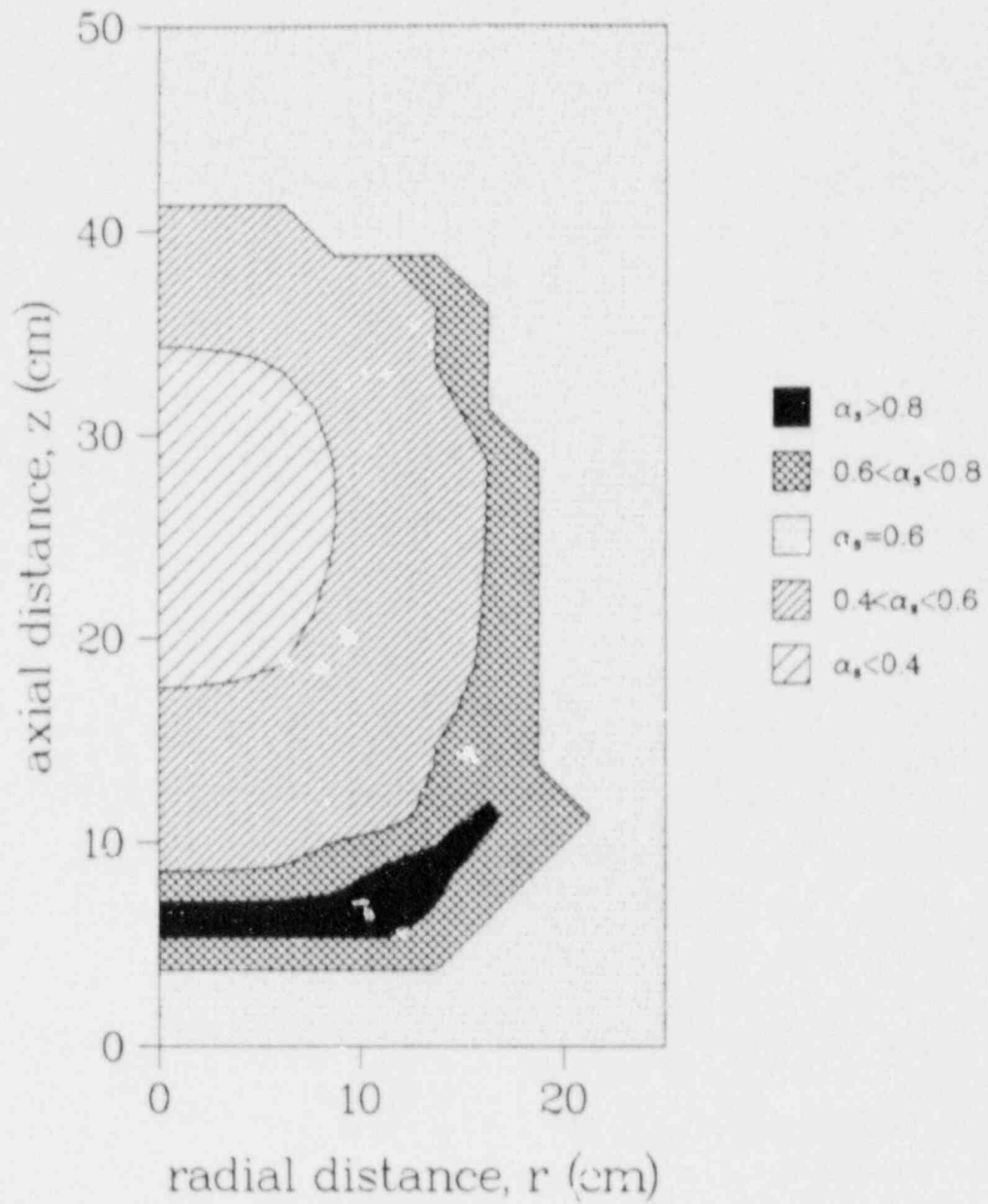


FIG 27: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4000$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

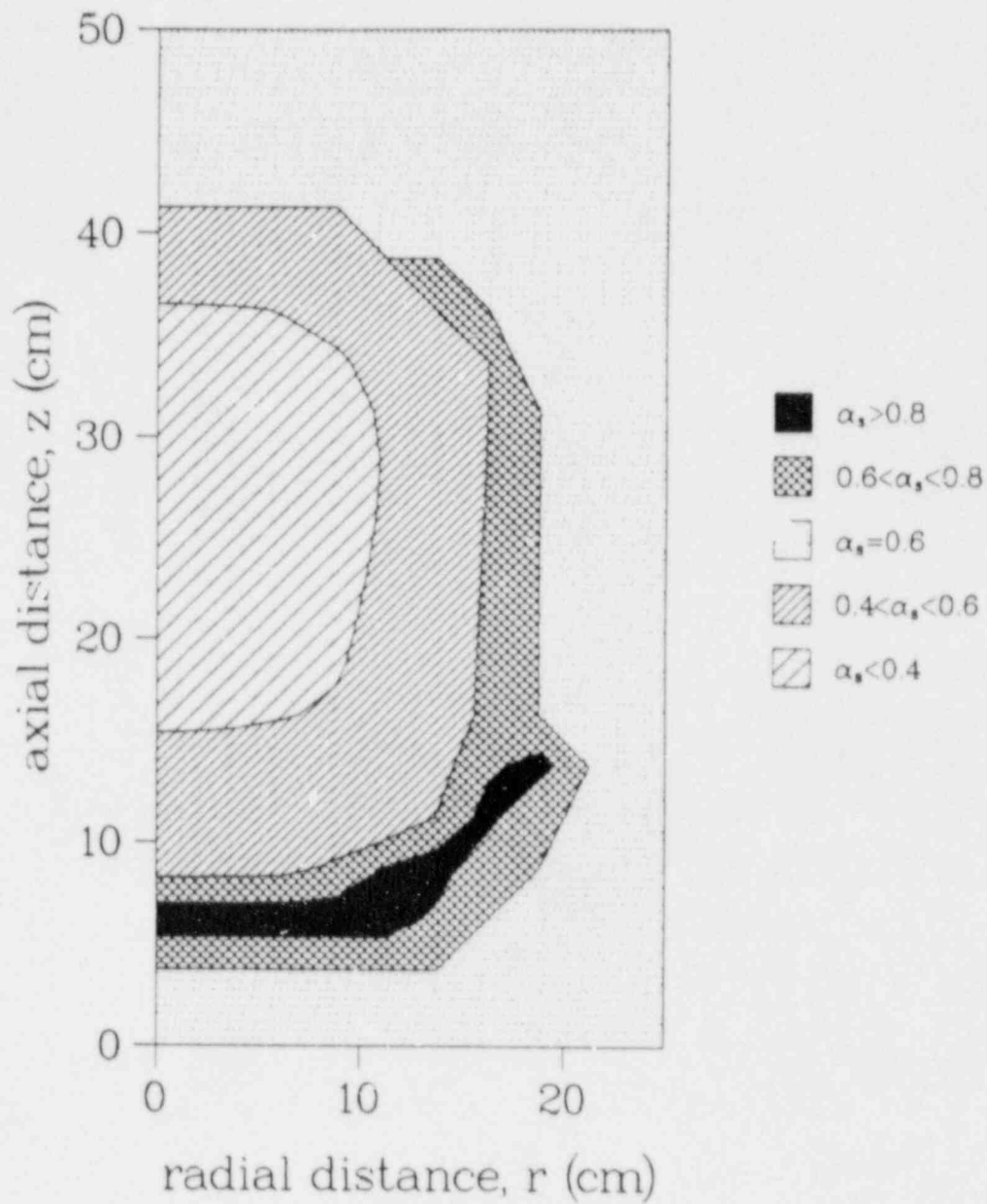


FIG. 28: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4200$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

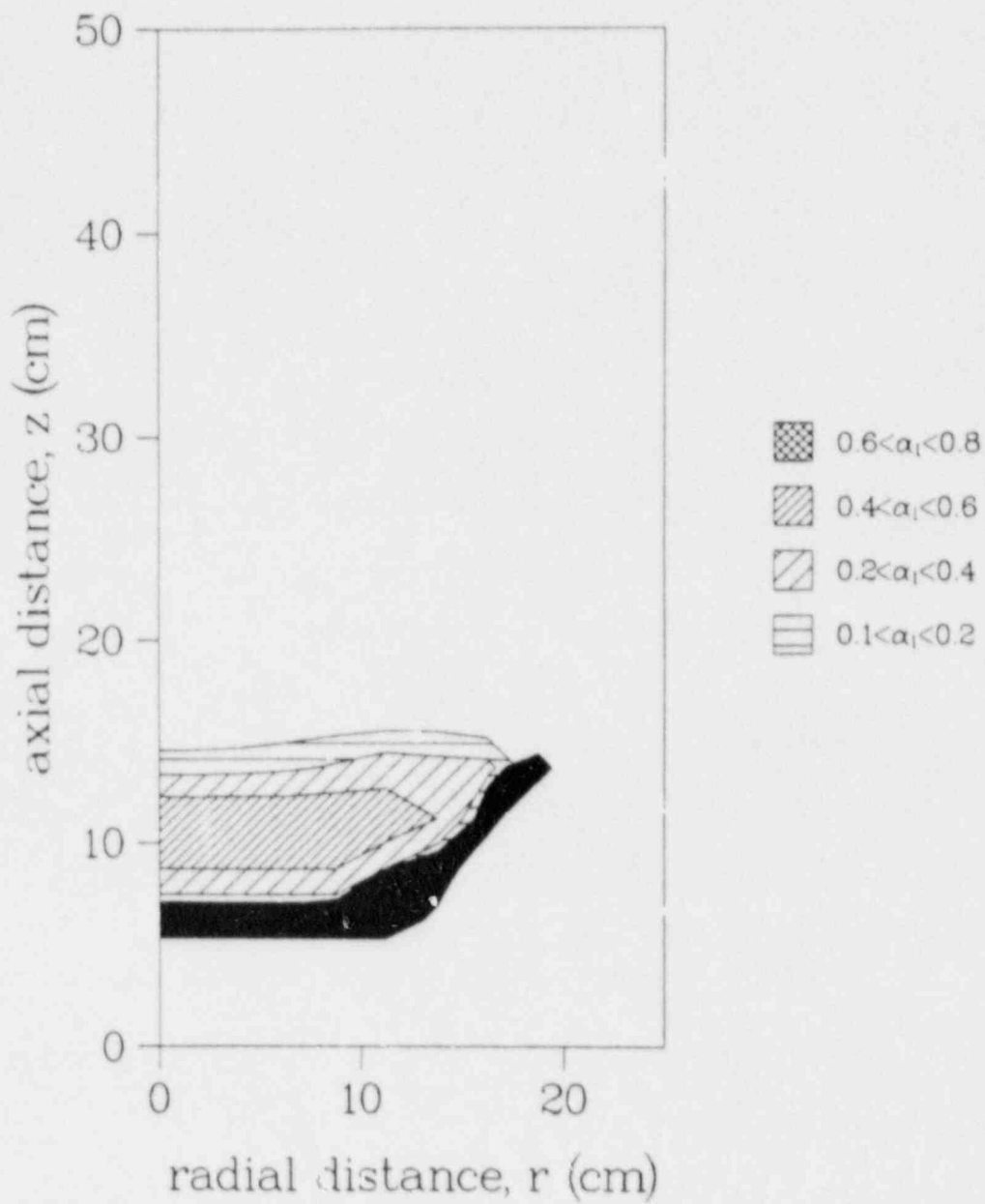


FIG. 29: Liquid volume fraction  $\alpha_l$  and the blockage region (in black) at  $t=4200$  s for a bed with 5 mm diameter particles.



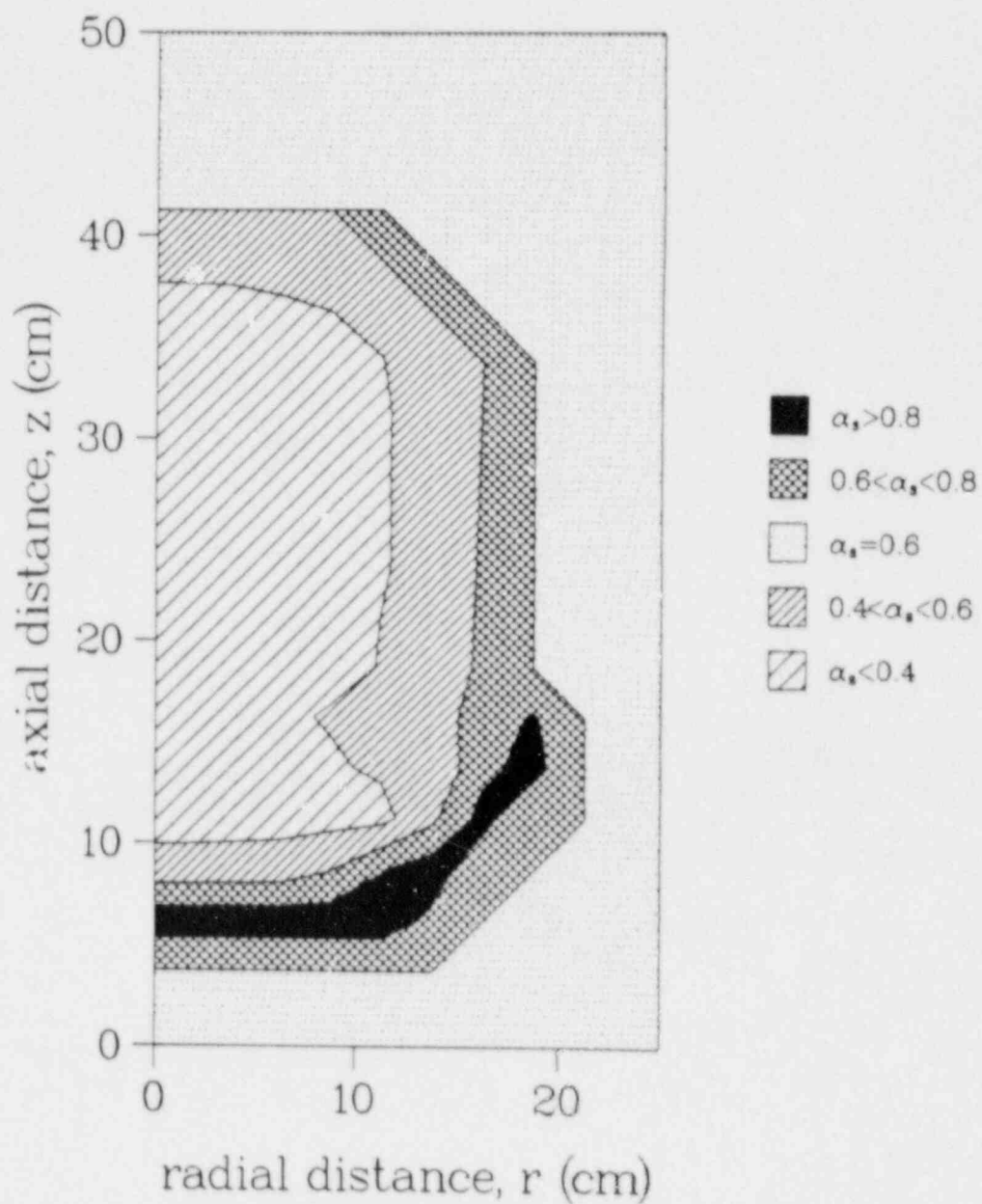


FIG. 30: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4400$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

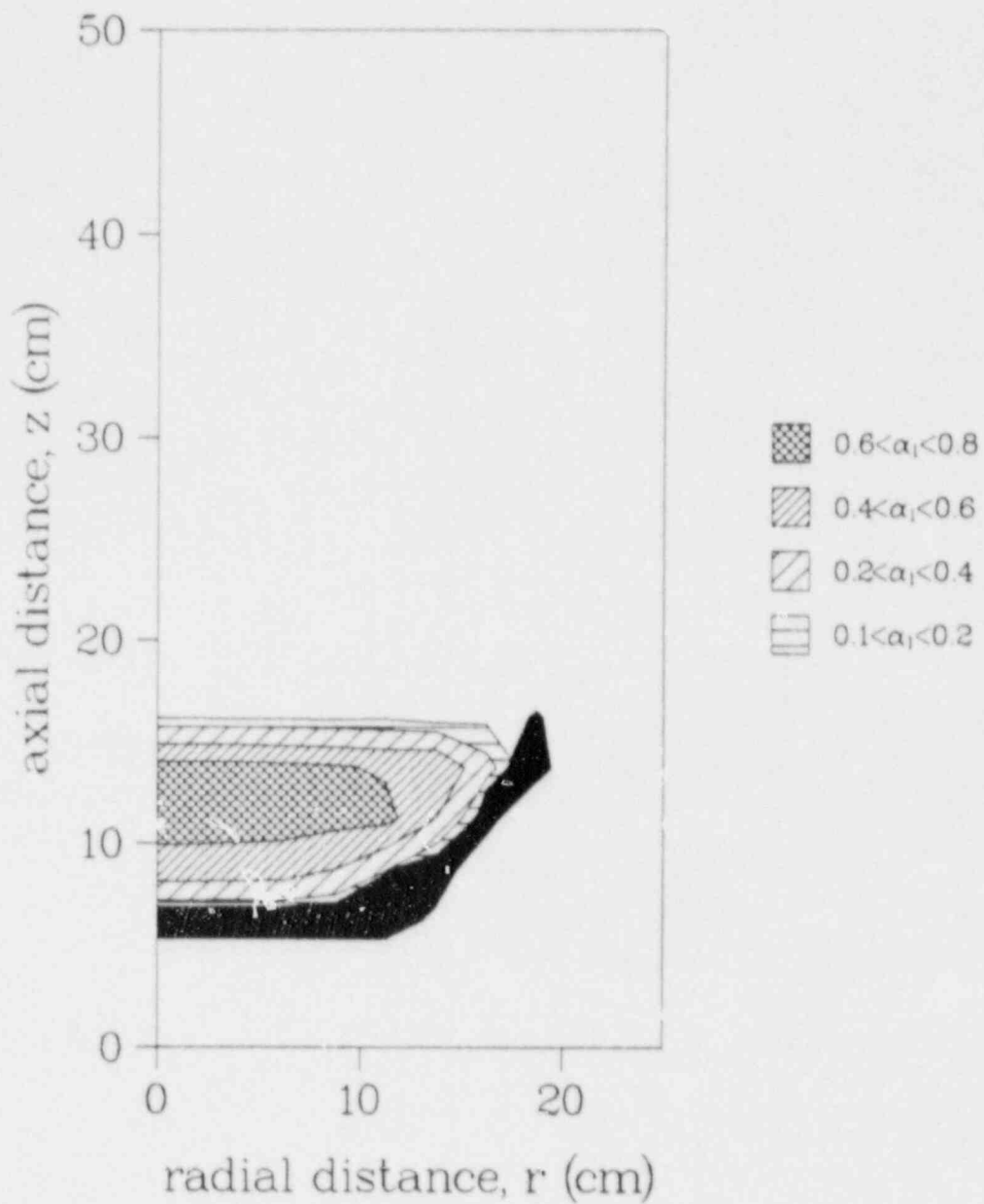


FIG 31 Liquid volume fraction  $\alpha_l$  and the blockage region (in black) at  $t=4400$  s for a bed with 5 mm diameter particles.

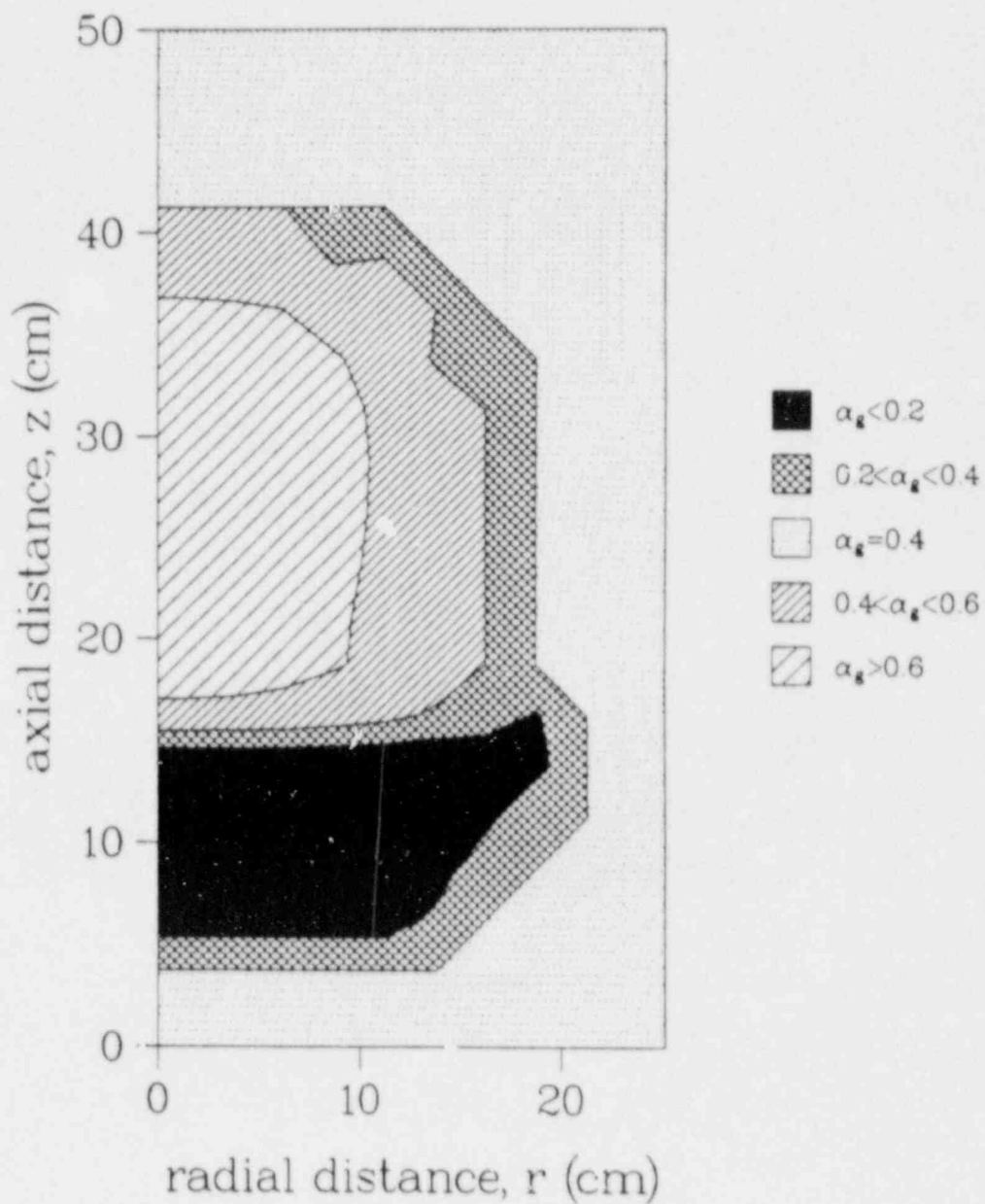


FIG. 32: Gas volume fraction  $\alpha_g$  in the rubble bed at  $t=4400$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform porosity of 0.4.

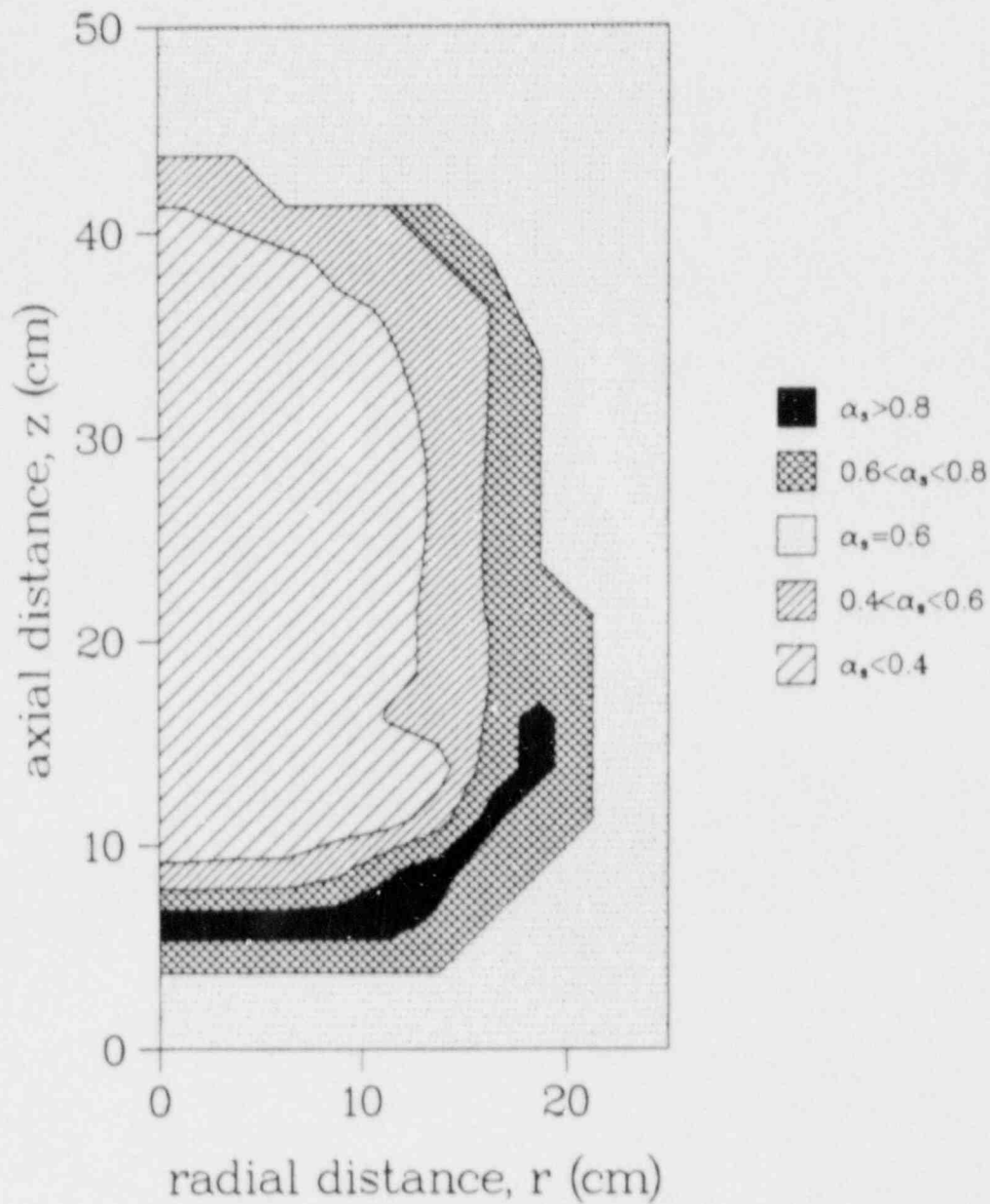


FIG 33: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4600$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.

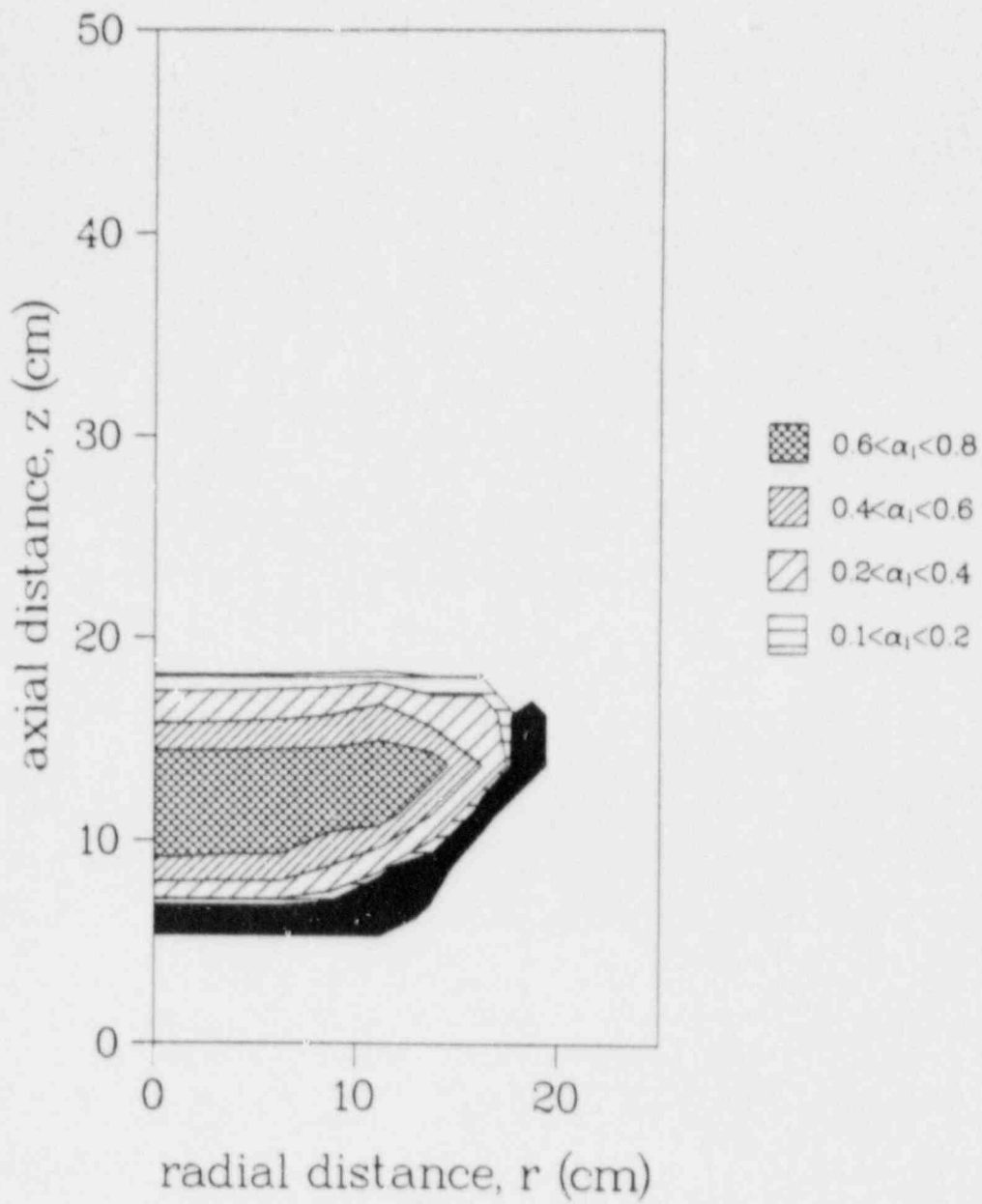


FIG. 34: Liquid volume fraction  $\alpha_l$  and the blockage region (in black) at  $t=4600$  s for a bed with 5 mm diameter particles.

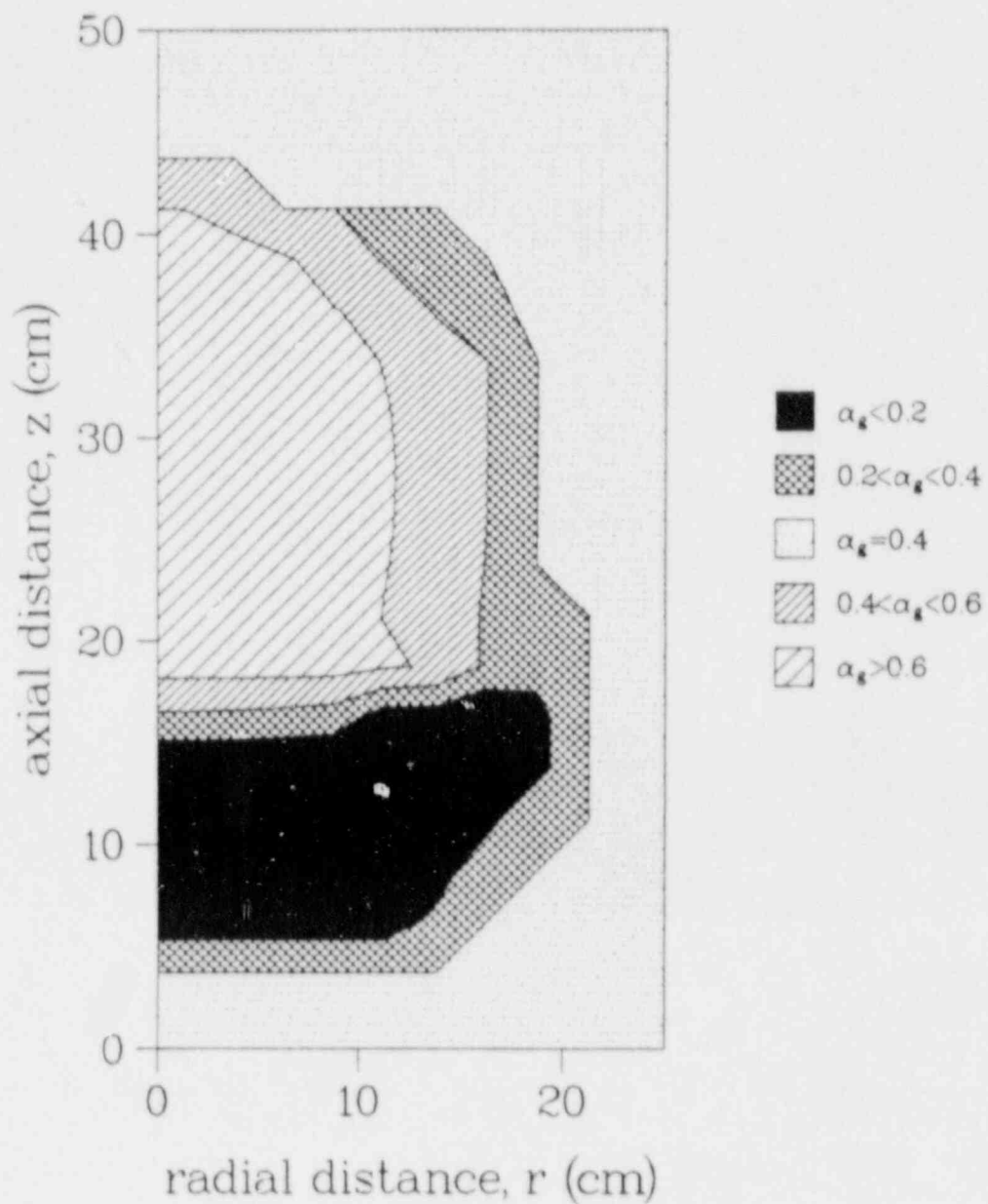


FIG. 35: Gas volume fraction  $\alpha_g$  in the rubble bed at  $t=4600$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform porosity of 0.4.

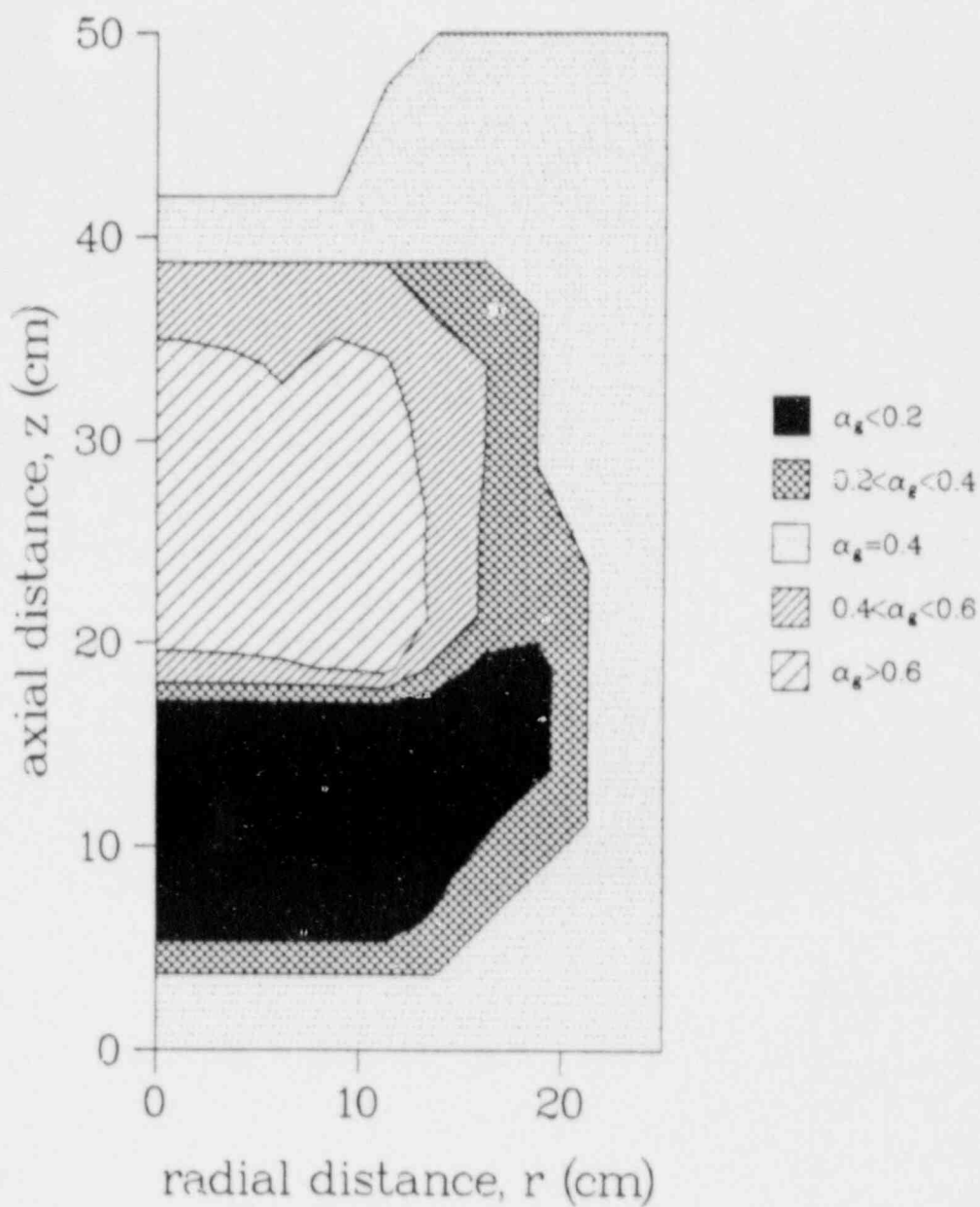


FIG. 36: Gas volume fraction  $\alpha_g$  in the rubble bed at  $t=4800$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform porosity of 0.4.

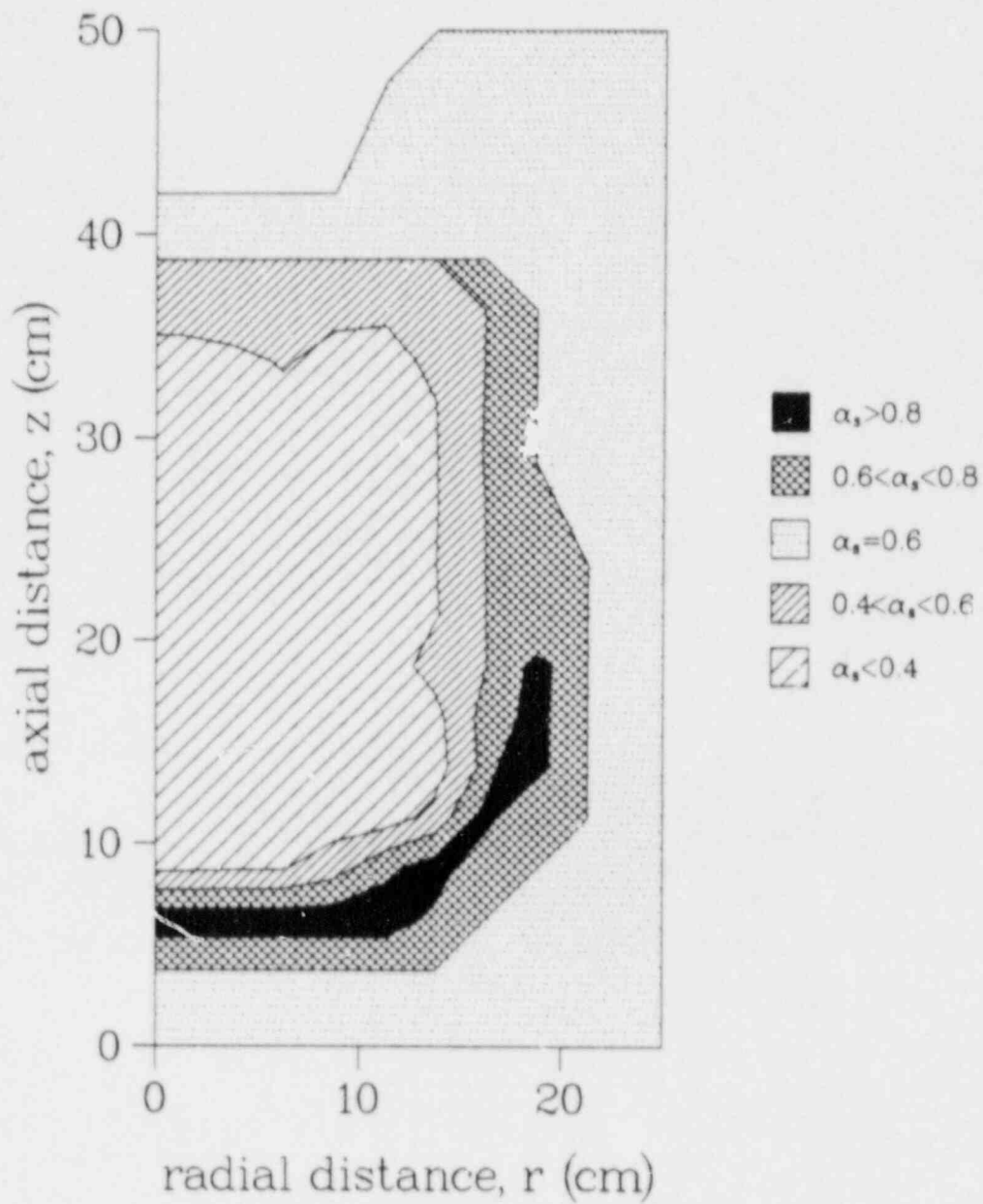


FIG. 37: Solid volume fraction  $\alpha_s$  in rubble bed at  $t=4800$  s for a bed with an average particle diameter of 5 mm. The rubble has an initial uniform solid volume fraction of 0.6.



## 4. CONCLUSIONS

### 4.1 Summary

A model of melt formation and relocation in a two-dimensional core rubble bed has been developed. The analysis included: mass conservation equations for the species of interest ( $UO_2$  and  $ZrO_2$ ); a momentum equation ( $z,r$ ) which represents a balance among drag, capillary and gravity forces; and an energy equation which incorporated the effects of convection by the melt, radiation and conduction through the bed and internal heat generation. An equilibrium  $UO_2$ - $ZrO_2$  phase diagram was prescribed and radiative heat transfer through the bed was incorporated utilizing a temperature dependent conductivity.

A typical solution was presented and the effect of varying the average particle diameter  $d_p$  was discussed. Varying  $d_p$  had important effects on melting, liquid relocation and crust formation. Increasing  $d_p$  resulted in higher effective thermal conductivities, delaying melt formation and decreasing the size of the melt zone. Melt relocation was faster, liquid flowed earlier and refreezing was more vigorous in the bed with large particles.

Materials such as zirconium and stainless steel are currently being incorporated into the present model. Models developed in this study will be implemented in the U.S. Nuclear Regulatory Commission's MELPROG computer code [48,49] and the modified version of the code will be used to analyze the accident at Three Mile Island in considerable detail.

### 4.2 Major Uncertainties

Experiments are needed both to validate the current core rubble model and to help guide future theoretical work. Major subjects of interest are (1) chemical interactions; (2) flow correlations; and (3) collapsing of high porosity particulate beds. A brief discussion of these issues follows.

Experiments using prototypical materials are needed to test the chemistry models in the analysis. In particular, the use of equilibrium phase diagrams must be assessed. Nonequilibrium behavior can result from both finite reaction rates and finite diffusion rates - the species of interest ( $UO_2, ZrO_2$ ) are initially separated in core rubble beds. If the equilibrium assumption is adequate, a U-Zr-O phase diagram that represents a minimization of the Gibb's free energy is essential. Especially important is the accurate determination of heats of fusion and dissolution energies for the compounds of interest. Such information will affect predictions of current models. For instance, a recent study indicates that the dissolution of  $UO_2$  by molten Zr is an exothermic process [73], with a heat release on the order of twice the heat of fusion of Zr. Because the current version of the DEBRIS module of MELPROG assumes no heat of dissolution for this reaction, it underpredicts the amount of  $UO_2$  dissolved. Numerical information regarding interactions of U and Zr with Fe, stainless steel, etc., will also be needed.

Melt relocation models in the analysis also need to be validated. Because melt relocation and crust formation is sensitive to flow correlations such as relative permeabilities, the residual saturation and the Leverett function, uncertainties in these correlations can lead to uncertainties in the timing of core slump. To date, none of these correlations have been tested in porous materials undergoing phase changes.

Another important subject of interest is bed collapse. As solid melts and flows downward, the solid density in the center of the bed decreases. Of interest here is the collapse of such high porosity particulate beds. Past experiments indicate that such beds can be self-supporting at fairly low densities. Two explanations have been proposed: (1) liquid trapped between particles by surface tension effects keep the particles in place as they melt; and (2) solid particles sinter as the bed temperature increases (mass diffusion rates increase rapidly with T). Experiments are needed to determine if bed collapse can be represented using a critical porosity. If such a model is adequate, this critical porosity must be determined as a function of particle diameter.

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