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DEVELOPMENT OF THE PLUTONIUM MODELING AND ASSESSMENT (PUMA) SIMULATION THROUGH THE COUPLING OF EXPERIMENTS AND MOLECULAR DYNAMICS

A.J. Parkison, D. Andersson, U. Carvajal-Nunez, F. Carver, M. Cooper, J. Harter, T. Matthews, H. Milenski, A. Nelson, S. Parker, J. White, M. Wilkin, and N. Wozniak

Los Alamos National Laboratory, Los Alamos, NM, 87505, ajparkison@lanl.gov

A finite element computer simulation has been developed at the Los Alamos National Laboratory (LANL) to simulate the production process of $^{238}PuO_2$ heat source pellets. This simulation, the Plutonium Modeling and Assessment (PUMA) code, is built upon the MOOSE framework. It was developed through an extensive experimental campaign using surrogates and validated for use in the PuO_2 system through molecular dynamics simulations. The PUMA code is currently being used at LANL to understand and improve upon the current fabrication process.

I. INTRODUCTION

The production of $^{238}PuO_2$ pellets for use in Multi-Mission Radioisotope Thermoelectric Generator (MMRTG) systems currently experiences substantial process losses, predominately through pellet fracture prior to encapsulation. An example may be seen in Figure 1. It would be prohibitively expensive to conduct a comprehensive study on the fabrication process in order to understand the root cause of these failures. This work consists of a systematic approach which couples experiments using surrogate materials, development of a finite element computer simulation (PUMA), and validation using molecular dynamics simulations.

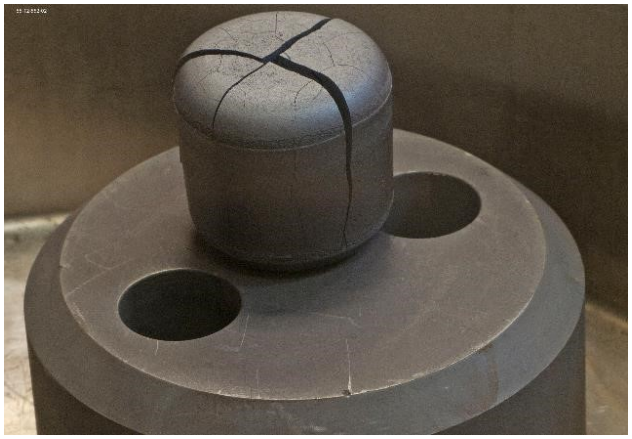

Fig. 1. Example of cracked plutonium oxide pellet during production

II. RESULTS

II.A Experimental

Cerium dioxide (CeO_2) was used as the surrogate material for all experimental studies due to its similar characteristics and phase structure when compared to PuO_2. While several material properties were measured such as thermal expansion as a function of stoichiometry and temperature, reduction rate, oxidation rate, heat capacity, surface interactions, and effect of impurities, no variable was found to have a larger influence on the presence of internal stresses within a $^{238}PuO_2$ pellet than thermal conductivity. This behavior was measured using Laser Flash Analysis (LFA) under flowing argon, and the results may be seen in Figure 2. Thermal gravimetric analysis (TGA) was used to verify there would be no stoichiometry changes under the conditions used to collect the LFA data.

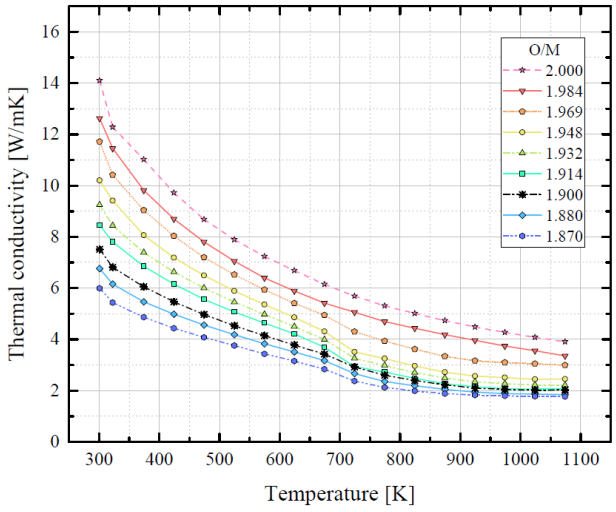

Fig. 2. Thermal conductivity of CeO_{2-x} as a function of stoichiometry and temperature.

The aforementioned variables were prioritized for experimental measurement by an iterative process through guidance from the PUMA simulation. This simulation was

then refined upon acquisition of this experimental information.

II.B Simulation

The PUMA simulation marks a distinct paradigm shift in how the $^{238}PuO_2$ program approaches understanding the pellet production process. It is fully capable of producing both spatial (Figure 3) and time-dependent (Figure 4) properties of $^{238}PuO_2$ pellets throughout the fabrication process.

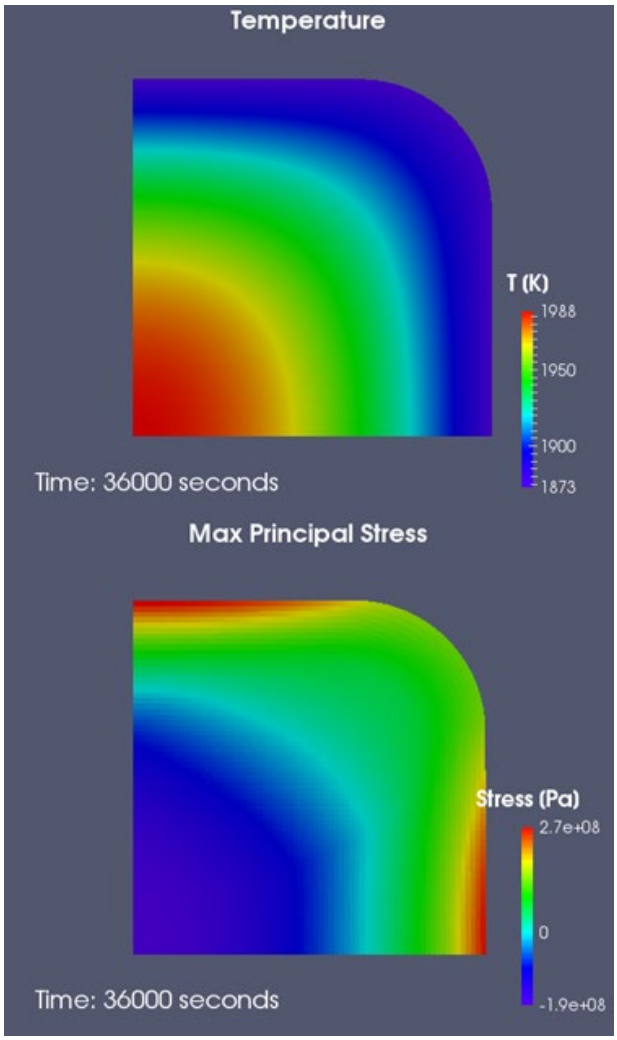

Fig. 3. Spatial dependence of temperature and principal stress for $^{238}PuO_2$ pellet at 1600 °C.

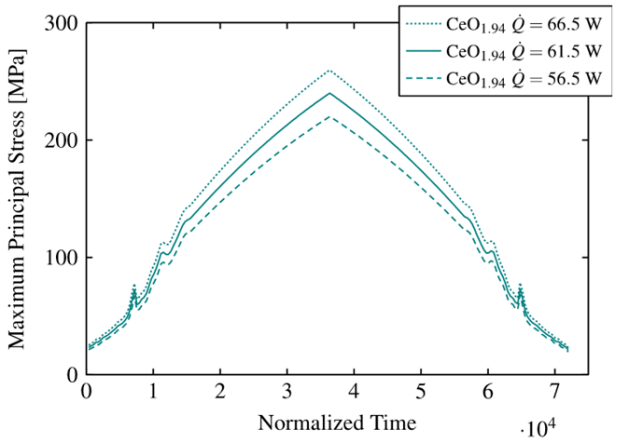

Fig. 4. Heating (RT-1600 °C) and cooling (1600 °C-RT) profile of self-heating $CeO_{1.94}$ as a function of power density.

The current state of the PUMA simulation allows for coupled, dynamic changes in material properties such as stoichiometry (Figure 5), thermal conductivity (Figure 7), and principal stress (Figure 8), among others. The heating profile shown in Figures 5, 7, and 8 was chosen arbitrarily simply for demonstration purposes and may be seen in Figure 6. This results in the most accurate depiction of the pellet fabrication process produced to date.

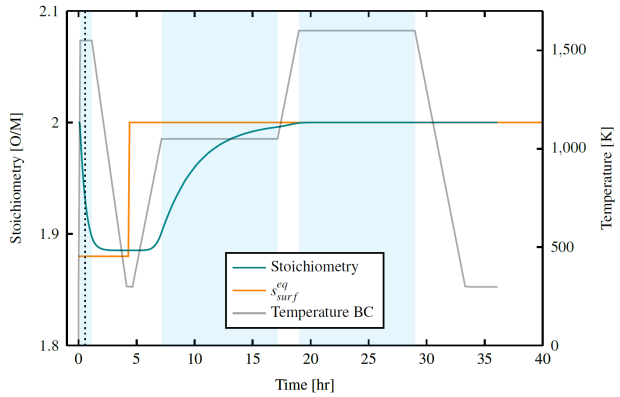

Fig. 5. Changes in pellet stoichiometry from reduction during the hot press operation and re-oxidation during sintering. Rates were determined through production experience and TGA analysis.

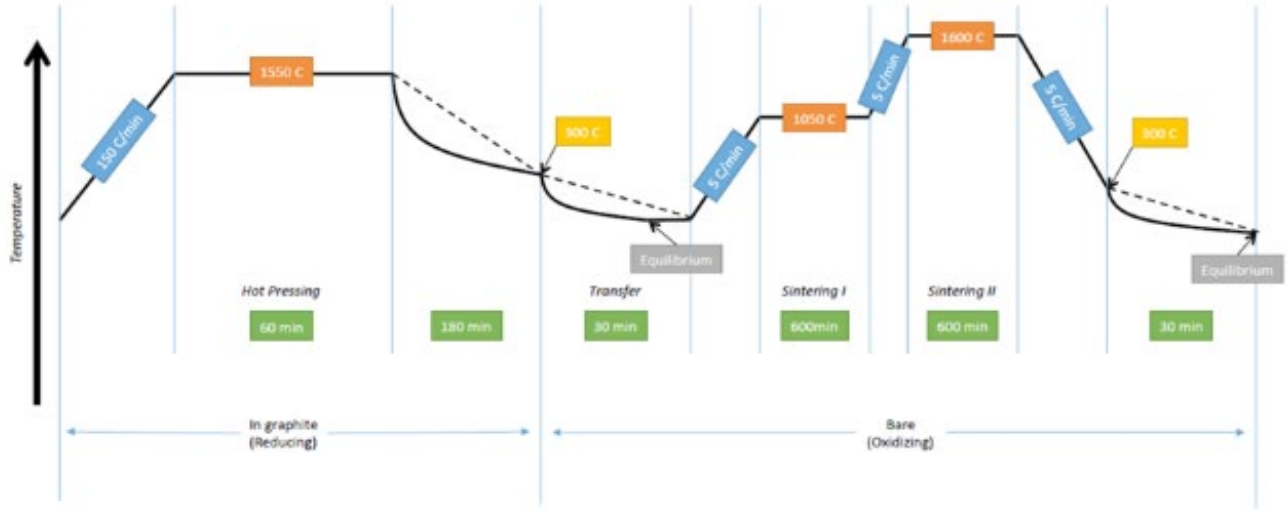

Fig. 6. Processing profile observed in Figures 5, 7, and 8.

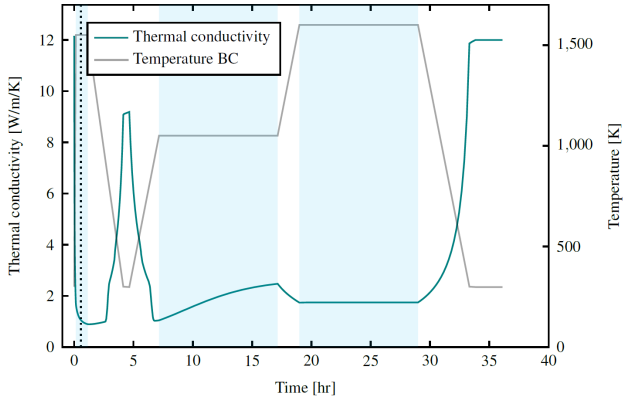

Fig. 7. Changes to thermal conductivity as a result of changes in temperature and stoichiometry.

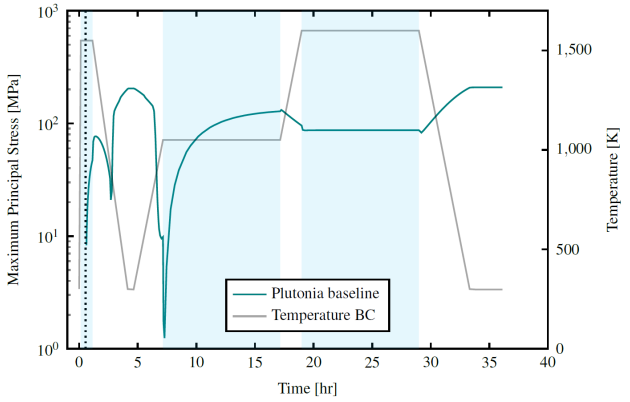

Fig. 8. Maximum principal stress observed by the pellet during the fabrication process.

II.C Molecular Dynamics

Molecular dynamics simulations were conducted using a combination of literature values, the experimental data acquired for this work, and interpolation. As stated previously, the thermal conductivity was the primary variable of concern, so emphasis was placed on ensuring its accuracy within the molecular dynamics simulation.

A sensitivity analysis of the variables used to simulate the thermal conductivity for a two phase CeO_x system was conducted. These variables may be seen in Equations 1-3.

$$\boldsymbol{\kappa = X_{Ce_{11}O_{20}}\kappa_{Ce_{11}O_{20}} + X_{F_1}\kappa_{F_1} + X_{F_2}\kappa_{F_2} + \kappa_{min}} \quad (1)$$

$$\boldsymbol{\kappa_{Ce_{11}O_{20}} = \frac{1}{A_{Ce_{11}O_{20}} + B_{Ce_{11}O_{20}}T}} \quad (2)$$

$$\boldsymbol{\kappa_F = \frac{1}{A_F + B_F T + c_F(2 - n_F)}} \quad (3)$$

It was found that the full data set for off-stoichiometric thermal conductivity could have been reliably acquired through interpolation using molecular dynamics by experimentally measuring the thermal conductivity of only CeO_2 and $CeO_{1.87}$ as a function of temperature. This may be seen in Figure 9. This greatly reduces the difficulty in acquiring the needed data set for the PuO_x system.

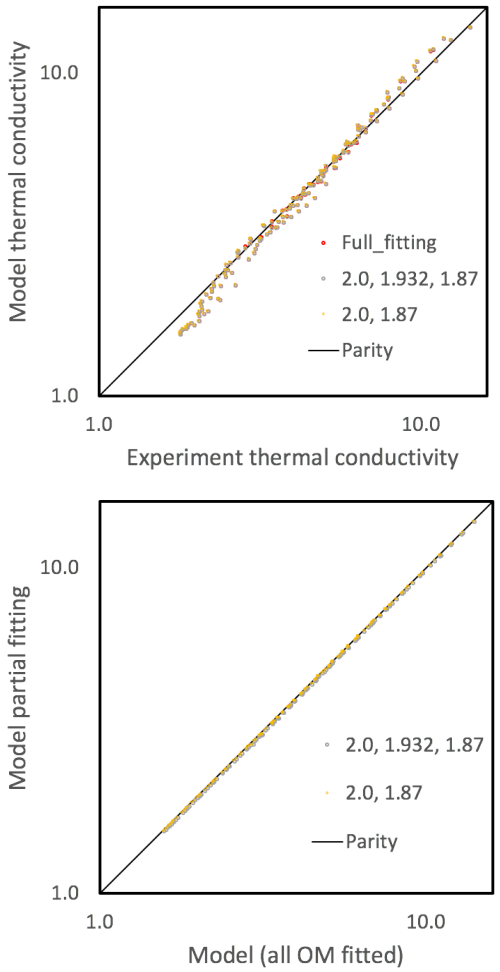

Fig. 9. Demonstration of ability for molecular dynamics simulations to predict material properties.

III. CONCLUSION

This work will continue to be applied to improving the fabrication process of $^{238}PuO_2$ pellets. Further refinements to the PUMA model and understanding of the applicability of molecular dynamics to this system is ongoing.

ACKNOWLEDGMENTS

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