

HOT ISOSTATIC PRESSING OF U-10ZR ALLOY NUCLEAR FUEL BY COUPLED
GRAIN BOUNDARY DIFFUSION AND POWER-LAW CREEP

A Thesis

Submitted to the Faculty

of

Purdue University

by

Sean M. McDeavitt

In Partial Fulfilment of the
Requirements for the Degree

of

Doctor of Philosophy

December 1992

'Could it be that questions tell us more than answers ever do?'
- Michael Card, 1983

ACKNOWLEDGEMENTS

I'd like to thank Dr. Alvin A. Solomon, my major professor, for his support, encouragement, and his consistent ability to generate new ideas when I felt I had reached a dead end. I am grateful to the School of Nuclear Engineering at Purdue University for the freedom to pursue my research interests in Materials Science. I also wish to thank Dr. G. Hofman, Dr. M. Dayananda, Dr. F. Clikeman, and Dr. T. Downar as members of my advisory committee and for their guidance at various stages in my research. A big thank you goes out to Dr. Mike Billone at Argonne National Laboratory for sponsoring me this past year through ANL's Lab-Grad program.

I want to thank my fellow students in the Nuclear Engineering Materials Group at Purdue University, especially Al Casagrande, who worked with me at the beginning of this project, and also Antai Xu, Mohammed Aslam, Kostas Davanas, and Pragna Bhakta for getting me started in the laboratory and showing me that it is possible to complete a thesis. I also thank Dennis Keiser, Mark Petri, and Pete Tortorici from Materials Engineering.

Most importantly, I want to thank God, for creating me with the ability to complete this work, my wife, Angela (to whom I dedicate this thesis), for loving and supporting me through this long road, and my children, Bethany and Stephen, who were born during this project (my most important results), and have been wonderful distractions from the frustrations of research.

This research supported by U.S. Department of Energy (DE-FG02-88ER12814). I want to thank Nuclear Metals, Inc. and Oak Ridge National Laboratory for the uranium and uranium hydride powders used in this study.

TABLE OF CONTENTS

	Page
LIST OF TABLES.....	vi
LIST OF FIGURES.....	vii
ABSTRACT.....	x
1. OVERVIEW.....	1
1.1 Metal nuclear fuels.....	2
1.2 The present work.....	6
2. MODELS AND BACKGROUND.....	9
2.1 Boundary diffusion controlled mechanism.....	9
2.2 Creep with interfacial control of grain boundary diffusion.....	13
2.2.1 Solute effects on diffusional creep.....	13
2.2.2 Particle by-pass controlled creep.....	17
2.3 Creep deformation enhanced and controlled mechanisms.....	20
2.3.1 Coupled boundary diffusion/dislocation creep mechanism....	20
2.3.2 Power-law creep control of HIP.....	25
2.4 Background information.....	28
2.4.1 HIP and swelling experimental background.....	28
2.4.2 Fuel performance of irradiated fuel	30
3. EXPERIMENTAL MATERIALS AND SYSTEMS.....	32
3.1 Experimental materials.....	32
3.2 Specimen fabrication.....	35
3.2.1 Inert atmosphere glovebox.....	35
3.2.2 Processing procedures.....	38
3.3 Sintering and hot-pressing methods.....	45
3.3.1 High pressure sintering system.....	45
3.3.2 Sintering and hot-pressing procedures.....	53
3.4 Sample preparation for microstructural evaluation.....	57

	Page
4. RESULTS.....	58
4.1 Fabrication and sintering results.....	58
4.1.1 List of experiments.....	65
4.1.2 Sintering results.....	68
4.1.3 Impurity phase identification.....	78
4.2 Microstructural characterization.....	81
4.2.1 Hydride-derived specimens.....	88
4.2.2 Metal-derived specimens.....	94
4.2.3 Grain size and pore spacing.....	102
4.3 Hot-isostatic pressing results.....	103
4.3.1 Hydride-derived specimens (general results).....	107
4.3.2 Metal-derived specimens (general results).....	111
4.3.3 HIP driving force dependence.....	124
4.3.4 HIP activation energy.....	129
5. DISCUSSION OF RESULTS.....	129
5.1 Threshold stress prediction.....	129
5.2 Grain boundary diffusion controlled HIP.....	132
5.2.1 HIP strain rate predictions.....	133
5.2.2 Mobility control vs. grain boundary diffusion control.....	135
5.3 Power-law creep controlled HIP.....	137
5.3.1 U-10Zr power-law creep correlation.....	139
5.3.2 HIP strain rate predictions.....	147
5.4 Coupled diffusion/power-law creep controlled HIP.....	148
5.4.1 HIP strain rate predictions.....	149
5.4.2 Comparison with experimental results.....	153
5.5 Time dependence of the coupled HIP model.....	158
5.6 Comparison with irradiated fuel behavior.....	158
5.6.1 Post-irradiation experiments at ANL.....	167
5.6.2 Fuel performance modelling.....	170
6. SUMMARY AND RECOMMENDATIONS.....	170
6.1 Summary.....	170
6.2 Recommendations.....	173
LIST OF REFERENCES.....	176
APPENDICES	
Appendix A: Specimen density measurements.....	182
Appendix B: Hydride-derived method development.....	186
VITA.....	189

ABSTRACT

McDeavitt, Sean M. Ph.D., Purdue University, December 1992. Hot Isostatic Pressing of U-10Zr Alloy Nuclear Fuel by Coupled Grain Boundary Diffusion and Power-Law Creep. Major Professor: Dr. A.A. Solomon.

Porous U-10Zr was produced using UH_3 and ZrH_2 powders and U and Zr metal powders. The powders were sintered under purified argon beyond pore closure to simulate fission gas bubbles. HIP experiments were performed at 700°C (γ phase) yielding an apparent driving force dependence of $n=3.4\pm 0.3$ and an apparent activation energy of $Q=187\pm 10$ kJ/mole. Experiments were attempted at 600°C (α' phase), but densification was not detectable. Post-HIP microstructures had modal pore sizes of 2.5 ± 0.1 and 1.7 ± 0.1 μm , pore spacings of 10 ± 1 and 7 ± 0.7 μm , and grain sizes of 34 ± 4 and 114 ± 12 μm for hydride- and metal-derived specimens, respectively. The hydride-derived specimens contained 25 vol.% of a ubiquitous Zr impurity phase stabilized by C, N, and O leaving ~ 5 wt.% Zr in solution, whereas the metal-derived specimens contained 5 vol.% of the Zr impurity phase leaving ~ 9 wt.% Zr in solution.

The coupled diffusion/creep cavitation model of Chen and Argon shows quantitative agreement with the measured HIP rates using $n=5$. The coupled model also predicts, for the first time, an asymmetry in HIP and swelling for identical driving forces due to differences in grain boundary stress. The difference in HIP behavior between metal- and hydride-derived material could be explained by differences in pore structure, whereas the differences in impurity particles and dissolved Zr content do not. A calculation of the

time-dependent HIP behavior was made which shows a varying time dependence that is consistent with the observed HIP rates.

The compressibility of U-10Zr is different from previously studied ceramics and metals and is significantly lower than U-5Fs, which appears to HIP by diffusive transport only. The differences could be due to higher grain boundary diffusivity and/or larger pore size-to-spacing ratios in U-5Fs causing diffusional growth to be rapid and dominant. The lack of detectable compressibility of U-10Zr in both the α' and $\alpha+\delta$ phases may lead to problems with fuel/cladding mechanical interactions during normal and off-normal conditions, especially at high burnup when solid fission products occupy available porosity.