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NUMERICAL SIMULATION OF HIGH-LEVEL RADIOACTIVE NUCLEAR WASTE GLASS PRODUCTION (U)

by

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ABSTRACT

Vitrification of radioactive waste has become an international approach for converting highly radioactive wastes into a durable solid prior to placing them in a permanent disposal repository. The technology for the process is not new. The conversion melter is a direct decendant of all electric melters used for manufacturing of some commercial glass types. Therefore, the vitrification process of radioactive wastes inherits typical problems of all electric furnaces and creates some other specific problems such as noble metal sedimentation. The noble metals and nickel sulfides in the melter are heavier than molten glass and have a low solubility. In a reducing condition, these metals amalgamate and tend to settle on the melter floor. The metal deposit resulting from this settling has a potential to short circuit the melter. The objective of this paper is to identify the typical problems that have been encountered in the waste melter operations and to address how these problems can be tackled using state-of-the-art numerical simulation techniques. It is believed that the large amount of pilot-scale melter experience throughout the world, combined with the knowledge gained from state-of-the-art computer modeling techniques would give assurance that the existing and future radioactive wastes can be effectively converted into a durable glass material and safely placed in a permanent repository.

INTRODUCTION

In the past twenty four years, the vitrification process of high-level radioactive waste has evolved from a small-scale batch operation to the present-day, high-capacity continuous process. Over this period, the process changed from a batch process, where the feed in a canister is directly heated to melting temperature, to a continuous process with all metal melters and, finally, a Joule-heating process with ceramic melters[1].

The Joule-heating process utilizes the glass as an electrolyte at high temperatures. An alternating electric current passing through the glass between electrodes generates heat by the Joule heating effect, thus melting the incoming feed and maintaining the molten state of the glass. The Joule-heated electric melting concept for the high-level waste started with bench scale melters in the U.S. in 1973[2] followed by the wide-spread use of the concept in the Soviet Union in 1974, Germany in 1976, Japan in 1977, and the United Kingdom in 1980. For Joule-heated melters,

convection currents are the principle means of mixing and need to be promoted. Enhanced convection currents, however, increase the rate of refractory erosion, which shortens melter life. Therefore, from a melter design point of view, the balance between the glass mixing and refractory erosion rate is the principle design criterion to be considered. This criterion can be determined only if interactions among glass temperature, velocity, and electric field in the melt pool are properly characterized.

Aside from design issues, there are melter operational issues that originate from combined effects of operation parameters. An operational issue that brought forth notable attention in recent years involves the noble metal deposition process in commercial waste melter operations. Noble metals or nickel sulfides in the melter feed are heavier and have lower solubility than other chemical species within the molten glass. In a reducing condition, these metals amalgamate and tend to settle on the melter floor, and the resulting deposits have a potential to short-circuit the melter.

Resolution of current operational issues such as the noble metal deposition problem requires an integrated approach of experiments, physical modeling, and mathematical modeling. Unlike the commercial glass manufacturing process, however, the nuclear waste process prohibits human access to the melter, in which direct measurements can be performed, so the need to develop modeling techniques as alternative numerical experiment tools is now more critical than ever.

Mathematical modeling techniques that can be used to study the fundamental phenomena of waste glass processing for advanced melter design are described in this paper. The noble metal deposit problem is addressed as a primary operational issue, and the process can be characterized in terms of noble metal deposit rate, size distribution, and residence time.

MELTER SYSTEM MODEL

The largest-scale, proto-type, liquid-fed, Joule-heated ceramic melter in the United States[3] (Figure 1) was constructed at the Savannah River Site (SRS) Defense Waste Products Facility (DWPF). Because SRS stores about 35 percent of the total volume and 70 percent (10^9 Ci) of the radioactivity of all Defense High-Level Waste (DHLW) accumulated in the U.S., the process adopted in the DWPF vitrification program might be considered as a reference process.

The DWPF melter vitrifies radioactive fission products by mixing them with borosilicate glass and water and feeding the resulting waste slurry directly into the melter. The molten glass waste in the melter is poured continuously into stainless steel canisters. In the feeding process, water vaporizes in the plenum space and the solid portion of the radioactive waste called "cold cap" melts into molten glass.

Because of similarities in design, most mathematical modeling techniques developed in the commercial glass industry can be applied to the waste-glass model. The theoretical development of mathematical models in the commercial glass industry was reviewed in the open literature[4,5,6]. Most of these models solve for velocity, temperature, and electric potentials of the glass melt in the furnace. Earlier 2-D modeling work involved glass properties such as color, viscosity, electric resistivity, and density as critical parameters. Later 3-D finite element and finite difference models involved furnace operation parameters such as minimum residence time, melting index, fining index, and recirculating ratio.

The batch-feed model provides the material source to and the necessary boundary conditions for the glass-melt model and also provides the glass-melt interface with the combustion gas in the crown. Combustion-gas and glass-melt models are usually treated separately, but because of the substantial mass and heat transfer among glass melt, batch-feed pile, and combustion gas, these models need to be integrated to form a closed system.

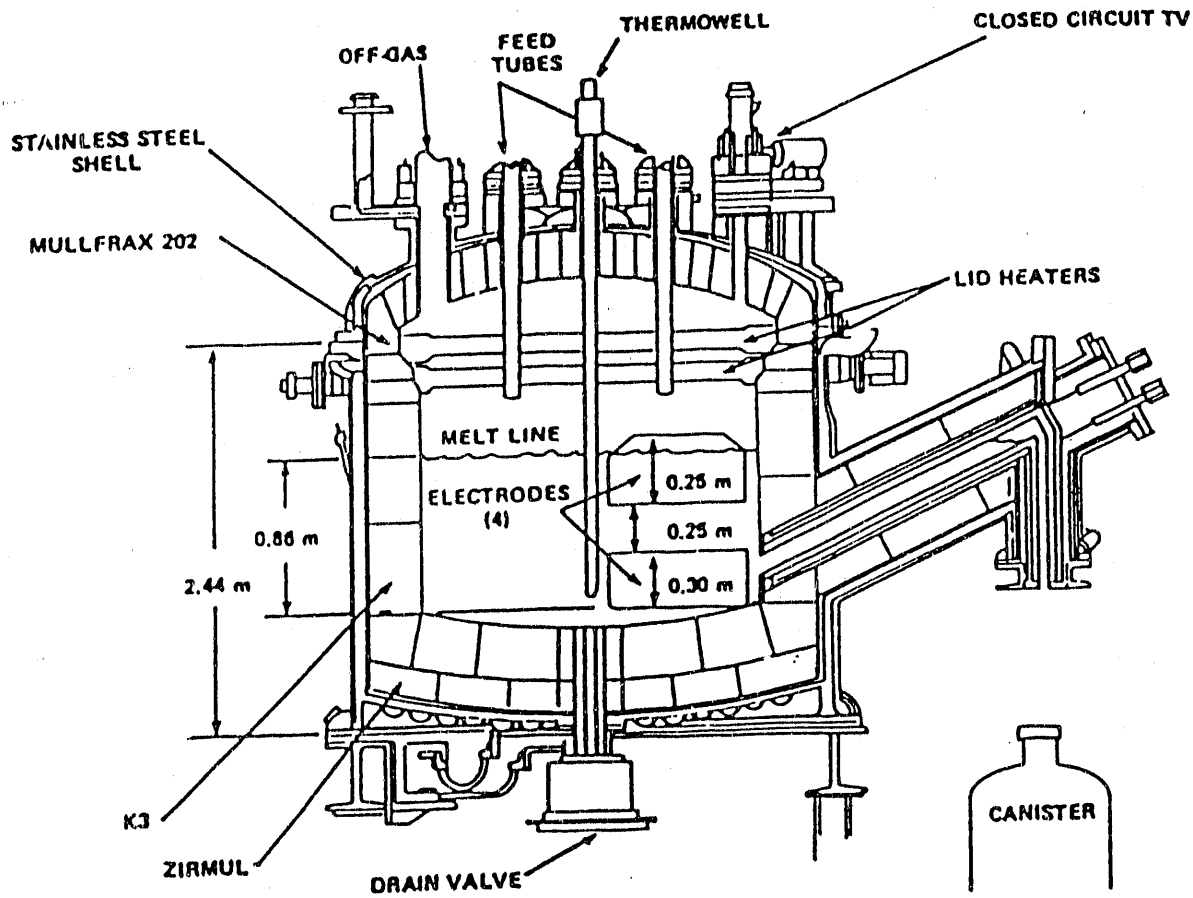

Figure 1. Cross Section of Defense Waste Processing Facility Melter

Nuclear waste melter models (Figure 2) can be catagorized similarly. The major difference in modeling strategy between the commercial glass and waste glass processes is derived from the melter feed (e.g. dry batch feed for the commercial glass furnace as opposed to wet slurry feed for the waste glass melter). The cold cap formed by wet slurry in the waste glass melter creates a highly unstable velocity and temperature field that creates difficulties in obtaining converging solutions.

Glass Melt Circulation

Several melter models exist at various Department Of Energy (DOE) nuclear waste facilities. Most, however, were developed to resolve specific melter design and operational issues in a relatively short time span in the early 1980's; therefore, they are too simplistic to be alternative tools to actual experiments. The most comprehensive model is the TEMPEST code developed at Pacific Northwest Laboratory (PNL).

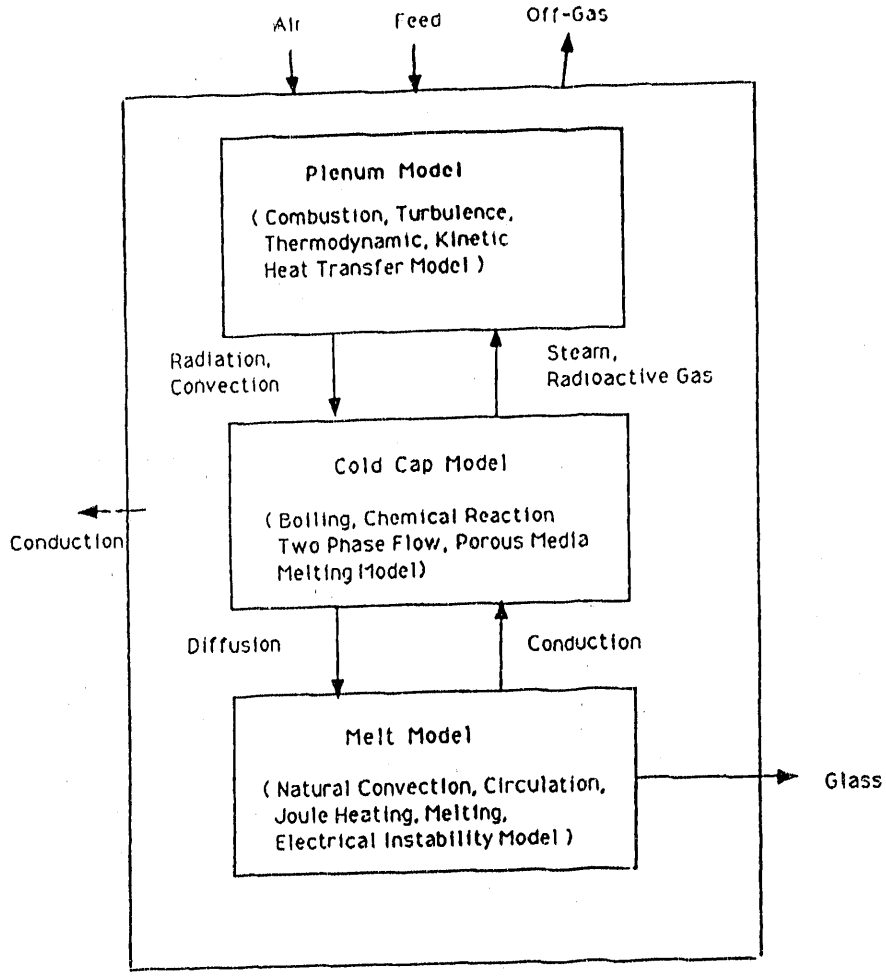

Figure 2. Schematic Representation of Melter Model

TEMPEST is a 3-D finite volume computer code that was specifically designed to model the radioactive waste-glass vitrification process. It has been used for melters at various DOE waste facilities - mostly at the Hanford site. TEMPEST is not available for public use but the contents are fairly well documented[7,8]. The recent modeling effort at SRS nvolves modification of the commercial general-purpose 3-D finite element codes that can solve momentum, energy, and electric equations simultaneously with temperature-dependent glass properties[9]. These models solve the fundamental conservation equations for the momentum, energy, and electric potentials as shown in equations (1) through (4).

$$\frac{\partial r_{\circ}}{\partial t} + \nabla.(\rho\bar{u}) = 0 \qquad (1)$$

$$\rho_0\left(\frac{\partial \bar{u}}{\partial t} + \bar{u}.\nabla\bar{u}\right) = -\nabla\rho + \nabla.(\mu\nabla\bar{u}) + \rho g[1-\beta(T-T_0)] + k_E EB \qquad -(2)$$

$$\rho_{o}C_{\rho}\left(\frac{\partial T}{\partial t}+\bar{u}\nabla T\right)=\nabla.(k_T\nabla T)+\nabla E.(k_E\nabla E) \tag{3}$$

$$C_E\frac{\partial E}{\partial t}=\nabla.(k_E\nabla E)+I+S_1(E)+S_2(Q) \tag{4}$$

The magnetic field form $K_E EB$, displacement current density $S_1(E)$, and charge density $S_2(Q)$ are assumed to be small. In addition to coupling all the governing equations through temperature-dependent properties, the energy equation is coupled to the momentum equation through the advection and the natural convection term, and the electric equation is coupled to the energy equation through the heat-source term [i.e. the last term in equation (3)].

It is worth remarking that these equations are highly nonlinear because of glass property variations with temperature; strong coupling of the momentum, energy, and strong temperature gradients developed by the high FeO contents in the electric equation; and the highly unstable mode of glass velocity and temperature field from the presence of the cold cap. In addition, past pilot-scale melter operations have indicated the possibility of an electric instability when glass electric resistivity exceeds the limit [10] per equation (5).

$$\frac{1}{\rho_E}\frac{\partial \rho_E}{\partial T})0.3(\%c) \tag{5}$$

This type of electric instability develops a positive feedback of electric currents that can result in electrode melting; thus, the glass-melt model should be able to account for this effect. In addition, the strong coupling between the governing equations requires many iterations to achieve convergence.

The choice of a Joule-heating melter for waste-glass processing has a significant advantage over direct heating methods. Unlike the commercial glasses, waste glass has high FeO and FeO_2 contents that make the glass a highly absorbing medium, meaning, from a glass-mixing point of view, that glass temperature uniformity is much harder to achieve. Therefore the electric resistivity and convection current control is the key operational strategy in waste glass processing.

Cold Cap Melting

More than half the waste-melter slurry feed is water; and during continuous melter feed operation, most water evaporates. The remainder piles up on top of the melt pool covering about 90 percent of the surface area. This feed pile, called "cold cap", continuously releases residue water vapor and various organic gases into the plenum. At the bottom of the cold cap, the solid waste frit continuously melts into molten glass. Between top and bottom, a transition layer exists where solid-to-liquid phase changes take place and, also, various chemical reactions.

The chemical reaction models are very complicated, as there are more than forty different chemical species in a typical waste-melter feed composition; thus, Arrhenius type of equations may not be applicable. From a thermal modeling point of view, however, these chemical reactions essentially provide heat sources. The amount of mass transfer of volatile and foaming gases through the cold cap is small and may be neglected; however, the effects on the cold-cap heat balance of these gases can be substantial.

In the commercial glass industry, several batch modeling studies[11,12] were conducted wherein the batch was dry, horizontally charged, and melting characteristics were determined by batch geometry and charging speed. However, in the waste melter, the cold-cap melt rate was small and

there was no appreciable movement within it except in the bottom interface with the molten glass. Therefore, a first-order cold cap model can be developed with an overall melter system energy balance alone.

Even with simplifications, the first-order cold cap thermal model involves a number of unknown parameters, the major ones being radiation flux from plenum heaters, forced convection by steam and gases in the plenum, and the heat transfer rate from the molten glass at the bottom. Therefore, the primary requirement for developing a better cold cap model is to determine these parameters. Sub-models that can be developed for this purpose include a radiation model for the plenum, a turbulence model for plenum steam and gases, and a diffusion model for glass melting. In addition to cold cap dimensions, the heat transfer model will involve, feed rate, melt rate, plenum heater temperature, glass-pool bulk temperature, steam enthalpy, and the heat transfer coefficient at the bottom interface. Because the cold cap has a combination of different phases, it should be treated as a composite material with different conductivities. Output of the cold cap model is temperature distribution.

One of the cold cap sub models involves the solid-to-liquid phase change, which may be treated as a Stephan type problem[13] where enthalpy changes and even natural convection at the solid-to-liquid interface can be included. A solution can be useful for estimating the extent of the glass-melt model boundary and the boundary conditions. Considering these assumptions, the conservation equation for the number of noble metal particles per unit volume and per unit size interval can be expressed as:

$$\frac{\partial}{\partial x}(uN)+\frac{\partial}{\partial y}(vN)+\frac{\partial}{\partial z}[(w-w_p)\frac{\partial N}{\partial z}]=\frac{\partial}{\partial x}(D\frac{\partial N}{\partial x})+\frac{\partial}{\partial y}(D\frac{\partial N}{\partial y})+\frac{\partial}{\partial z}(D\frac{\partial N}{\partial z})-\frac{\partial}{\partial s}(SN) \qquad (6)$$

where u,v, and w represent glass velocities and w_p is the relative velocity of noble metal particles in the vertical direction. In the last term on the right side of equation (6), s is the size of the particle (i.e., radius or square of the radius) and S is the rate of change of particle size, which can also be interpreted as velocity of the particle in geometrical size space. The first three terms on the right represent diffusion of the particle within the glass bath. Their contribution is usually so small in laminar flows that they are neglected. The utility of equation (6) is enhanced if one recognizes that:

$$NM = m\rho \qquad (7)$$

where M is the mass of a single particle. In accordance with the assumption that mass is a function of a size parameter only, the symbol m denotes the mass fraction of grains per unit size interval. Substituting equation (7) into equation (6) and rearranging the terms, yields:

$$\frac{\partial}{\partial x}(\rho um)+\frac{\partial}{\partial y}(\rho vm)+\frac{\partial}{\partial z}[\rho(w-w_p)m]=-M\frac{\partial}{\partial s}(\dot{S}\frac{\rho m}{M}) \qquad (8)$$

Dissolution of a noble metal particle may be expressed by the following size change equation[14].

$$\dot{S}=\frac{ds}{dt}=\frac{d}{dt}(r^2)=2[\frac{D}{\rho}(C_\infty - C_1)] \qquad (9)$$

The geometrical size is characterized by the square of the particle radius, C_∞ is the noble metal concentration in the bulk glass, and C_1 is the liquid-grain interface at equilibrium. If the range is

divided into a finite set of intervals numbered 1,2...,n-1,n,n+1,.,k, the average values in the intervals can be expressed as:

$$\overline{m}_R = \int^{s_+} \frac{m ds}{s_+ - s_-} \tag{10}$$

where s_+ and s_- are the size parameters at the boundaries of interval n in Figure 3; and because

$$M = f[s(r^2)]^{3/2} \tag{11}$$

using integration-by-parts, the right side of equation (8) can be expressed as:

$$\int^{s_+} M \frac{\partial}{\partial s}\left(\frac{S\rho m}{M}\right) ds = \rho\{(\dot{S}m)_{s_-} - (\dot{S}m)_{s_+} + \frac{3}{2}\int^{s_+} \frac{Sm}{s} ds\} \tag{12}$$

as S does not depend on s [equation (9)]. e q. (12) can be rewritten as:

$$\int^{s_+} -M \frac{\partial}{\partial s}\left(\frac{S\rho m}{M}\right) ds = \rho \dot{S}\{m_{s_-} - m_{s_+} + \frac{3}{2}\int^{s_+} \frac{m}{s} ds\} \tag{13}$$

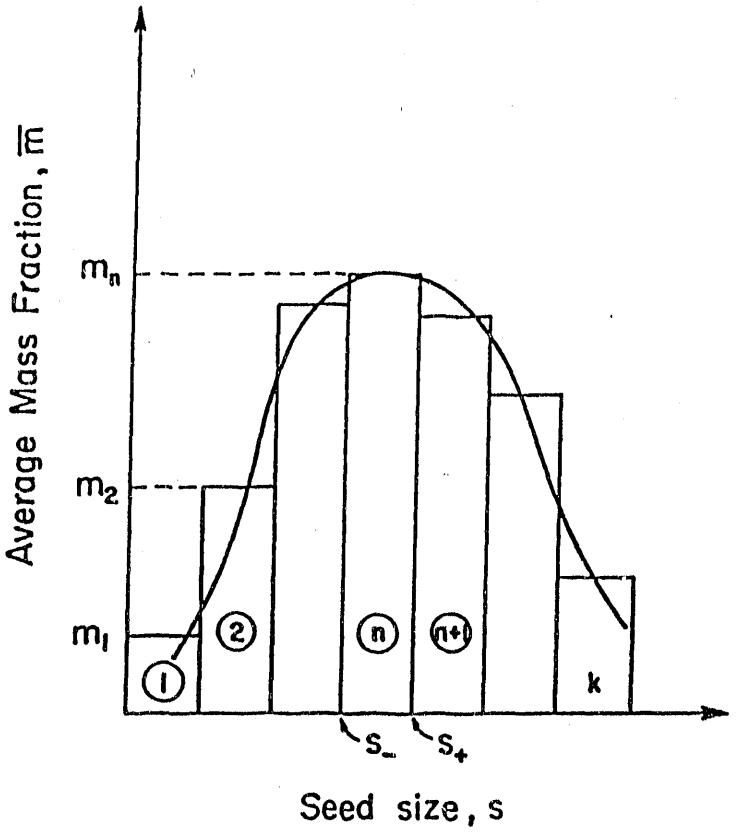

Figure 3. Approximate Noble Metal Size Distribution Within Waste Glass Melt

From Eqs. (14), (15), and (16), it is shown that the average mass fraction of noble particles in interval n can be found from:

$$\frac{\partial}{\partial x}(\rho u\overline{m}_n)+\frac{\partial}{\partial y}(\rho v\overline{m}_n)+\frac{\partial}{\partial z}[\rho(w-w_p)\overline{m}_n]=\frac{\rho\dot{S}}{(s_+-s_-)}\{\overline{m}_{s_-}-\overline{m}_{s_+}+\frac{3}{4}\int^{s_+}\overline{m}_s ds\} \quad (14)$$

Although the obvious approximations for m_{s-} and m_{s+} are

$$m_{s_-}=\frac{1}{2}(m_{n-1}+m_n) \quad (15a)$$

$$m_{s_+}=\frac{1}{2}(m_n+m_{n+1}) \quad (15b)$$

such formulations would lead to numerical oscillations and physically unrealistic results; therefore, a scheme akin to the upwind differencing scheme[15] may be employed. Accordingly, as noble particles are dissolving (i.e., S< 0), the following formulations are assumed to be appropriate.

$$\overline{m}_{s_-}=\overline{m}_R \quad (16a)$$

$$\overline{m}_{s_+}=\overline{m}_{n+1} \quad (16b)$$

Substituting the eqs. (16a) and (16b) into equation (14) and arranging the terms yields:

$$\frac{\partial}{\partial x}(\rho u\overline{m}_n)+\frac{\partial}{\partial y}(\rho v\overline{m}_n)+\frac{\partial}{\partial z}[\rho(w-w_p)\overline{m}_n]=\rho\dot{S}[\frac{\overline{m}_n-\overline{m}_{n+1}}{s_+-s_-}+\frac{3\overline{m}_n}{s_++s_-}] \quad (17)$$

Of course, there are as many equations of this type as there are intervals (Figure 2), and they are coupled to each other by the source (last) terms.

The mathematical model predicts noble metal size distribution within the glass pool and bottom sedimentation. This information may be augumented by the streaklines of noble metal particles (i.e. the path traced by particles in the glass pool). Streaklines are computed by starting at a point in the cold cap and calculating the subsequent point after a small time interval using:

$$\frac{D\bar{r}}{Dt}=\bar{u}_p \quad (18)$$

where r is the position vector and u_p is the velocity vector of the particle.

CONCLUSIONS

Numerical simulation techniques are proven to be a powerful tool in design and development of commercial glass furnaces and are expected to find the same utility in waste-glass melters. The purpose of this study was to facilitate these expectations and to channel efforts towards designing and developing safe waste glass melters. Although powerful, numerical models have some

limitation, their success largely depends on the availability of relevant empirical data, the vision of the model designer, and interpretation of the results by the user/furnace engineers. Usually some iterations among these three are necessary to fill the blank, or difficult, areas.

NOMENCLATURE

C_C	electric capacitance	$\rightarrow r$	position vector of a noble metal particle
C_l	concentration glass-noble metal interface at equilibrium	$\dot{S}$	size growth rate
C_∞	concentration of noble materials in the bulk glass	s	size of a particle (radius square)
C_p	heat capacity	s_+, s_-	particle size at the boundaries of band n
E	voltage	T	temperature
g	gravity	T_o	reference temperature
I	current density per unit length	t	time
k_E	electric conductivity	u	velocity vector
k_T	thermal conductivity	x,y,z	axial leteral vertical velocity components
M	mass of single noble metal particle	.	scalar product
$\overline{m}_n, \overline{m}_{n+1}$	average mass fraction at band n	β	thermal expansion coefficient
m_{S+}, m_{S-}	Mass fractions at the boundaries of a band	Δ	vector operator
N	number density	μ	viscosity
p	pressure	ρ	density
r	radius of a noble metal particle	ρ_o	density at reference temp

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REFERENCES

1. McElroy, J. L., W. J. Bjorklund, and W. F. Bonner, "Waste Vitrification:..A Historical Perspective", The Treatment and Handling of Radioactive Wastes, 1983.

2. Daken, R. B., C. C. Chapman, and A. K. Postma, "Ceramic Electrode Heated Melter" Quaterly Progress Report, Research and Development Activities, Waste Fixation Program, Compiler A. M. Platt, BNWL-1809, Pacific Northwest Laboratory, Richland, WA, 1973.

3. Bradley, R. F., "Savannah River Waste Vitrification Program" Waste Management '79, University of Arizona, Tucson, Arizona, 1979.

4. Nolet, D. A.and R. A. Murnane, "Development of Modeling Techniques for Glass Furnaces", Advances in the Fusion of Glass, 1988.

5. Ungan, A., "Three Dimensional Numerical Modeling of Glass Melting Process", Ph.D. Thesis, Purdue University, West Lafayette, IN, 1985.

6. Rawson, H., "Mathematical Modeling", J. Non-Crystalline Solids, Vol. 73, 1985.

7. Eyler, L. L., et . al., "Physical and Numerical Modeling of Joule Heated Melters", PNL-5491, Pacific Northwest Laboratory, Richland, WA, 19985.

8. Trent, D.S. and L. L. Eyler, "TEMPEST:..A Three Dimensional Time Dependent Computer Program for Hydro-Thermal Analysis",Vol. 1:..Numerical Methods and Inout Instructions", PNL-4348, Vol. 1, Rev. 2, Pacific Northwest Laboratory, Richland, WA, 1989.

9. FIDAP Ver. 5.01 Manual, Fluid Dynamics International, Evanston, IL, 1990.

10. Bickford, D. F., et . al., "Control of Radioactive Waste Glass Melters", Part 3 - Glass Electrical Stability, 1989.

11. Mase, H. and K. Oda, "Mathematical Model at Glass Tank Treemace with Batch Melting Process", J. Non-Crystalline Solids, Vol. 38/39, 1980.

12. Ungan, A. and R. Viskanta, "Melting Behavior of Continuously Charged Loose Batch Blanckets in Glass Melting Furnances", Glastechn. Ber., Vol 59, No. 10, 1986.

13. Dantzig, J. A. , "Mathematical Modeling of Solidification Processes", Symposium on Interdisciplinary Issues in Materials Processing and Manufacturing", ASME Winter Annual Meeting, Boston, MA, 1989.

14. Gamm, H. D., "Mass Transfer Problems in Glass Furnance Simulation", Sc.D. Thesis, M.I.T., Cambridge, Massachusetts, 1975.

15. Patankar, S. V., Numerical Heat Transfer and Fluid Flow, McGraw-Hill, 1980.

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