

DISCRETE AND CONTINUOUS MODELLING OF REACTIVE PELLETS AND GAS TRANSPORT IN PYROMETALLURGICAL BATHS

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ABSTRACT

In many pyrometallurgical processes, reactive pellets are added to a melt. Heat is transferred to the pellets and reaction products such as gases, metal and slag are formed. Many traditional grid based CFD methods, such as the Finite Element Method (FEM) and Finite Volume (or Control Volume methods), produce suitable predictions. However, the inclusion of solids in the bath presents significant challenges for these methods. The formation and transport of bubbles in the melt and/or slag also creates similar problems. Smoothed Particle Hydrodynamics (SPH), a Lagrangian simulation method, is able to simulate all three phases; the fluid component of the bath, any immersed solid materials including the tracking of their motion and their interaction with the fluid, and gas generation and transport. The use of coupled discrete bubbles can enable more realistic modelling of the gas phase transport. In this paper, we summarize the key aspects of the SPH method and show a series of examples illustrating the behaviour of the different physics sub-systems that are used in the overall pyrometallurgical model.

INTRODUCTION

The use of computational fluid dynamics (CFD) to model pyrometallurgical processes has traditionally been by grid based finite element or finite volume methods. These simulation tools are quite useful in predicting the overall flow patterns and heat transfer in the molten bath. However, in many pyrometallurgical processes reactive solids are added to the molten bath in the form of pellets or briquettes. Heat is transferred from the bath into the porous solids and reaction proceeds with possible metal, slag and gaseous products being formed depending on the specific chemistry of the system. For example, in the carbothermic reduction of iron oxide pellets in slag, molten oxides, carburized iron droplets and a carbon monoxide rich gas are formed (Street et al., 1998). It is computationally prohibitive to simulate the individual pellets using grid-based CFD methods (Johannessen et al., 2001). For grid-based methods one would also need to incorporate complicated mesh deformations to account for the interaction between the fluid and solid pellets.

The modelling of the gas phase transport also represents a substantial challenge for grid based methods. The traditional approach is to assume that the gas phase can be approximated as a continuous inter-penetrating fluid phase. In many circumstances this is a reasonable approach. But if the superficial gas velocities become large then numerical diffusion problems can emerge

arising from the grid based solution of the gas advection. If the gas phase consists of bubbles of sufficient size then the assumption of a continuous gas phase is also no longer reasonable. This assumption is also not reasonable if some of the physical processes are inherently discrete and occur on the bubble scale. In these cases, it is necessary to model the bubbles as discrete entities interacting with both the melt and with any solids in the bath.

Smoothed Particle Hydrodynamics (SPH) is a Lagrangian (grid-free) simulation method that has been used extensively to model fluid flows with heat transfer for different applications such as high pressure die casting (Cleary and Ha, 2003), ingot casting of aluminum (Cleary et al., 2004), geophysical flows (Cleary and Prakash, 2004) and modelling of bio-pumps (Prakash et al., 2003). Due to its grid-free nature it has the following advantages over other CFD tools:

- Free surface flows can be handled easily and accurately.
- Solid/fluid interactions with complicated motions of the solid parts can be simulated with few additional computational penalties.
- Complicated physics can be added easily (examples include non-Newtonian rheology, solidification, freezing, solid stress and history dependence of fluid variables).
- Gas phases can be modelled using either an advected Eulerian approach or by inclusion of discrete bubbles.

Due to these inherent advantages, this method is well suited to modelling pyrometallurgical processes involved in a molten phase bath, discrete reactive solids and gas phases. This paper presents a summary of the key aspects of the SPH methodology with the required extensions. A series of examples illustrating the physics sub-systems incorporated are presented.

THE SPH METHOD

SPH Interpolation

The basic SPH method is given by Monaghan (1992, 1994). A general review of SPH is also given in Monaghan (2005). Basically, the interpolated value of a function A at any position \mathbf{r} can be expressed using SPH smoothing as:

$$A(\mathbf{r}) = \sum_b m_b \frac{A_b}{\rho_b} W(\mathbf{r} - \mathbf{r}_b, h), \quad (1)$$

where m_b and \mathbf{r}_b are the mass and density of particle b and the sum is over all particles b within a radius $2h$ of \mathbf{r} . Here

$W(\mathbf{r},h)$ is a C^2 spline based interpolation or smoothing kernel with radius $2h$ that approximates the shape of a Gaussian function. The gradient of the function A is given by differentiating the interpolation equation (1) to give:

$$\nabla A(\mathbf{r}) = \sum_b m_b \frac{A_b}{\rho_b} \nabla W(\mathbf{r} - \mathbf{r}_b, h). \quad (2)$$

Using these interpolation formulae and suitable finite difference approximations for second order derivatives, one is able to convert parabolic partial differential equations into ordinary differential equations for the motion of the particles and the rates of change of their properties.

Continuity Equation

From Monaghan (1992), the SPH continuity equation is:

$$\frac{d\rho_a}{dt} = \sum_b m_b (\mathbf{v}_a - \mathbf{v}_b) \cdot \nabla W_{ab} \quad (3)$$

We denote the position vector from particle b to particle a by $\mathbf{r}_{ab} = \mathbf{r}_a - \mathbf{r}_b$ and let $W_{ab} = W(\mathbf{r}_{ab}, h)$ be the interpolation kernel with smoothing length h evaluated for the distance $|\mathbf{r}_{ab}|$. This form of the continuity equation is Galilean invariant (since the positions and velocities appear only as differences), has good numerical conservation properties and is not affected by free surfaces or density discontinuities. The use of this form of the continuity equation is very important for predicting free surface flows of the present kind.

Momentum Equation

The momentum equation then becomes the acceleration for each particle:

$$\frac{d\mathbf{v}_a}{dt} = \mathbf{g} - \sum_b m_b \left[\left(\frac{P_b}{\rho_b^2} + \frac{P_a}{\rho_a^2} \right) - \frac{\xi}{\rho_a \rho_b} \frac{4\mu_a \mu_b}{(\mu_a + \mu_b)} \frac{\mathbf{v}_{ab} \cdot \mathbf{r}_{ab}}{\mathbf{r}_{ab}^2 + \eta^2} \right] \nabla_a W_{ab} \quad (4)$$

where P_a and μ_a are pressure and viscosity of particle a and $\mathbf{v}_{ab} = \mathbf{v}_a - \mathbf{v}_b$. Here ξ is a factor associated with the viscous term (Cleary, 1996), η is a small parameter used to smooth out the singularity at $\mathbf{r}_{ab} = 0$ and \mathbf{g} is the gravity vector.

Equation of State

Since the SPH method used here is quasi-compressible one needs to use an equation of state, giving the relationship between particle density and fluid pressure. A suitable one is:

$$P = P_0 \left[\left(\frac{\rho}{\rho_0} \right)^\gamma - 1 \right] \quad (5)$$

where P_0 is the magnitude of the pressure and ρ_0 is the reference density. For water or molten phases we use $\gamma = 7$. This pressure is then used in the SPH momentum equation (3) to give the particle motion.

Energy Equation

The SPH heat equation is based on the internal energy one developed in Cleary and Monaghan (1999), but modified to use an enthalpy formulation for solidifying metals (see Cleary et al., 1998) and including contributions from viscous heating.

$$\frac{dH_a}{dt} = \sum_b \frac{4m_b}{\rho_a \rho_b} \frac{k_a k_b}{k_a + k_b} T_{ab} \frac{\mathbf{r}_{ab} \cdot \nabla_a W_{ab}}{(\mathbf{r}_{ab}^2 + \eta^2)^2} \quad (7)$$

where the summation is the heat conduction term. The enthalpy per unit mass is defined by:

$$H = \int_0^T c_p(\theta) d\theta + L[1 - f_s(T)] \quad (8)$$

where c_p is the temperature dependent specific heat, L is the latent heat and $f_s(T)$ is the volume fraction of material that is solid at temperature T , k_b is the conductivity, and $T_{ab} = T_a - T_b$.

Equation (7) has an explicit conductivity which can be temperature dependent and ensures that heat flux is automatically continuous across material interfaces, such as between a die and a liquid metal. This allows multiple materials with substantially different conductivities and specific heats to be accurately simulated.

Model for Reactive Solids in Fluid

The discrete solids are modelled by connected assemblies of SPH particles. The solid SPH particles are used in the solution of the continuity, momentum and heat equations in the normal way. This gives forces on each particle making up the solids. The net force and net torque on the overall discrete solid is calculated using the instantaneous point forces on each part of the solid. Newton's equations of motion are then solved for the linear and rotational motion of the solid.

There are no continuous approximations used in this formulation and there is no dependence on empirical drag laws. This model relies on calculating the detailed flow field around each solid body. The interaction between the solids and the fluid arise from the fundamental fluid forces of pressure and viscous stress generated on the surface of the solids. This is a feasible approach for up to tens of thousands of reasonable sized solid particles, but not for very fine ones or ones existing in larger numbers.

The ability to model immersed discrete solids in this manner is one of the key strengths of SPH because it allows prediction of the detailed coupled flow field and has no requirement for expensive and diffusive re-meshing because there is no mesh in SPH that needs to be updated to exactly surround the solids. The SPH fluid particles automatically do this.

Continuum Model for Gas Transport

If the gas phase consists of reasonable volumes of small bubbles in a liquid, then it is reasonable to represent the gas as an inter-penetrating fluid. The two separate phases, treated as continua, have their own momentum and mass conservation laws and interact through a drag force that each exerts on the other. This is an extremely common approach in traditional Eulerian CFD modelling of multiphase systems. Here we do something quite similar but with a key difference relating to the Lagrangian nature of the SPH particles. In the traditional Eulerian solution, the gas phase is solved using the full advection velocity of the gas which can be quite large leading to potential artificial diffusion. Here an SPH particle represents a combination of fluid and gas. The SPH particles are automatically advected with mixture speed. The differential gas transport is obtained by solving an advection-diffusion equation on the SPH particles (when normally we solve only a diffusion equation) but where the advection velocity is the superficial speed of the gas

compared to the liquid. This will be typically smaller than the absolute gas speed and so the numerical diffusion resulting in the SPH solution will be smaller than for the purely Eulerian solution.

We use the subscript l for the liquid phase and g for the gas phase. A critical variable is the gas volume fraction ε . In terms of its effect on liquid flow we assume the density of the gas is zero. The density of the liquid here is ρ . As usual v represents the velocity, while η is the viscous stress in the liquid (assumed unaffected by the gas). The model uses the speed of sound in liquid c , the liquid viscosity μ , and the average bubble diameter d . The assumption that the gas has zero density is appropriate for these applications which has gas in a dense liquid. We also assume that the gas is incompressible since hydrostatic pressure will cause only small density variations. The gas conservation law is then just an advection equation:

$$\varepsilon_l + v_g \cdot \nabla \varepsilon + \varepsilon \nabla \cdot v_g = 0 \quad (9)$$

The conservation law for the liquid can be written as:

$$\varepsilon_l + v_l \cdot \nabla \varepsilon = \frac{1}{\rho c^2} P_t + \nabla \cdot v_l \quad (10)$$

The subscript t refers to a time derivative. Without gas the terms on the left would be zero; the terms on the left represent the change in gas volume for which there is an equal change in liquid volume fraction. The momentum or force balance term for the gas can be simplified to:

$$H(v_g - v_l) + \nabla P = 0 \quad (11)$$

Here H is the inter-phase drag term and P the pressure. In this preliminary implementation there was no attempt made to model the effect of bubble size. Instead a constant drag factor H was assumed for the calculations which gave a superficial bubble rise velocity of around 1 m/s. More sophisticated representation of the gas phase and of the inter-phase drag are well known in the Eulerian literature and can be used here without change.

The momentum equation for the liquid can be written as:

$$\rho(v_l)_t + \rho v_l \cdot \nabla(v_l) = (1 - \varepsilon)(\rho g - \nabla P) + \nabla \cdot (\tau_l) \quad (12)$$

Discrete Model for Gas Transport – Bubbles

Gas-liquid models have the simplifying feature that the momentum of the gas is quite small. However, unlike solid-liquid flows, the actual size of the bubbles is often poorly known and may change quite radically and quickly in time and space. At higher bubble loadings the effects of bubble aggregation and the variation of shape of the resulting larger bubbles need to be considered. Furthermore, if the bubbles are large compared to other elements of the system or any of the reaction or transport physics occurs on the bubble scale then the discreteness of the gas cannot be easily ignored. In these cases we need to be able to model the gas as discrete entities.

The Discrete Element Method (DEM) is a method used to model the motion and interaction of discrete entities, Cleary (2004). Normally, this is used for discrete solids in circumstances where collisional interactions are the dominant physical process.

Small and medium sized bubbles are relatively well approximated as discrete entities with fixed shapes (most

easily spherical). Here a DEM method is used to model the collisional interaction of discrete spherical bubbles with each other and with other solids and boundaries. The bubbles can be coupled to the liquid phase via an empirical drag law. For these demonstrations we use a simple drag law for isolated bubbles in a large volume of fluid (a dilute approximation) and neglect the effect of the bubble momentum on the liquid phase. The coupling of the gas phase back to liquid phase and the use of drag laws that take account of large volume fractions of bubbles are both relatively straight forward to implement.

The discrete bubbles can also:

1. Split during collision
2. Coalesce during collision
3. Float on the surface to form a foam
4. Burst when in the surface foam
5. Interact with fine particulates in flotation like processes
6. Can be fragmented by the fluid in regions of high shear.
7. Can grow due to absorption of gas from the liquid through which they pass
8. Can be nucleated on surfaces or on suspended solids within the liquid
9. Bubble nucleation can be linked to the amount of dissolved gas locally available at the nucleation sites
10. Dissolved gas can be tracked by solving another diffusion equation on the SPH fluid particles and can be connected to chemical reactions either in the melt or in the immersed reactive solids.

This type of model has significantly more bubble scale physics and allows, in principle, the capturing of many important processes that cannot be directly modelled in traditional Eulerian systems.

Next we provide a series of relatively simple example problems that demonstrate key aspects of the SPH/DEM model described above.

SOLID PELLETS IN A FLUID WITH BUOYANCY

Solid pellets are represented by collections of SPH particles. The forces on these particles are determined in the same way as for fluid particles but with an overriding constraint of rigid-body motion. Buoyancy effects due to the density difference between the pellets and fluid are automatically dealt with by the method. Thermally induced buoyancy effects are small compared to these and are omitted. Equations for heat transfer and species transport are solved using fluid and solid particles with appropriate material properties. Table 1 gives the properties of the fluid and pellet for the cases in the first example.

	Fluid	Pellet
Conductivity (W/mK)	53.0	19.0
Specific Heat (J/kgK)	241.0	1434.0
Density (kg/m ³)	5860.0	1860.0

Table 1: Properties for pellets and fluid.

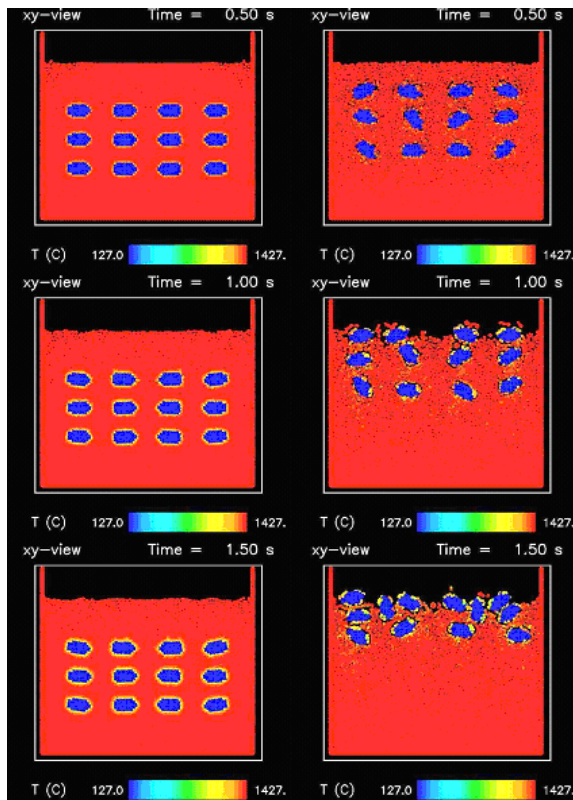


Figure 1: Motion of cold pellets in a hot fluid: (left) neutrally buoyant and (right) strongly buoyant pellets. In both cases we solve a coupled heat transfer equation.

Figure 1 shows the motion of initially submerged pellets for the cases of neutrally buoyant and very light pellets. The liquid bath and the pellets are coloured according to their temperature. When there is no difference in densities for the two materials, the pellets are neutrally buoyant and remain in their original positions throughout the simulation (as they should). This is independent of the inclusion of heat transfer. There is very little movement of the pellets due to convective heat transfer. When the pellets are much less dense than the surrounding liquid bath, they float up to the surface of the fluid at speeds dependent on the density difference between the two materials, (again as they should). The progress of the heat conduction is indicated by the change in colour of the pellet surfaces (from blue to light green and towards yellow) as well as that of the surrounding fluid (from red to yellow in certain regions) as the cold pellets heat up and cool the surrounding fluid. This simple test shows that the method produces qualitatively sensible predictions for pellet motion and heating.

GAS TRANSPORT FROM REACTIVE PELLETS USING THE CONTINUOUS GAS MODEL

Next we enable gas generation at the surfaces of the submerged solids arising from some form of temperature driven chemical reaction. This generates gas which we will model using a continuous gas phase model. The gas is then transported to the surface of the liquid in complex transient plumes. This is shown in two consecutive parts in Figures 2 and 3. The neutrally buoyant case is shown on the left of each of the figures and the positively buoyant pellet case is shown in the right side. The fluid is coloured by gas fraction with blue corresponding to no gas

and red being a volume fraction of 25% gas. The generation of gas and its transport through the liquid bath is clearly visible as transient plumes rising from the surfaces of the reactive pellets.

For the neutrally buoyant case, the gas motion through the liquid bath reduces the effective density and makes the liquid buoyant. The combination of this buoyancy and the gas motion around and above each pellet creates a buoyant plume that entrains fluid which in turn pushes up the pellet. The resulting updrafts are strong enough to lift the neutrally buoyant particulates to the surface. The combination of the buoyant plumes produces two weak bulk recirculation patterns in the fluid.

When the pellets are lighter than the liquid bath (right column of Figures 2 and 3), then the recirculation is much less noticeable and there is no coherent fluid flow. The buoyant pellets simply float and cluster at the surface. The generation of the buoyant gas plumes and the entrainment of fluid enhances the natural buoyancy of the pellets causing them to rise faster. Note that the gas generation is far from uniform on the surfaces of the pellets. The shape of the particulate strongly affects the local heat transfer to it and therefore the local rate of reaction and therefore the rate of gas generation. Corners are more exposed to the hot fluid and so heat up faster. For many of the particulates shown the gas generation is preferentially from these corners. This creates separate gas plumes each of which produces a torque on the particulate which then spins in response to the balance of the gas generated torques. Similarly, parts of the pellets which protrude above the bath surface are no longer able to be heated in these areas and cease to react on these surfaces.

Figure 4 shows an analogous rectangular tank in 3D. The colour on the pellet surface again represents the volume fraction of gas being generated there. In the liquid bath, the gas distribution is represented by spheres whose size and colour show increasing gas fraction. The larger the sphere the higher is the local volume fraction of gas. This is a discrete graphical representation but the underlying solution remains continuous. The flow behaviour predicted in 3D is broadly similar to that shown in 2D.

These examples indicate that SPH is able to predict at least qualitatively, gas generation from pellets with adequate representation of buoyancy effects and gas transport through the liquid bath, including fluid convection generated by gas plumes rising from the pellets. This system is quite complex and has an important five way coupling, where the heat transfer controls gas generation via chemical reactions which generates gas phase plumes which produce buoyancy driven motion that move the particulates around. This in turn affects the local rates of heat transfer and the loop is closed. This model captures many of the key processes involved in pyrometallurgy at the scale of the particulates giving a much more detailed ability to understand the specific dynamics occurring.

GAS TRANSPORT FROM REACTIVE PELLETS USING A DISCRETE BUBBLE MODEL

Figure 5 shows the transport of discrete gas bubbles from the bottom of a tank. Again there is a positively buoyant

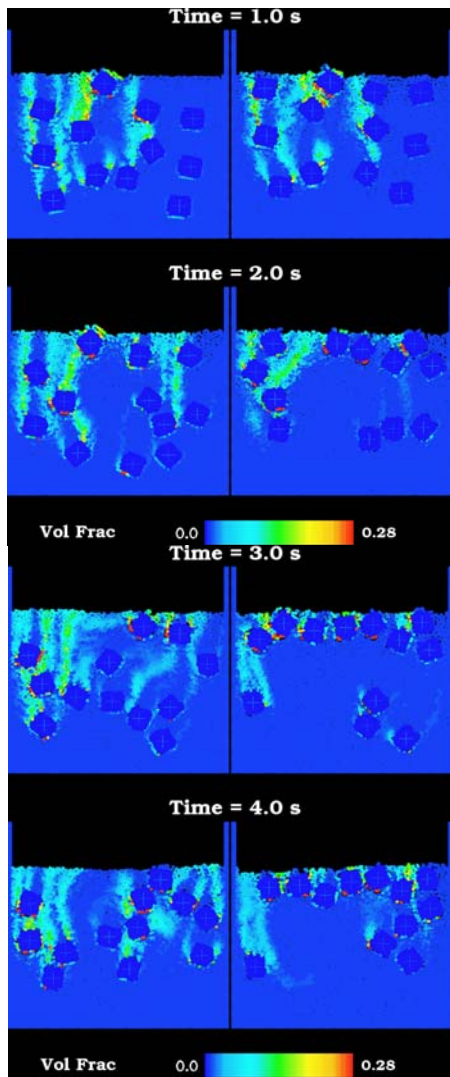


Figure 2: First half of an example of gas generation and its transport in 2D using a continuous gas model: (left) pellet and liquid bath have the same density, and (right) pellets are significantly lighter than the bath.

solid body floating towards the fluid surface and interacting with the bubbles. The bubbles are strongly buoyant and are coupled to the fluid motion via a suitable drag term. They rise to the surface, colliding with each other, the walls and the floating body. At the surface they cluster to form a foam layer on top of the bath. The continuous supply of bubbles from the bottom leads to a steady increase in the thickness of the foam which is dependent on the balance between the rate of bubble arrival and the rate of bursting. The bubbles can coalesce or split during collision and can be sheared by the fluid.

Bubble generation can occur wherever it is needed. If the gas is dissolved in the bath then the advection-diffusion of this dissolved gas to the bubble nucleation sites can also be solved for. If the gas is generated at the surface of reactive solid bodies, then the bubble nucleation can occur directly on the surface in accordance with the amount of gas locally available. If there is dissolved gas in the bath, then the bubbles can also grow by diffusion from adjacent liquid as the bubbles pass through. Gas generation rates can again be driven by the chemistry and heat transfer.

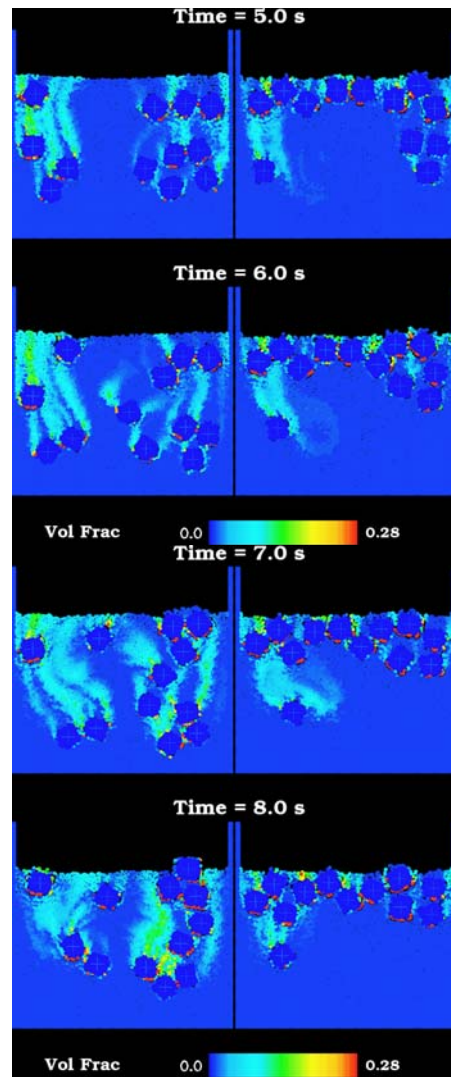


Figure 3: Second half of an example of gas generation and its transport in 2D using a continuous gas model: (left) pellet and liquid bath have the same density, and (right) pellets are significantly lighter than the bath.

CONCLUSION

SPH is a versatile numerical method that is well suited to modelling pyrometallurgical systems. In particular it is well able to:

- Simulate solid particulates/pellets interacting with fluid including buoyancy effects;
- Model gas generation with real reaction chemistry in the solid pellets including heat transfer effects;
- Predict the convective and diffusive transport of gas from the pellets to the fluid free surface using a continuous gas model in realistic transient plumes;
- Predict nucleation of discrete bubbles from solid surfaces based on dissolved gas availability
- Predict discrete bubble motion interacting with fluid and including inter-bubble collision, splitting, and coalescence.
- Simulate formation of foams on the fluid surface consisting of assemblies of sticky discrete bubbles and including bursting of these bubbles.

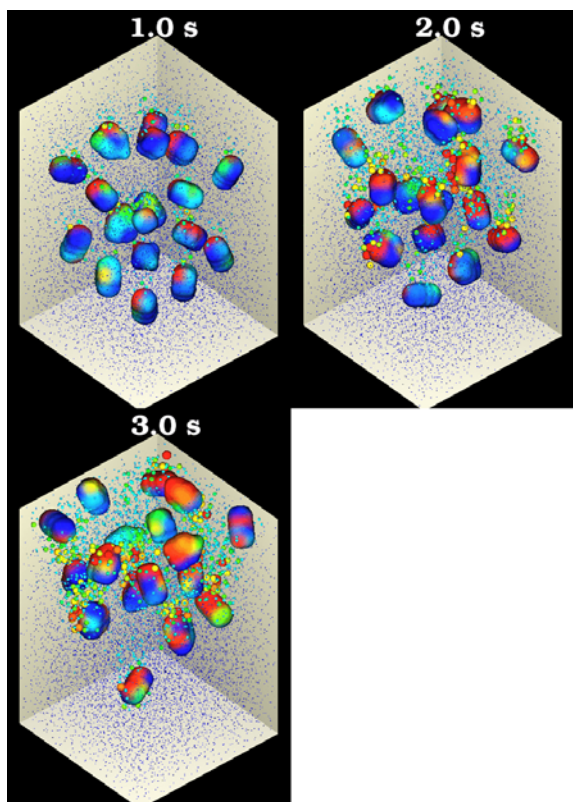


Figure 4: Gas generation and transport in 3D using a continuous gas model. The pellets are much less dense than liquid bath.

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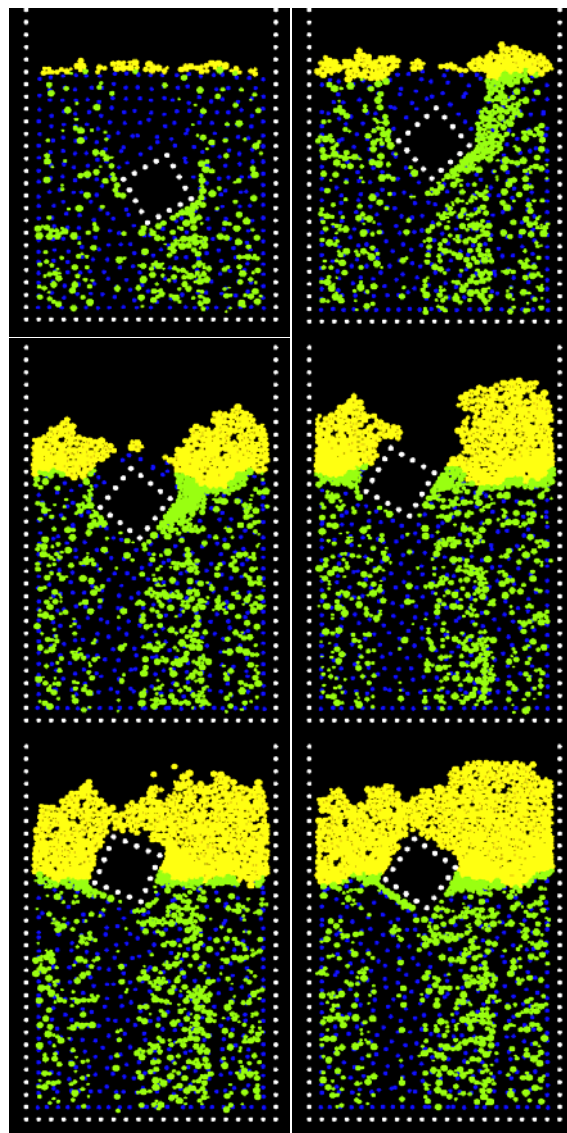


Figure 5: Gas transport (in 2D) through the liquid bath using the discrete bubble model. The blue particles are the background SPH fluid. The green particles are submerged bubbles and the yellow ones are bubbles above the fluid free surface.

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