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Densification and Shape Change of Calcined High Level Waste during Hot-Isostatic Pressing

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Abstract: Hot Isostatic Pressing (HIP) has been selected as the means of treating calcined high level waste (HLW) for the Calcine Disposition Project. The HIP process combines high temperature and pressure to consolidate powder metals and ceramics into a monolithic densified structure. This study uses COMSOL Multiphysics to simultaneously predict temperature, densification, and shape deformation of a stainless steel can filled with HLW. Once in the HIP machine, the can is heated and compressed using Argon gas. At the specified temperature and pressure, densification of the HLW occurs and the HLW turns in to a mineral similar to the geologic formulation of granite. Two approaches were used to model densification of HLW during the HIP process including: development of a volumetric thermal change coefficient implemented through thermal expansion and the development of an ordinary differential equation to approximate deformation mechanics. Preliminary models suggest that COMSOL can be used to predict densification and shape change as indicated in model results which predicts HIP can shape change and temperature change similar to experimental test results. With initial models resulting in the successful reproduction of experimental data, it is anticipated that COMSOL will be a valuable tool in the successful design of a HLW HIP can.

Keywords: Densification, shape change, structural mechanics, hot isostatic pressing.

1. Introduction

Hot isostatic pressing is generally used to reduce porosity and influence density of metal and ceramic materials for manufacturing processes including the consolidation of powder metals and ceramic composites. The HIP process has been selected as the means for treating HLW calcine. During the HIP process, HLW material placed inside a steel HIP can is subjected to a high temperature, high pressure environment. The high temperature and pressure applied to the HIP can causes the can to compress and the granular calcine inside to densify. The process results in mineralization of the calcine into a material similar to the geologic formulation of granite. The consolidated HIP can is then cooled and prepared for shipment to a permanent off-site repository.

Calcine behavior during the HIP process is driven by a complex interaction of multiple physics phenomena, including structural mechanics with large deformations, heat transfer, and material volume change due to thermal effects and densification.

This paper presents two methods used for modeling the densification of calcine during the HIP process. The first approach develops a volume change coefficient implemented in COMSOL in place of the thermal expansion coefficient. The second approach uses the ODE Multiphysics to implement densification equations, as developed by Ashby and others, to model the powder densification mechanisms including plastic yielding, diffusion, creep and grain growth.

2. Model Approach

Figure 1 is a free body diagram representation of a steel can filled with Calcine during the HIP process.

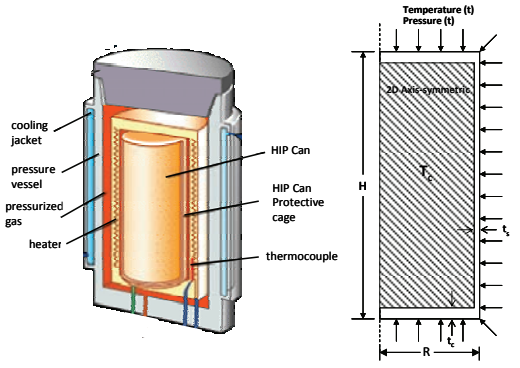

Figure 1. Representation of HIP Can filled with Calcine for COMSOL Analysis.

The free body diagram represents a 2D axis-symmetric, straight-walled HIP can. The can is at an initial temperature (T_c) and subject to the temperature and pressure profile shown in Figure 2. The can is of height, H, with a wall thickness, t_s and end-cap thickness of t_c. This basic set-up was used for both densification modeling techniques.

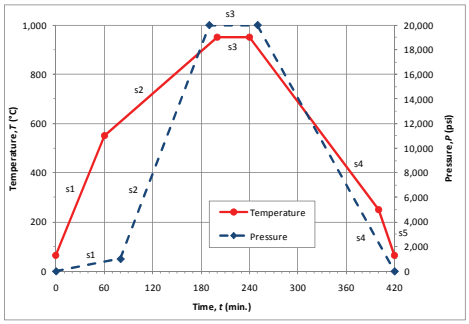

Figure 2. Time-Temperature-Pressure profile for Densification Analysis.

It is important to note that the structural performance of the steel can is not being investigated in the COMSOL models presently. The surrogate calcine material used in the densification and heat transfer model was placed in a basic steel can in order to understand model coupling and deformation characteristics based on the additional structural integrity of a steel shell.

3. Densification Modeled by Volume Change Coefficient

Densification equations are used to describe the relationship between densification rate and volume shrinkage. However, the driving mechanisms behind the densification of bulk powders are difficult to model accurately without knowing very specific material properties and particulate geometric parameters at the molecular level. Effectively modeling the three-dimensional shrinkage of material and the evolution of its density with time as it is HIPped is a difficult task. Usually identified by the term densification rate, this variable is a function of pore and particle size, microstructure, packing density and geometric shape. In order to accurately model the can behavior during the HIP process, a correct representation of the can contents is imperative. Due to the lack of material property information, modeling of this densification behavior has been approximated through the use of equations of state.

Equations of state are often used to describe the volumetric properties of amorphous polymers as a function of temperature and pressure. Equations such as the Tait equation or the Spencer-Gilmore equations provide a valuable first-level approximation of such volumetric properties. While these equations of state can be quite accurate for a molten polymer, volume characteristics in the glassy-state are extremely influenced by formation history and not only the current temperature and pressure. As such, this method may be inadequate if higher degrees of accuracy are required [5].

The method below is a simplified approach at modeling the densification process based on volume shrinkage as defined by a transition from a bulk solids density to the density of a low-porosity, densely-packed material [3]. Equations of state were used to derive an equation for the thermal expansion coefficient as a function of temperature and pressure. This coefficient is then used in COMSOL to represent a temperature and pressure dependent volume change.

The equation for the linear volume change in terms of temperature, pressure and density was derived to be the following:

$$\alpha_L = \frac{\left(\frac{\rho_0}{\rho} + \beta\Delta P\right)^{\frac{1}{3}} - 1}{\Delta T}$$

Where ρ and ρ_0 are the final and initial material density, β is the material compressibility coefficient, Δp is the localized change in pressure and ΔT is the localized change in temperature.

For the initial simulation, the compressibility term was assumed to be negligible until sufficient compressibility data could be obtained. Setting the compressibility term (β) equal to zero resulted in the volumetric expansion coefficient used for this study:

$$\alpha_L = \frac{\left(\frac{\rho_0}{\rho}\right)^{\frac{1}{3}} - 1}{\Delta T}$$

3.1 Modeling Volume Change Coefficient using COMSOL Multiphysics

To model the densification of HLW calcine using the derived volume change coefficient, the structural mechanics and heat transfer modules of COMSOL were used. The steel can and the HLW calcine were both modeled as linear elastic; however, the HLW calcine linear elastic model included the thermal expansion sub-physics where the volume change coefficient was inserted in place of the expansion coefficient. The calculated volume change coefficient is then used to approximate volumetric shrinkage within the calcine material. The pressure applied on the outside of the steel can causes the can to conform to the deformed shape of the calcine.

3.2 Model Results

The model results produced by COMSOL yield a deformed geometry similar to those produced by experiment HIP tests. **Figure 3** is a von mises stress plot showing the deformed can geometry following the HIP process.

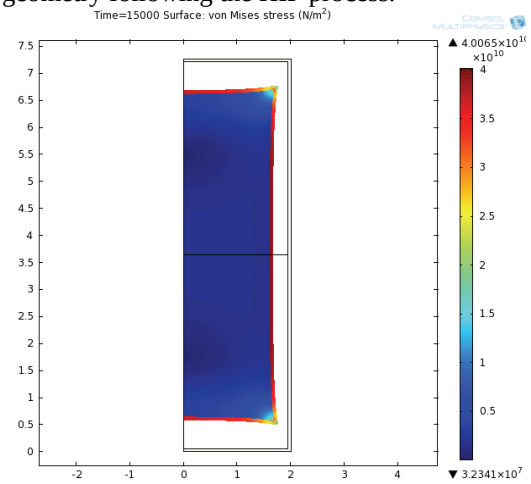

Figure 3. HIP can stress and deformation change using volumetric change coefficient approach

Figure 4 shows modeled and experimental data for the hot isostatic pressing of metal powder [6]. This figure shows similar deformation characteristics as those being produced by COMSOL for the HIP can.

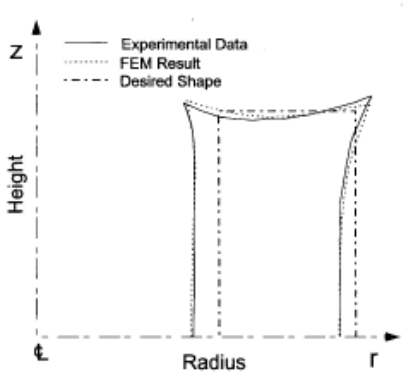

Figure 4. Shape comparisons for 316L stainless steel powder compacts after HIP process

Results from the COMSOL simulation were compared against experimental tests conducted in 1995 by the Idaho National Engineering Laboratory [5].

The goal of the calcine densification model is to predict the final radius of the HIP process. **Figure 5** shows the COMSOL results for the diameter of the HIP can as a function of time compared with the experimental results for can diameter during the experimental HIP tests.

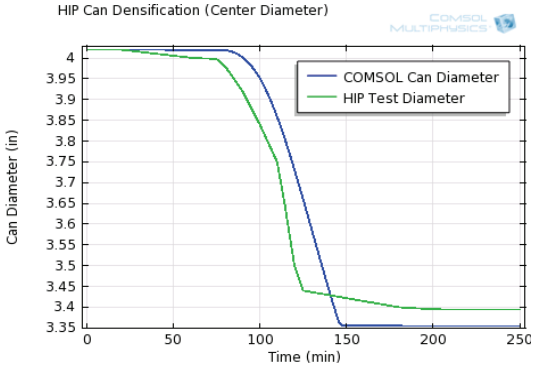

Figure 5. Small cylinder diameter block during HIP simulation vs. experiment HIP test results

The above plots indicate that the COMSOL model is giving a reasonable approximation of the final state for the can radius using the volumetric change coefficient approach. Discrepancies between the two curves are believed to be due to assumptions made during model development including: linear approximation for density as a function of temperature and pressure and assuming material compressibility is negligible.

This model is a prediction for the final state of a process based on well defined material properties. In order to accurately predict can shape, the final expected density based on process temperature and pressure is required.

The accuracy of this simulation could be greatly increased upon the accrual of more accurate material properties, such as a detailed density curve as a function of temperature and pressure.

4. Densification Model Using Deformation Mechanisms

Densification of powders during HIP happens by several mechanisms. The primary mechanisms considered in this study are plastic yielding, power-law creep, and diffusion. M. F. Ashby and others have done extensive research to define the mechanics of high pressure and high temperature deformation to predict the final shape and density of powders during the HIP process. Li et al [7] has published densification rate equations which predict the densification rate of sintering powders based on a microscopic approach. This study attempts to incorporate these rate equations into a Multiphysics COMSOL model that can be used to predict the temperature, density, and final shape of a can filled with HLW calcine.

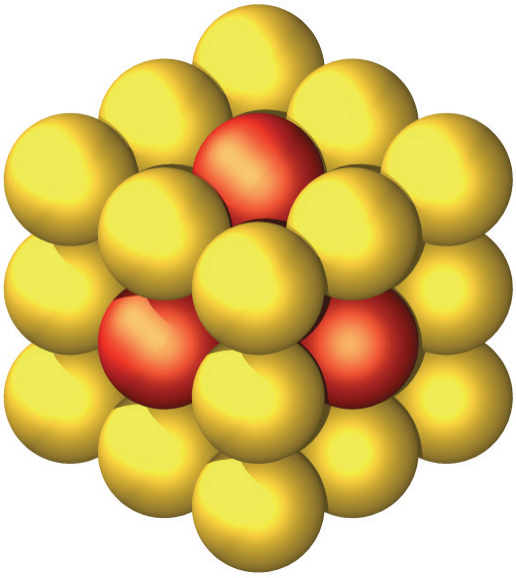

Figure 6. Randomly packed powder particles.

In a randomly packed material, each particle is in contact with six other particles [1], **Figure 6**. For densification to occur the particle centers must move closer together. When pressure is applied, the particles first begin to yield elastically and the centers begin to move together. As the pressure increases the force at the contact points also increases and plastic yielding begins. As the particles undergo plastic deformation the contact area of the particles increase and the forces are distributed over larger areas. Plastic yielding stops when the contact areas can support the forces from the applied pressure. When the pressure is no longer sufficient to cause yielding time dependant mechanisms for deformation begin and determine how the densification will progress. Both plastic yielding and time dependant deformation increase the number of contact points as well as the contact area and cause the particle centers to move closer together increasing the density of the material. **Figure 7** depicts particle deformation and densification during the HIP process.

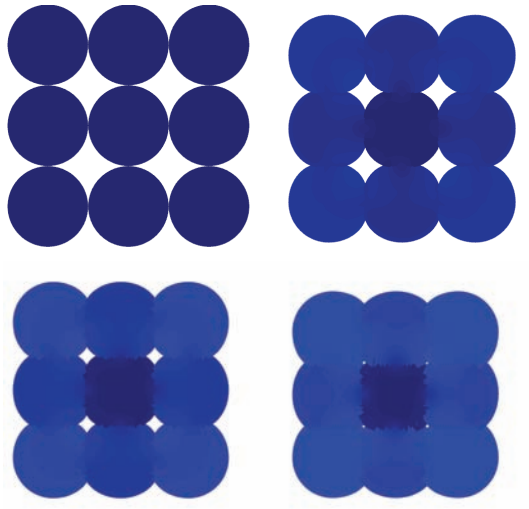

Figure 7. Particle deformation and densification during HIP process.

4.1 Use of COMSOL Multiphysics

To predict deformation and temperature in a material, a Multiphysics model is used. The deformation equations used to predict relative density, the ratio of the current density to the solid density of the material, are dependant on temperature and pressure. **Figure 8** shows the

physics used in the model and how they interact. A distributed ODE was used to incorporate the densification rate equations. The relative density calculated from the ODE is then used to calculate properties used in heat transfer and solid mechanics physics. The moving mesh is necessary to incorporate the large deformation that the material undergoes, increasing the accuracy of the temperature calculations and the solid mechanics model.

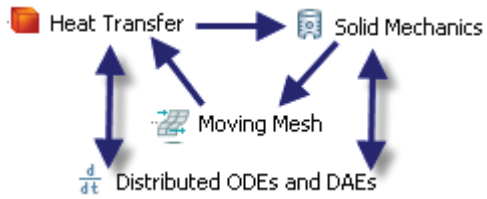

Figure 8. Multiphysics model schematic.

To model the shape change of the material an initial strain is calculated from the relative density determined by the ODE and is inserted into the structural model as an initial strain. At this time in our research we have not closely investigated the structural integrity of the can with the use of COMSOL due to the large deformation and large strain that the can undergoes. COMSOL is not currently capable of adequately predicting the stresses in the can. COMSOL is in the process of developing a module to handle large strain and large deformation. When that work is complete it will be incorporated into the model and the model refined. The primary area of focus has been to determine the temperature in the material during the HIP process and the final density and shape of the material.

4.2 Model Results

The results from the COMSOL model were compared against a lab experiment that was conducted in 1993 by the Idaho National Engineering Laboratory [4]. The can wall temperature and center point temperature of the material was measured during the course of test. The measured can wall temperature was used as a boundary condition in the heat transfer module to drive the temperature of the material. The experimental center point temperature was then compared to the COMSOL results. Two scenarios were evaluated. First, the thermal conductivity was held constant and not allowed to vary with temperature to bound the center point temperature. Next, the thermal conductivity was allowed to vary as a function of relative density which is a function of temperature and pressure. As seen in **Figure 9**, the center point temperature results from COMSOL, when using a constant thermal conductivity of the solid material, is higher than the experimental results, as is expected. When the thermal conductivity was varied the results from COMSOL closely resemble the experimental data.

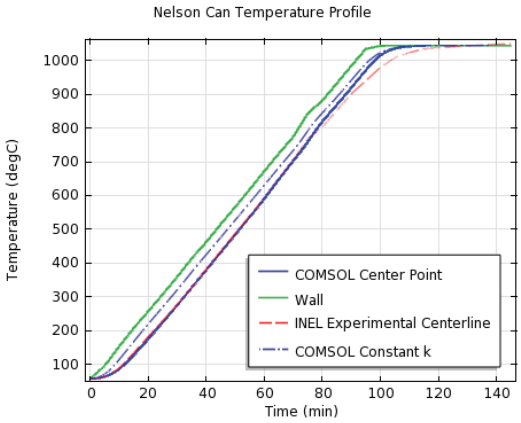

Figure 9. Temperature profile using deformation rate equations.

When determining the temperature in the can it is important to incorporate the deformation of the can in the heat transfer model by using a moving mesh. As the calcine begins to densify, the particle radiuses get closer together. This reduces the distance that the heat must transfer to reach the next particle. The model was run using a moving mesh and without. A small difference in the temperature profile was seen. The reduction in can size is expected to play a larger role in the temperature of the material as the initial can size increases. By incorporating the moving mesh with heat transfer the accuracy of the results are expected to increase.

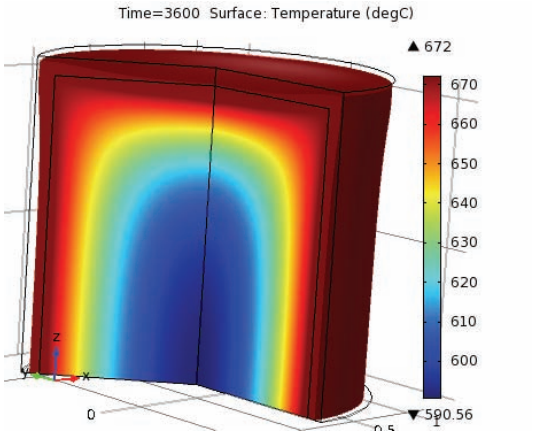

Figure 10. Intermediate temperature profile and shape change.

Figure 11 shows the intermediate relative density and shape of the COMSOL model. The model needs additional refinement to obtain more accurate results, but it does show that the material is denser near the edge of the can with less dense material near the center. Both the final shape and relative density are similar to those observed in the INEL experiment.

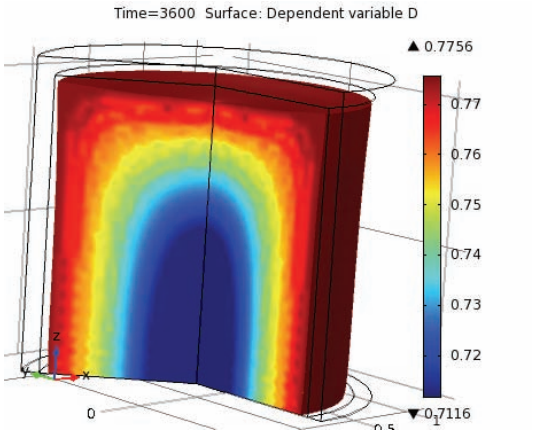

Figure 11. Intermediate relative density and shape.

5. Conclusions

Preliminary results of both models are promising. Implementing the volumetric change coefficient in place of the thermal expansion coefficient results in a model that is able to predict volume reduction as a function of pressure and temperature dependent density change. Improved accuracy may be obtained by refining the model to implement the volume change coefficient as an initial strain as opposed to thermal expansion. Future models with focus on refinement of material properties, implementation of the initial strain approach and inclusion of the material compressibility.

When applying the deformation mechanism rate equations COMSOL accurately predicts the temperature within the can and the final shape, see **Figure 12**. COMSOL also gives a good approximation of the relative density but additional refinement on the implementation of these equations is necessary to obtain reliable results. The rate equations are highly sensitive to material properties. More reliable results will be obtained as the accuracy of the material properties increase and are incorporated into the model.

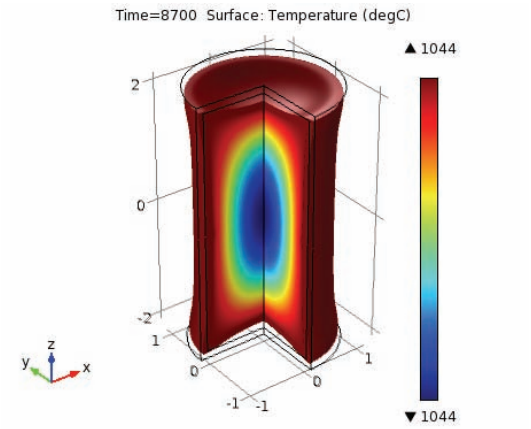

Figure 12. Final shape and temperature using rate equations.

6. Future Work

Future models will incorporate all aspects of the physical process. The models will be expanded to include plasticity in the can, large strain, large deformation, contact between the can and calcine, chemical reactions, and variable particle size. Currently Abaqus and COMSOL are being used to model the HIP process with the desire to combine all aspects of the process into a single Multiphysics model within COMSOL.

High fidelity analytical modeling is essential for the Calcine Disposition Project due to the prohibitive cost and complexity of testing

radioactive calcine. Analytic results will validate HIP can performance before the first radioactive can is treated. Thorough testing with non-radioactive surrogate materials will demonstrate scalability of the analytic models. With the use of these analytical models the design process will be improved and a safe and effective can will be designed.

7. References

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