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WHC-EP-0713

Ferrocyanide Safety Program: Waste Tank Sludge Rheology Within a Hot Spot or During Draining

Homogeneous Flow Versus Flow Through a Porous Medium

Prepared for the U.S. Department of Energy
Office of Environmental Restoration
and Waste Management

Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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Date Published
November 1993

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Date Received: 12/8/93 KMB	INFORMATION RELEASE REQUEST	Reference: WHC-CM-3-4

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ID Number (include revision, volume, etc.): WHC-EP-0713

List attachments.

Date Release Required: 11/24/93

Title: Ferrocyanide Safety Program: Waste Tank Sludge Rheology Within a Hot Spot or During Draining

Unclassified Category: UC-600

Impact Level: 4

New or novel (patentable) subject matter? [X] No [] Yes
If "Yes", has disclosure been submitted by WHC or other company?
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Will material be handed out? [] Yes [] No

Title of Journal

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Applied Technology/Export Controlled Information or International Program	[]	[X]			
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Communications	[X]	[]	W. P. Whiting	WPWhiting	11-30-93
RL Program/Project	[X]	[]	R. F. Christensen	RF Christensen	12/2/93
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Other Program/Project	[]	[]			

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References Available to Intended Audience	[X]	[]
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Date Cancelled

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BD-7600-062 (08/91) WEF074

Part 1

ABSTRACT

The conditions under which ferrocyanide waste sludge flows as a homogeneous non-Newtonian two-phase (solid precipitate-liquid) mixture rather than as a liquid through a porous medium (of stationary precipitate) are examined theoretically, based on the notion that the preferred rheological behavior of the sludge is the one which imposes the least resistance to the sludge flow. The homogeneous two-phase mixture is modeled as a power-law fluid and simple criteria are derived that show that the homogeneous power-law sludge-flow is a much more likely flow situation than the porous medium model of sludge flow. The implication of this finding is that the formation of a hot spot or the drainage of sludge from a waste tank are not likely to result in the uncovering (drying) and subsequent potential overheating of the reactive-solid component of the sludge.

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1.0 INTRODUCTION

Some of the Hanford Site high-level nuclear waste storage tanks contain solid (precipitate) ferrocyanide compounds as components of an aqueous sludge. The major safety issue associated with these tanks is the possibility of the existence or formation of local hot spot(s) resulting in dry precipitate that can self-heat, due to the concentration of radioactive Cs and Sr isotopes. This may lead to uncontrolled runaway reaction and consequent possibility of radioactive material release. It is important to note at the outset that this issue stems from the notion that the waste sludge behaves as a porous medium rather than as a slurry of fine particulate. In a slurry the liquid and particulate phases move together in a homogeneous manner, i.e., the velocities of the two phase are approximately equal. Obviously the separation of the liquid phase from the solid phase within a slurry is difficult, if not impossible. However, in the porous medium model of the sludge the precipitate particles are assumed to be stationary and liquid (or vapor) flows in the "void space" between particles. Accordingly, the uncovering of the solid phase is readily visualized and two mechanisms that may result in the overheating of the precipitate have been identified.

The first mechanism is based on the formation of a small but finite size region in the waste tank sludge that is highly concentrated in heat-generating ferrocyanide compounds. In discussing this so-called "hot-spot problem" it should be recognized that it is, at present, an imaginary waste configuration in that no means of calculating or even explaining the formation of a hot spot from physical principles has yet been suggested. If the heat generating material is sufficiently concentrated, the interstitial liquid will reach its boiling point. Once this occurs an evaporation front, separating a vapor region (or bubble) from the surrounding liquid will propagate into the porous medium. This drying process will be followed by the overheating of the stationary solid precipitate. Note that the heat generation rate required to bring the porous medium sludge to the boiling temperature of the interstitial liquid is usually much larger than the critical dryout heat flux. This is because the precipitate particles are very small (< 2 μm) and, according, the predicted permeability of the postulated porous medium sludge is very low (κ - $10^{-14} m^2$). Thus as soon as the boiling point of the liquid component is achieved, drying of the hot spot begins.

The second mechanism that may lead to drying of the precipitate is the drainage of the liquid from the porous-medium ferrocyanide sludge. The drainage may occur as a result of a leaking tank wall.

The main goal of the work reported here is to quantitatively examine the validity of the porous medium model of the waste tank sludge. To accomplish this we consider an alternative and equally valid model of the sludge rheology, namely non-Newtonian, homogeneous particle-liquid flow. It seems reasonable that the expected rheological behavior of the sludge is the one which imposes the least resistance to the motion of the sludge, driven by either a hot spot vapor bubble or gravity in the case of drainage. This flow resistance criterion enables rational predictions of the conditions under which interstitial liquid flow through a stationary medium of solid ferrocyanide is possible. We begin with a discussion of the importance of sludge rheology with respect to hot spot thermal stability.

2.0 FLOW RHEOLOGY AND HOT SPOT THERMAL STABILITY

As mentioned in the Introduction, it seems that the fate of a hypothetical, thermally unstable hot spot within a waste-tank sludge depends on the hydrodynamic response of this two-phase, liquid-solid particle mixture to vapor bubble nucleation and subsequent bubble growth at the center of the overheated hot spot. In the past the sludge has been viewed as a porous medium (see, e.g. Wong, 1992). In this case the liquid component of the sludge is displaced by the vapor bubble and flows radially outward through a stationary medium of sludge particulate. The final state of the hot spot is a voided (dry) and, perhaps uncoolable region of heat generating, solid waste material (see Fig. 1a). The rheological equation of the sludge is, then, Darcy's law for liquid flow through a rigid porous medium. On the other hand, the rheological equation of the sludge may be quite different from that of Darcy. The sludge may behave externally as a pure liquid in that both the liquid and particle components move together as a homogeneous two-phase flow. The bubble interior now contains only vapor material. The original spherical hot spot is ultimately converted to a thin, thermally stable annular region at the termination of the bubble growth process (see Fig. 1b). By comparing Fig. 1a with Fig. 1b we conclude that, in terms of hot spot thermal stability, a homogeneous convecting sludge mixture is a much more favorable flow situation (Fig. 1b) than the Darcy law model of the sludge (Fig. 1a). The expected rheological sludge behavior is the one which imposes the least resistance to the growth of the expanding vapor cavity.

In the following, we examine the momentum equation for the early stages of bubble growth in a porous medium sludge and in a homogeneous, non-Newtonian sludge. In a subsequent section the bubble growth rates for these two different rheological models of sludge behavior are then compared with one another to determine the likely rheology and final hot spot configuration.

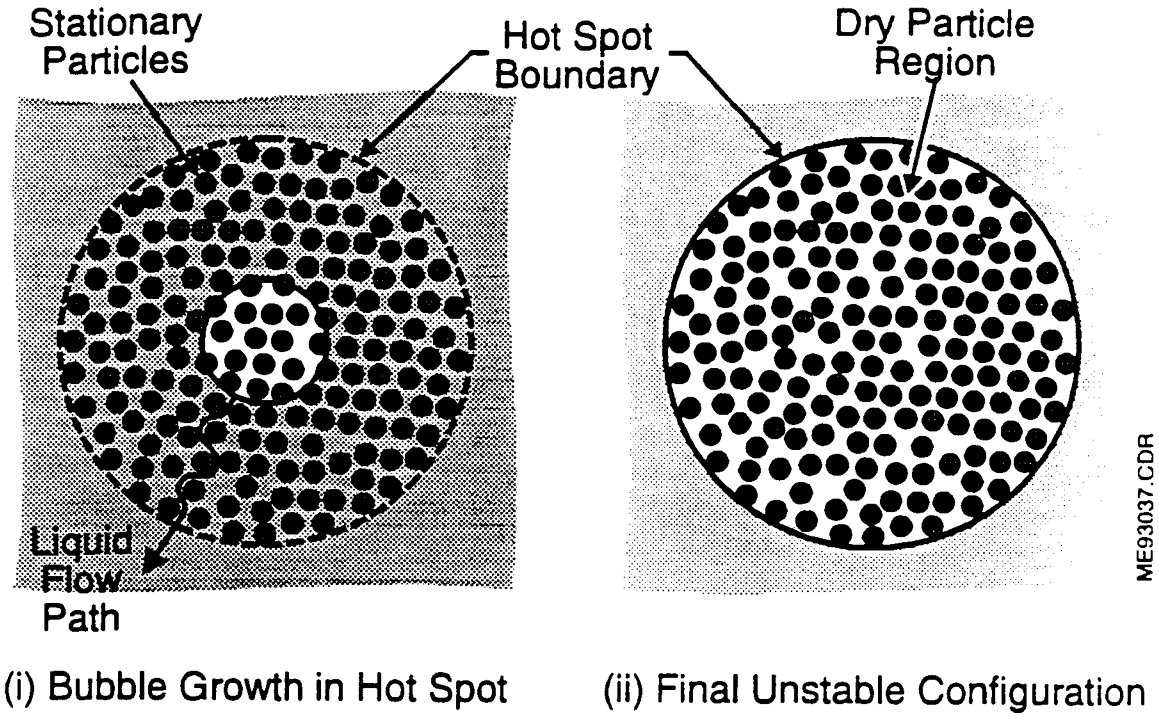

Figure 1a Porous medium model of dry spot formation from initial hot spot.

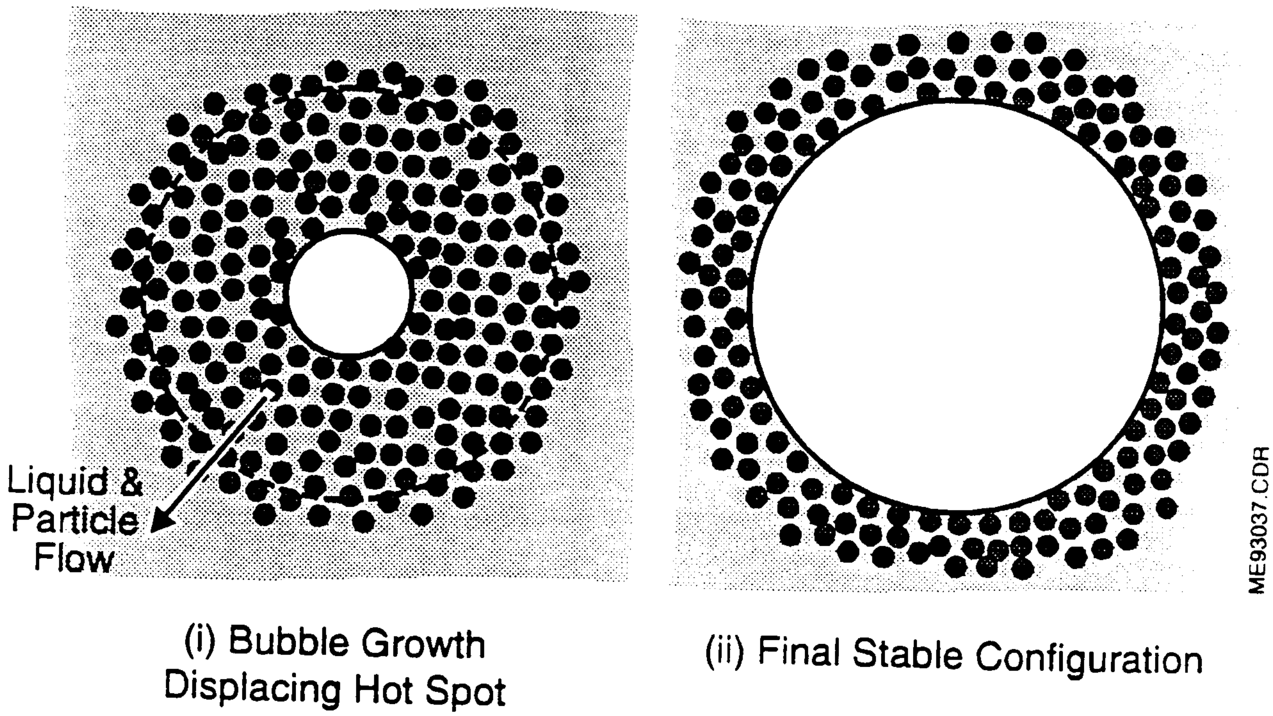

Figure 1b Homogeneous flow model of hot spot behavior.

FIGURE 1 Porous medium and homogeneous flow models of waste-tank sludge behavior. Note that for the sake of clarity the particulate material that is initially present outside the hot spot boundary is not shown.

3.0 BUBBLE GROWTH MOMENTUM EQUATIONS

The temperature of the waste-tank liquid will first rise above the boiling point T_{bp} of its liquid component at or very near the center of a thermally unstable hot spot. We consider the situation in which a small bubble of radius R_o containing vapor appears at the center of the hot spot. Note that we rule out multiple bubble nucleation events, or, equivalently, consider only that stage of the process following the coalescence of many small bubbles born near the center of the hot spot.

3.1 Bubble Growth in a Homogeneous non-Newtonian Two-Phase Flow

The continuity equation, when written for the spherically symmetrical incompressible liquid surrounding a bubble of instantaneous radius R(t), can be integrated at once to give

$$r^2 u = R^2(t) \frac{dR}{dt}(t) \qquad (1)$$

where r is the radial coordinate measured from the bubble center, u is the local radial velocity, and t is time. Note that in the case of radial liquid flow through a porous medium, u represents the actual liquid velocity in the pores (see Section 3.2). In writing Eq. (1) we have assumed that the bubble vapor density is negligible compared with the liquid density.

The force balance across the spherical bubble interface is

$$P(R^+) + \frac{2\sigma}{R} + \tau_{rr}(R) = P(R^-) = P_b \qquad (2)$$

where $P(R^+)$ is the pressure on the liquid side of the interface, σ is the surface tension of the liquid component of the waste tank mixture, τ_{rr} is the shear stress exerted in the r-direction on the liquid side of the interface, and P_b is the pressure within the bubble. The shear stress exerted by the vapor at the interface is ignored. The general form of the momentum equation for spherically symmetrical incompressible liquid flow is

$$\rho \frac{\partial u}{\partial t} + \rho u \frac{\partial u}{\partial r} = - \frac{\partial P}{\partial r} - \frac{\partial \tau_{rr}}{\partial r} + \frac{\tau_{\theta\theta} + \tau_{\phi\phi} - 2\,\tau_{rr}}{r} \tag{3}$$

where ρ is the density of the surrounding liquid and the τ_{ij}'s are the non-zero components of the shear stress for radial symmetric flow.

In order to transform Eqs. (2) and (3) into useful forms we must introduce appropriate expressions for the shear stress components. As pointed out by Wong (1992), the range of particle sizes found in waste tank sludges is typical of silts and clays. In fact Wong states that all the hydraulic property tests performed on ferrocyanide sludge simulants indicate that the properties of the sludge are similar to those of silts and clays. Therefore, for purposes of modeling the sludge as a homogeneous liquid/particle mixture, it is appropriate to employ the Ostwald-de Waele ("power law") model to express the relationship between shear stress and velocity gradient. For purely one-dimensional flow in a rectangular (x,y) coordinate system, the power law model is

$$\tau_{yx} = - m \left| \frac{du}{dy} \right|^{n-1} \frac{du}{dy} \tag{4}$$

where m and n are empirically determined parameters. This rheological law has been used to model silts, clays, cements, etc. (see, e.g., Bird, et al., 1960). The general form of Eq. (4) is

$$\boldsymbol{\tau} = - \left\{ m \left(\frac{1}{2}\, \Delta : \Delta \right)^{\frac{n-1}{2}} \right\} \Delta \tag{5}$$

where $\boldsymbol{\tau}$ is the stress tensor and Δ is the "rate of deformation tensor". From this equation we can determine the components of $\boldsymbol{\tau}$ for a non-Newtonian power law liquid undergoing a pure radial expansion by following the procedure outlined in Bird, et al., (1960).

The components of τ for a symmetrically spherical system are

$$\tau_{rr} = -2\eta \frac{\partial u}{\partial r} \tag{6}$$

$$\tau_{\theta\theta} = \tau_{\phi\phi} = -2\eta \frac{u}{r} \tag{7}$$

where the scalar viscosity for a power-law fluid is

$$\eta = m\left[2\left(\frac{\partial u}{\partial r}\right)^2 + 4\left(\frac{u}{r}\right)^2\right]^{\frac{n-1}{2}} \tag{8}$$

Thus the sum of the shear stress terms in Eq. (3) becomes

$$\frac{\tau_{\theta\theta} + \tau_{\phi\phi} - 2\,\tau_{rr}}{r} = 4m\left[2\left(\frac{\partial u}{\partial r}\right)^2 + 4\left(\frac{u}{r}\right)^2\right]^{\frac{n-1}{2}} \cdot \left(\frac{1}{r}\frac{\partial u}{\partial r} - \frac{u}{r^2}\right) \tag{9}$$

Substituting Eq. (9) into (3), using Eq. (1) to replace u in favor of r and R(t), and integrating the resulting momentum equation from the bubble surface (at r = R) to infinity yields

$$\rho R\ddot{R} + \frac{3}{2}\rho\,\dot{R}^2 + \frac{2\sigma}{R} + \frac{m\,12^{\frac{n+1}{2}}}{3n}\left(\frac{\dot{R}}{R}\right)^n = P_b - P_\infty \tag{10}$$

where P_∞ is the ambient pressure and the dot above R signifies differentiation with respect to time. Here we made use of Eq. (2) to relate the pressure in the liquid at the bubble wall to the bubble pressure. Now a complete model of bubble growth would ordinarily require an energy equation. However for the present we are only concerned with the growth behavior of the bubble very early in time, in fact at its birth, when the rate of supply of heat from the surrounding liquid is more than sufficient to keep up with the bubble growth rate (or, equivalently, the liquid evaporation rate). In a subsequent section we will consider energy transfer to the bubble wall. Early in time, the inertial resistance of the liquid to bubble growth is negligible compared with frictional resistance due to the rheological properties of the liquid. Thus we may ignore the first two terms of Eq. (10), and the momentum equation becomes

$$N\left(\frac{\dot{R}_H}{R_H}\right)^n = P_b - P_\infty - \frac{2\sigma}{R_H} \tag{11}$$

where

$$N = \frac{m\ 12^{(n+1)/2}}{3n} \tag{12}$$

The subscript H indicates that Eq. (11) describes bubble growth in a homogeneous sludge, that is when particles and liquid move together in accord with power law rheology. The subscript serves to distinguish this type of growth from bubble growth in a "porous medium sludge" of stationary particles, which is considered below.

3.2 Bubble Growth in a Porous Medium

Unlike the development of Eq. (10), there is no rigorous way of deriving an analogous momentum equation for flow in a porous medium. The theory of laminar flow in a porous medium is based on a classical experiment originally performed by Darcy (1856). His semi-empirical expression relates the average velocity through the porous medium to the pressure drop across the porous medium. For purposes of determining the local flow velocity in a porous medium, the following differential form of Darcy's law is normally used (see, e.g., Scheidegger, 1957).

$$u_s = -\frac{\kappa}{\mu}\frac{\partial P}{\partial r} \tag{13}$$

in which κ is the permeability of the porous medium and μ is the viscosity of the liquid component. The velocity u_s in this equation is the superficial velocity which when multiplied by the local, total flow cross-sectional area yields the volume flow of liquid. The pressure drop term in Eq. (13) is assumed to replace all the frictional drop terms on the right-hand side of Eq. (3), resulting in the following empirical modification of the momentum equation for flow in a porous medium:

$$\rho \frac{\partial u_s}{\partial t} + \rho u_s \frac{\partial u_s}{\partial t} = - \frac{\partial P}{\partial r} - \frac{\mu u_s}{\kappa} \tag{14}$$

In order to use the mass conservation equation, Eq. (1), it is necessary to relate the superficial velocity to the liquid velocity in the pores via the definition of the porosity ϵ of the porous medium, namely

$$u_s = \epsilon u \tag{15}$$

In Darcy flow the viscous forces dominate over inertia forces so that the left-hand side of Eq. (14) may be set equal to zero. Integrating the right-hand side from $r = R$ to $r = \infty$ and using Eqs. (1), (2) and (15) to eliminate $P(R^+)$ and u_s gives the desired result

$$\frac{\mu\epsilon}{\kappa} R_P \dot{R}_P = P_b - P_\infty - \frac{2\sigma}{R_P} \tag{16}$$

where the subscript P refers to bubble growth in a porous medium model of the waste tank sludge.

An examination of the right-hand sides of Eq. (11) or Eq. (16) indicates that the nucleus bubble will not begin to grow unless the initial bubble size satisfies the condition

$$R_o > \frac{2\sigma}{P_b - P_\infty} \tag{17}$$

4.0 STRINGENT CONDITION FOR HOMOGENEOUS PARTICLE-LIQUID FLOW

We now have two rheological models (momentum equations) for bubble growth in the waste tank sludge. These models are represented mathematically by Eqs. (11) and (16). Based on the available physical properties of the waste liquid (Wong, 1992), both models would appear to be appropriate. When the bubble is at its initial size R_o and ready to expand, it seems reasonable to suppose that the behavior of the surrounding liquid will be dictated by that mixture rheology which minimizes the effective liquid viscosity felt by the bubble. If we demand that the effective liquid viscosity associated with homogeneous, non-Newtonian flow be less than that associated with flow through a porous medium, the condition we wish to satisfy is

$$\dot{R}_H > \dot{R}_P \; ; \text{ at } R_H = R_P = R_o \tag{18}$$

for a given pressure drop $P_b - P_\infty - 2\sigma/R_o$. Note from Eqs. (11) and (16) that the velocity $\dot{R}_H$ of the wall of a bubble that "drives" a homogeneous flow increases with increasing bubble size while the opposite is true for the bubble that expands with velocity $\dot{R}_P$ against a surrounding liquid in a porous medium. Therefore, if the criterion given by Eq. (15) is satisfied when the bubble is formed, that is at its initial radius R_o, it remains satisfied during the subsequent bubble growth process.

Substituting Eqs. (11) and (16) into Criterion (18) and setting $R_H = R_P = R_o$ yields, after some algebraic manipulations,

$$P_b - P_\infty > \frac{2\sigma}{R_o} + \left(\frac{\kappa \; N^{1/n}}{\mu \; \epsilon \; R_o^{\,2}}\right)^{\frac{n}{1-n}} \tag{19}$$

We see that the criterion given by Eq. (19) implies that the bubble pressure, which in reality is the equilibrium vapor pressure of the liquid, must exceed some threshold value in order for the subsequent radial flow in the surrounding liquid-particle mixture to be homogeneous. To cause a bubble of size R_o to form, the liquid must be superheated to supply the additional vapor pressure $2\sigma/R_o$ so that the bubble achieves mechanical equilibrium with the liquid (see Eq. 17). The last term in Eq. (19) is the incremental pressure (or superheat) increase that is required to cause simultaneous bubble growth and homogeneous flow in the surrounding liquid-particle mixture. This pressure increase can be converted to an equivalent liquid superheat ΔT by invoking the linearized form of the Clasius-Clapeyron equation, namely,

$$\Delta T = \frac{T_{bp}}{\rho_g h_{fg}}\left(P_b - P_\infty - \frac{2\sigma}{R_o}\right) \tag{20}$$

to get

$$\Delta T > \frac{T_{bp}}{\rho_g h_{fg}}\left(\frac{\kappa\ N^{1/n}}{\mu\ \epsilon\ R_o^{\ 2}}\right)^{\frac{n}{1-n}} \tag{21}$$

In Eqs. (20) and (21) ρ_g is the density of the bubble vapor evaluated at the boiling point T_{bp} and h_{fg} is the latent heat of evaporation of the volatile liquid component of the waste sludge.

We are now in a position to estimate the level of liquid superheat that is required to ensure radial, outward movement of sludge in the form of a homogeneous particle-liquid flow. If the sludge is viewed as a silt or clay, the appropriate power law model parameters are $n = 0.229$ and $m = 5.6$ kg s^{n-2} m^{-1} (Bird, et al., 1960). The porous media model would suggest $\kappa = 10^{-14}$ m^2 and $\epsilon = 0.7$ (Wong, 1992). The remaining parameters are based on water-like properties for the liquid component: $h_{fg} = 2.2 \times 10^6$ J kg^{-1}, $\rho_g = 0.54$ kg m^{-3}, $\mu = 2.2 \times 10^{-4}$ kg m^{-1} s^{-1}, and a boiling point $T_{bp} = 400K$ at $P_\infty = 10^5$ Pa. Table 1 gives calculated threshold superheats for several practical nucleus bubble sizes. Given the small predicted values of ΔT it is obvious that homogeneous particle-liquid flow, as illustrated in Fig. 1b, is the preferred rheology.

Table 1

MINIMUM SLUDGE SUPERHEAT COMPATIBLE WITH HOMOGENEOUS SLUDGE FLOW

R_o, μm	10^2	10^1	10^0
ΔT, K	8.3×10^{-3}	3.3×10^{-2}	1.3×10^{-1}

5.0 ZERO INITIAL SUPERHEAT CRITERION FOR HOMOGENEOUS PARTICLE-LIQUID FLOW

In the previous section we demanded that the effective liquid viscosity associated with homogeneous, non-Newtonian flow be less than that associated with flow through a porous medium when the bubble is at its initial size R_o. This most stringent condition for homogeneous flow implies the existence of a small but finite initial superheat ΔT above that required to grow the critical bubble of size R_o. We now require the bubble growth process to be started with $\Delta T = 0$. In this situation ΔT increases at a rate proportional to the volumetric heat generation rate Q (see below). The sludge surrounding the bubble first behaves as a porous medium, with the liquid component moving within the pores formed by stationary sludge particulate, since, for a given bubble wall velocity ($\dot{R} = \dot{R}_H = \dot{R}_p$), the resistance to flow in a porous medium is directly proportioned to R (see Eq. 16) while the resistance to homogeneous flow is inversely proportional to R (see Eq. 11). Ultimately, however, the resistance seen by the bubble to its growth in a homogeneous sludge will fall below that in a porous medium sludge. We denote by R^* ($R^* > R_o$) the size of the bubble when this flow regime transition is made.

For $R < R^*$ the expansion of the bubble and the consequent motion of the surrounding liquid proceeds within the interstices of the porous media. We will assume that the growth of the bubble is limited by the rate of arrival of heat necessary for evaporation at the bubble surface. This assumption will be justified later on. It follows that, except for the earliest stages of bubble growth when surface tension is important, the bubble pressure P_b is for all practical purposes equal to P_∞. Thus there is no need to utilize the momentum equations, Eqs. (11) and (16) to predict the bubble growth history. However, the frictional terms in these equations are compared to determine the condition for homogeneous flow. We can expect such flow when the viscous resistance to bubble growth in a porous medium exceeds the viscous resistance to bubble growth in a homogeneous particle/liquid flow; that is, when

$$\frac{\mu\epsilon}{\kappa} R_p \dot{R}_p \geq N\left(\frac{\dot{R}_H}{R_H}\right)^n \tag{22}$$

at $R_H = R_p = R*$ and when $\dot{R}_p = \dot{R}_H = \dot{R}$. Thus, solving Eq. (22) for $\dot{R}$ gives the condition

$$\dot{R} \geq \left(\frac{\kappa N}{\mu\epsilon}\right)^{\frac{1}{1-n}} (R*)^{-\frac{1+n}{1-n}} \tag{23}$$

In order to transform Eq. (23) into a practical form we require an expression for the bubble wall velocity in a porous, heat-generating medium. The energy equation for heat-transfer controlled, spherically symmetric bubble growth in a porous medium is

$$\rho c_p \frac{\partial T}{\partial t} + \rho_f c_{p,f} u_s \frac{\partial T}{\partial r} = \frac{k}{r^2} \frac{\partial}{\partial r}\left(r^2 \frac{\partial T}{\partial r}\right) + Q \tag{24}$$

where c_p and k are, respectively, the specific heat and thermal conductivity of the sludge mixture; ρ_f and $c_{p,f}$ are, respectively, the density and specific heat of the liquid component of the sludge; T is the local temperature in the thermal boundary layer surrounding the bubble; and Q is the sludge volumetric heating rate (in W m^{-3}). An energy balance at the surface of the bubble provides the (moving) boundary condition

$$\epsilon h_{fg} \rho_g \dot{R} = k\left(\frac{\partial T}{\partial r}\right)_{r=R} \tag{25}$$

For heat transfer controlled bubble growth we have the additional condition at the bubble surface:

$$T(R,t) = T_{bp} \tag{26}$$

It will be demonstrated later on that, under the slow growth conditions of interest here, all the terms in Eq. (24) are unimportant in comparison with the conduction term within the thermal boundary layer that surrounds the bubble. Thus Eq. (24) has the simple solution

$$T(r,t) = T_{\infty}(t) - \frac{R}{r}(T_{\infty} - T_{bp}) \tag{27}$$

Outside the thermal boundary layer only the first and last terms in Eq. (24) are important and, therefore, we have the solution

$$T_{\infty}(t) = T_{bp} + \frac{Qt}{\rho c_p} \tag{28}$$

Substituting Eqs. (27) and (28) into Eq. (25) and integrating the result from $R = 0$ at $t = 0$ to $R < R^*$ at time t gives

$$R = \left(\frac{\alpha Q}{\epsilon h_{fg} \rho_g}\right)^{1/2} t \tag{29}$$

where α is the thermal diffusivity of the sludge mixture ($\alpha \approx 4.2 \times 10^{-7} m^2 s^{-1}$; see Wong, 1992).

Since Eq. (29) is valid for bubble sizes less than or equal to R^* (but much greater than R_o), we can combine this equation with Eq. (23) to obtain the following inequality for homogeneous particle-liquid flow:

$$Q \geq \frac{\epsilon h_{fg} \rho_g}{\alpha} \left(\frac{\kappa N}{\mu \epsilon}\right)^{\frac{2}{1-n}} (R^{*2})^{-\frac{1+n}{1-n}} \tag{30}$$

Equation (30) states that for a given instantaneous bubble size R^* ($R^* >> R_o$), the radial flow in the surrounding sludge is homogeneous for bubble growth beyond R^* when Q is sufficiently high. Table 2 gives the required calculated values of Q for two choices of R^*. Also shown in the table is the radius R_{HS} of a hot spot absorbing its heat load limit Q based on cooling by radial heat conduction to a surrounding ambient at 20°C. If the initial liquid superheat is zero there is always an early domain of bubble growth during which porous flow resistance is less than homogeneous flow resistance. However, as can be seen from Table 2, this domain ends well before the bubble grows to a size that is significant on the scale of the hot spot dimension R_{HS}. For example, a bubble that grows from, say, initial size $R_o = 1.0$ μm, in a sludge internally heated at a rate of only 15.3 W m^{-3}, would, according to the present theory, feel a switch from porous-medium to homogeneous-flow limitations when it grows through the size R* = 300 μm.

Table 2

MINIMUM HEAT GENERATION RATE COMPATIBLE WITH HOMOGENEOUS SLUDGE FLOW

R*, μm	Q, W m^{-3}	R_{HS}, m[+]	R_{HS}/R*
100	508	0.92	9.2×10^3
300	15.3	5.3	1.8×10^4

[+] $R_{HS}^2 = 2k(T_{bp} - 20°C)/Q$

6.0 EXAMINATION OF ASSUMPTIONS USED IN HOT SPOT ANALYSES

In deriving Eq. (21) of Section 4 we have neglected the effect of sludge inertia. The inertia terms in the momentum equations, Eqs. (10) and (14), are of the order $\rho\dot{R}^2$. An examination of the momentum equations indicates that inertial limitations to the early stage of bubble growth (in a homogeneous mixture or porous medium) will be negligible provided that $\rho\dot{R}^2$ is small compared with the "frictional pressure drop"; that is

$$\frac{\rho\dot{R}^2}{P_b - P_\infty - 2\sigma/R} << 1 \tag{31}$$

Using Eq. (11) to evaluate $\dot{R}$ and Eq. (20) to eliminate the pressure drop terms in favor of the liquid superheat ΔT, Eq. (31) leads us to expect negligible inertial limitations when:

$$\frac{\rho R_o^2}{N^{2/n}}\left(\frac{\rho_g h_{fg} \Delta T}{T_{bp}}\right)^{\frac{2}{n} - 1} << 1 \tag{32}$$

We find that for all waste tank cases of practical interest this inequality is indeed satisfied. For example, for $R_o = 1.0\ \mu m$ and $\Delta T = 1.3 \times 10^{-1}$ (see Table 1), the left-hand side of Eq. (32) is 2.8×10^{-3}.

We have also neglected the effects of liquid inertia in the conduction-controlled bubble growth analysis of the previous section. In particular we have assumed that the pressure drop in the liquid due to inertia, namely $\rho\dot{R}^2$, is small compared with the maximum possible pressure drop based on the instantaneous liquid superheat given by Eq. (28), or, in mathematical terms, we have assumed that (see, also, Eq. 20)

$$\frac{\rho\dot{R}^2}{\left(\frac{\rho_g h_{fg}}{T_{bp}}\right)\left(\frac{Qt}{\rho c_p}\right)} << 1 \tag{33}$$

By using Eq. (29) to obtain $\dot{R}$ and t in terms of R, we may rewrite the above inequality as

$$\frac{\rho^2 c_p T_{bp}}{\rho_g h_{fg} Q R}\left(\frac{\alpha Q}{\epsilon h_{fg}\rho_g}\right)^{3/2} \ll 1 \tag{34}$$

We note that this inequality can not be satisfied for vanishingly small bubble sizes ($R \to 0$). However, if we are willing to ignore the very earliest stages of bubble growth we find that the left-hand side of Eq. (34) is indeed small; it is only, for example, 2.9×10^{-5} for bubbles growing past $R = 1.0\ \mu m$ in a sludge volumetrically heated at the rate $Q = 10^3\ W\ m^{-3}$.

Recall that Eq. (24) was simplified by ignoring (i) the transient heat storage term, (ii) the convective term and (iii) the heat generation term within the liquid near the phase change interface. Assumption (i) is valid providing that the time it takes for a thermal wave to traverse the boundary layer thickness, namely $t \sim R^2/\alpha$, is short compared to the time it takes the bubble to grow to size R*. This is the case if (see Eq. 29)

$$R^* \cdot \left(\frac{Q}{\epsilon h_{fg}\rho_g \alpha}\right)^{1/2} \ll 1.0 \tag{35}$$

The left-hand side of this inequality is approximately 2×10^{-3} (see Table 2 for input values). Assumption (ii) is valid if the convective term in Eq. (1), namely $\rho_f c_{p,f} u_s (T_\infty - T_{bp})$, is small compared to the instantaneous conduction heat flux $k[T_\infty(t) - T_{bp}]/R$ to the bubble wall, or

$$\frac{\rho_f c_{p,f} R^*}{k}\left(\frac{\epsilon \alpha Q}{h_{fg}\rho_g}\right)^{1/2} \ll 1 \tag{36}$$

This inequality was obtained by using Eqs. (1) and (15) to eliminate u_s and then by using Eq. (29) for $\dot{R}$. The left-hand side of Eq. (36) is typically of the order 2×10^{-3}. Finally, in order to demonstrate the validity of Assumption (iii), we consider the ratio of the effective heat flux QR due to volumetric heating to the actual instantaneous conduction heat flux $k[T_\infty(t)$

$- T_{bp}]/R$. With the aid of Eqs. (28) and (29), this ratio can readily be shown to be identical to the left-hand side of Eq. (35), which we already know is small.

7.0 DRAINING FROM WASTE TANKS

Suppose a liquid/solid particulate sludge is allowed to drain from a waste tank through a circular opening in the tank wall or tank bottom. The question of interest is whether or not only the liquid will drain, thereby leaving behind the dry, heat generating solid particulate. Our approach to this problem is to first assume that the particulate remains in the tank and, therefore, the rate of draining is controlled by liquid flow through the porous medium established by the stationary particulate. Then we analyze the problem of two-phase homogeneous flow of solid particulate and liquid into the opening. The homogeneous mixture is assumed to flow as a power-law (Ostwald-de Waele) fluid. Of these two rheological behaviors (flow in porous medium or homogeneous flow), the one that results in the least resistance to drainage is deemed to be the actual mode of fluid drainage.

7.1 Flow Through Porous Medium Into Circular Opening

We consider an axisymmetric (r,z) flow of liquid from a semi-infinite porous medium to a circular opening in the tank boundary (wall). The radial coordinate r is measured from the center of the opening and lies in the plane that coincides with the inside surface of the tank wall. The z-coordinate is perpendicular to the tank wall and is measured from the center of the opening into the tank. Within the tank the liquid moves through the porous solid in the negative z-direction toward the opening.

The superficial velocity components (u_s, v_s) in the r- and z-directions are, in accordance with Darcy's law for flow in porous media (see Eq. 13),

$$u_s = -\frac{\kappa}{\mu}\frac{\partial P}{\partial r} \qquad (37)$$

$$v_s = -\frac{\kappa}{\mu}\frac{\partial P}{\partial z} \qquad (38)$$

where P is the local liquid pressure. Differentiating Eq. (37) with respect to z and differentiating Eq. (38) with respect to r and eliminating $\partial^2 P/\partial r \partial z$ between the resulting equations gives

$$\frac{\partial u_s}{\partial z} - \frac{\partial v_s}{\partial r} = 0 \tag{39}$$

The remaining equation that must be satisfied is the continuity equation for an axisymmetric system:

$$\frac{1}{r}\frac{\partial}{\partial r}(r u_s) + \frac{\partial v_s}{\partial z} = 0 \tag{40}$$

The velocity components are now related to the velocity potential ϕ as

$$u_s = \frac{\partial \phi}{\partial r} \quad ; \quad v_s = \frac{\partial \phi}{\partial z} \tag{41}$$

so that Eq. (39) is automatically satisfied and Eq. (40) becomes

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r}\frac{\partial \phi}{\partial r} + \frac{\partial^2 \phi}{\partial z^2} = 0 \tag{42}$$

If V_s is the velocity (superficial) of draining in the plane of the opening, the boundary condition for ϕ along the tank wall (i.e., at z = 0) is

$$v_s(r,0) = \frac{\partial \phi}{\partial z}(r,0) = \begin{cases} -V_s & \text{for} \quad r < R \\ 0 & \text{for} \quad r > R \end{cases} \tag{43}$$

where R is the radius of the circular opening. Far from the opening, we have the boundary conditions

$$\phi \to 0, \ \frac{\partial \phi}{\partial z} \to 0, \ \frac{\partial \phi}{\partial r} \to 0 \quad \text{as} \quad z \to \infty \tag{44}$$

To solve Eq. (42) we introduce the infinite Hankel transform

$$\Phi(\xi,z) = \int_0^\infty r\phi(r,z)J_0(\xi r)dr \tag{45}$$

Multiplication of both sides of Eq. (42) by $rJ_0(\xi r)$ and integration with respect to r from 0 to ∞, we find, as a result of integration by parts, that the transform satisfies the equation

$$\frac{d^2\Phi}{dz^2} - \xi^2\Phi = 0 \tag{46}$$

Hankel transforming the boundary conditions (43) and (44) gives

$$\frac{d\Phi}{dz} = - V_s \int_0^R rJ_0(\xi r)dr = - \frac{V_s R}{\xi} J_1(\xi R) \quad \text{at} \quad z = 0 \tag{47}$$

$$\Phi \to 0, \ \frac{d\Phi}{dz} \to 0 \quad \text{as} \quad z \to \infty \tag{48}$$

The solution of Eq. (46) subject to boundary conditions (47) and (48) is

$$\Phi = \frac{V_s R}{\xi^2} J_1(\xi R)e^{-\xi z} \tag{49}$$

Inverting the above equation by means of the Hankel inversion theorem, namely

$$\phi(r,z) = \int_0^\infty \xi\Phi(\xi,z)J_0(r\xi)d\xi \tag{50}$$

we obtain

$$\phi(r,z) = V_s R \int_0^\infty \frac{1}{\xi} J_1(R\xi)J_0(r\xi)e^{-\xi z}d\xi \tag{51}$$

The velocity components follow from Eq. (41):

$$u_s = - V_s R \int_0^\infty J_1(R\xi) J_1(r\xi) e^{-\xi z} d\xi \tag{52}$$

$$v_s = - V_s R \int_0^\infty J_1(R\xi) J_0(r\xi) e^{-\xi z} d\xi \tag{53}$$

Now the pressure difference $P_\infty - P_0$ required to maintain the liquid velocity V_s in the plane of the opening can be obtained by integrating either Eq. (37) from $r = 0$ to $r = \infty$ in the plane $z = 0$ or Eq. (38) from $z = 0$ to $z = \infty$ along the centerline $r = 0$. Choosing the former method, we have

$$P_\infty - P_0 = - \frac{\mu}{\kappa} \int_0^\infty u(r,0) dr \tag{54}$$

Note that P_∞ is the pressure within the tank far from the opening and P_0 is the pressure in the plane of the opening. Substituting Eq. (52) into Eq. (54) gives, after reversing the order of integration,

$$\begin{aligned} P_\infty - P_0 &= \frac{\mu V_s R}{\kappa} \int_0^\infty J_1(R\xi) \left[\int_0^\infty J_0(\xi r) dr \right] d\xi \\ &= \frac{\mu V_s R}{\kappa} \int_0^\infty \frac{J_1(R\xi)}{\xi} d\xi = \frac{\mu V_s R}{\kappa} \cdot \frac{\Gamma(1/2)}{2\Gamma(3/2)} \end{aligned} \tag{55}$$

Since $\Gamma(1/2) = 2\Gamma(3/2)$, we have the simple result

$$P_\infty - P_0 = \frac{\mu V_s R}{\kappa} \tag{56}$$

which, interestingly enough, could have been readily obtained by dimensional analysis. The volumetric flow rate $\dot{V}$ through the opening (in $m^3\ s^{-1}$) is

$$\dot{V} = \pi R^2 V_s = \frac{\pi R \kappa (P_\infty - P_0)}{\mu} \tag{57}$$

If the liquid (water at ~ 20°C, $\mu = 10^{-3}$ kg m^{-1} s^{-1}) drains under a 1.0-m gravity head through a porous medium with $\kappa = 10^{-14}$ m^2 (Wong, 1992) into a circular opening of size R = 0.1 m, we estimate $\dot{V} = 3.1 \times 10^{-8}$ m^3 s^{-1}. The time required to lower the liquid level in the tank from its initial value H_0 to H ($H < H_0$) via gravity is derived from the simple mass balance $A dH/dt = - \dot{V}$ and is

$$t_{drain} = \frac{\mu A \ln(H_0/H)}{\pi R \kappa \rho_f g} \tag{58}$$

where A is the cross-sectional area of the tank. Referring to the previous example, with A = 400 m^2, we find that it requires

$$t_{drain} = 3 \times 10^{10} s = 950 \text{ yr}$$

to reduce the liquid level by 90 percent.

7.2 Flow of Power-Law Liquid/Particle Mixture Into Circular Opening

The power law model for the fluid shear stress is given by Eq. (5). While one can use this relationship to write down the components of τ for a non-Newtonian axisymmetric flow into a circular opening, the solution of the resulting momentum equation is a formidable undertaking. A simple way of arriving at an approximate relationship between $P_\infty - P_0$ and the efflux velocity V in the plane of the opening is to employ dimensional analysis. Since m, V, and R are the only obvious dimensional parameters and since m has the units kg s^{n-2} m^{-1}, we immediately arrive at the formula

$$P_\infty - P_0 = C \frac{mV^n}{R^n} \tag{59}$$

where C is an unknown proportionality constant. In using Eq. (59) one has little choice but to assume that C is of order unity.

In what follows we will strive for a more accurate formula than Eq. (59) by attempting to estimate C. If it is assumed that the non-Newtonian flow toward the opening is entirely radial, then the local flow velocity u is only a function of the distance r measured from some virtual point sink at which the fluid is absorbed continuously. The equation of motion for such a radial (spherically symmetric) creeping flow is (see Eq. 3)

$$\frac{\partial P}{\partial r} = -\frac{\partial \tau_{rr}}{\partial r} - \frac{\tau_{\theta\theta} + \tau_{\phi\phi} - 2\tau_{rr}}{r} \tag{60}$$

Experience with this class of incompressible flow problems indicates that inside the tank, away from the walls and far from the opening, the streamlines are "aimed" at the center of the opening. Thus it seems reasonable to locate the virtual sink (or origin) of the system at the center of the opening. Unfortunately, in the near field of the opening, say within a radial distance of the origin roughly equivalent to the radius R of the opening, the streamlines bend away from the origin and pass through the opening some lateral offset distance from the central streamline (z-axis). Thus we cannot extend our postulated radial flow into the near field and, therefore, we cannot estimate the pressure drop associated with the flow close to the opening. One way of circumventing this difficulty is to assume that the flow velocity V in the plane of the opening is achieved in our purely radial flow field at r = R. The continuity equation then becomes

$$ur^2 = VR^2 \tag{61}$$

The equation for the pressure drop $P_\infty - P_0$ is obtained by integrating Eq. (60) from r = R to r = ∞:

$$P_\infty - P_0 = -\tau_{rr}(\infty) + \tau_{rr}(R) - \int_R^\infty \frac{\tau_{\theta\theta} + \tau_{\phi\phi} - 2\tau_{rr}}{r}\, dr \tag{62}$$

The components of τ for a symmetrically spherical system are, from Eqs. (6) through (8) and (61),

$$\tau_{rr} = - 2\eta \frac{\partial u}{\partial r} = 4\eta \frac{VR^2}{r^3} \tag{63}$$

$$\tau_{\theta\theta} = \tau_{\phi\phi} = - 2\eta \frac{u}{r} = - 2\eta \frac{VR^2}{r^3} \tag{64}$$

where the scalar viscosity η for a power-law fluid is

$$\eta = m\left[2\left(\frac{\partial u}{\partial r}\right)^2 + 4\left(\frac{u}{r}\right)^2\right]^{\frac{n-1}{2}} = m(12)^{\frac{n-1}{2}}\left(\frac{VR^2}{r^3}\right)^{n-1} \tag{65}$$

Substituting Eqs. (63) through (65) into Eq. (62) and performing the indicated integration gives the sought result; namely,

$$P_\infty - P_0 = \frac{12^{\frac{n+1}{2}}}{3} \cdot \left(1 + \frac{1}{n}\right) \cdot m \left(\frac{V}{R}\right)^n \tag{66}$$

The accuracy of this expression can be checked in the limit of Newtonian flow ($n=1$; $m=\mu$):

$$P_\infty - P_0 = \frac{8\mu V}{R} \tag{67}$$

The available empirical data (Miller, 1989) on low-Reynolds flow through an orifice indicates that the coefficient 8 should be changed to a value somewhere between 9 and 9.5.

7.3 Criterion for Homogeneous Flow During Draining

We may now estimate the critical pressure drop $\Delta P = (P_\infty - P_0)_{crit}$ above which the flow of sludge toward the opening is a homogeneous power-law flow. For a given pressure drop ΔP we require the velocity V_s in Eq. (56) for flow through a porous medium to be less than V in Eq. (66), or

$$R\left(\frac{\Delta P}{M}\right)^{\frac{1}{n}} > \frac{\kappa \Delta P}{\mu R} \tag{68}$$

where M is defined as

$$M = \frac{12}{3}^{\frac{n+1}{2}} \cdot \left(1 + \frac{1}{n}\right) \cdot m \tag{69}$$

Solving Eq. (69) for ΔP, the criterion for the minimum ΔP consistent with homogeneous flow from the waste tank is

$$\Delta P > \left(\frac{\kappa}{\mu R^2}\right)^{\frac{n}{1-n}} (mM)^{\frac{1}{1-n}} \tag{70}$$

Again using properties typical of water-saturated clay at 20°C ($\kappa = 10^{-14}$ m^2, $\mu = 10^{-3}$ kg m^{-1} s^{-1}, m = 5.6 kg s^{n-2} m^{-1}, and n = 0.229), we estimate that ΔP need only exceed 0.3 Pa in order for the sludge to flow as a homogeneous particle/liquid mixture into a 0.2-m diameter opening. This pressure drop is equivalent to a minimum sludge depth of only 30 μm.

8.0 CONCLUDING REMARKS

A homogeneous two-phase model of ferrocyanide sludge flow has been suggested. In this model the solid precipitate and liquid components of the sludge flow together at the same velocity as a non-Newtonian power-law fluid. At this stage of our knowledge of the hydraulic properties of ferrocyanide waste sludges, this model would appear to be just as reasonable as the popular porous medium model in which liquid flows through the pores of stationary solid precipitate. Comparisons of the frictional resistance of the porous medium sludge rheology with that of the homogeneous flow rheology for sludge flow due to "hot spot bubble growth" or due to drainage from a waste tank leads to the conclusion that homogeneous solid/liquid flow is more likely in these flow situations. This finding precludes the overheating of the reactive solid phase since the separation of the liquid and solid components can only occur when the sludge flow obeys the porous medium rheology.

Of course, our conclusion that homogeneous flow is favored over flow through a porous medium may depend to some extent on our choice of power-law homogeneous-flow rheology. Different theoretical results could be obtained if, for example, homogeneous flow were modeled as a Binghan plastic with a high yield stress. Thus, in order to guide future theoretical developments in the general area of the thermal stability of ferrocyanide waste sludges, it is imperative to obtain experimental data on sludge flow behavior under conditions that appropriately simulate bubble growth within hot spots and sludge drainage from waste tanks. Such a program should concentrate on determining whether or not the liquid phase separates from the solid phase of the sludge.

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10.0 NOMENCLATURE

A	Cross-sectional area of tank.
C	Proportionality constant; Eq. (59).
c_p	Specific heat of sludge mixture (liquid + particulate).
$c_{p,f}$	Specific heat of liquid component of sludge.
g	Gravitational constant.
H_o	Initial height of sludge in tank.
H	Instantaneous height of sludge in tank.
h_{fg}	Latent heat of evaporation of liquid component of sludge.
k	Thermal conductivity of sludge mixture (liquid + particulate).
M	Modified power law coefficient; Eq. (69).
m	Coefficient in power-law shear stress model; Eq. (4).
N	Modified power law coefficient; Eq. (12).
n	Exponent in power-law shear stress model, Eq. (4).
P	Local pressure in liquid sludge.
P_b	Pressure in bubble.
P_o	Pressure in the plane of the drainage opening.
P_∞	Pressure in sludge far from the bubble or far from the opening through which sludge drains.
Q	Volumetric heat generation rate in sludge mixture.
R	Instantaneous bubble radius or radius of opening in tank for sludge drainage analysis.
R_{HS}	Radius of hot spot at incipient boiling condition.
R_o	Size of nucleus bubble.
$R*$	Bubble size at which liquid flow transition from porous medium to homogeneous rheology is made.
$\dot{R}$	dR/dt.

$\ddot{R}$ — d^2R/dt^2.

r — Radial coordinate measured from the center of the bubble or from the center of the opening in the tank wall during sludge drainage.

T — Local temperature in liquid sludge.

T_{bp} — Boiling point of liquid component of sludge.

T_∞ — Instantaneous temperature of sludge far from bubble wall.

t — Time.

t_{drain} — Drainage time; Eq. (58).

u — Local-radial-liquid velocity during bubble growth.

u_s — Local-radial-superficial-liquid velocity during bubble growth, or during axisymmetric drainage through opening.

V — Sludge velocity of draining (homogeneous flow model) in the plane of the opening.

$\dot{V}$ — Volumetric rate of drainage.

V_s — Superficial velocity of draining (porous medium model) in the plane of the opening.

v_s — Local-axial-superficial-liquid velocity during axisymmetric drainage through opening.

z — Coordinate perpendicular to tank wall in liquid drainage analysis.

Greek Letters

α — Thermal diffusivity of sludge; $k/(\rho c_p)$.

η — Scalar viscosity; Eqs. (6) and (7).

Δ — Rate of deformation tensor; Eq. (5).

ΔP — Critical pressure drop above which homogeneous sludge flow to drain opening is expected.

ΔT — Superheat in liquid above that required to produce bubble growth and homogeneous particle-liquid flow.

ϵ — Porosity in porous-medium model of sludge.

Φ — Hankel transform of velocity potential; Eq. (45).

ϕ — Velocity potential; Eq. (41).

κ Permeability in porous-medium model of sludge.

ξ Dummy integration variable in Hankel transform; Eq. (45).

μ Viscosity of liquid component of sludge.

ρ Density of sludge mixture (liquid + particulate).

ρ_f Density of liquid component of sludge.

ρ_g Density of bubble vapor evaluated at T_{bp}.

σ Surface tension of liquid component of sludge.

τ Stress tensor; Eq. (5).

τ_{ij} Components of the shear stress for spherically symmetric flow.

Subscripts

H Refers to homogeneous sludge rheology.

P Refers to porous media sludge rheology.

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