Materials Research Proceedings **10** (2019) 11-17 doi: http://dx.doi.org/10.21741/9781644900031-2

Hot Isostatic Pressing of Radioactive Nuclear Waste: The Calcine at INL

Dr. Anders Eklund[1,a*], Dr. Regis A. Matzie[2,b]

[1]Quintus Technologies AB, Quintusvägen 2, SE-72166 Västerås, Sweden

[2]RAMatzie Nuclear Technology Consulting, LLC, 15 Winhart Drive, Granby, CT 06035, USA

[a]anders.eklund@quintusteam.com, [b]regismatzie@gmail.com

Keywords: Heat Treatment, Hot Isostatic Pressing (HIP), Radioactive Waste, Calcine, Vitrification, Immobilizing, Radioactive Isotopes, Leak-Before-Burst, Pre-Stressed Wire-Wound, Safety

Abstract. Hot Isostatic Pressing (HIPing) is a technology that has been around for 60+ years. By using high temperature and high gas pressure, dry metal and ceramic powders can be consolidated and a volume decrease can be achieved. This paper presents the simulations of using the HIPing process at the Idaho National Laboratory to treat the calcine radioactive waste. Once loaded into collapsible canisters, the HIPing would be used to treat the waste before final disposal. The resulting volume reduction was shown to be 20-70% and the cost ratio vs vitrification is 1:1.74.

Introduction

Hot Isostatic Press (HIP)

Hot Isostatic Press has fundamentally two different designs when it comes to contain the high pressurized gas, typically Argon. The two methods are called mono-lithic, sometimes referred to as mono-block, and pre-stressed wire-wound technology. An example of the wire-wound pressure vessel can be seen in Fig. 1.

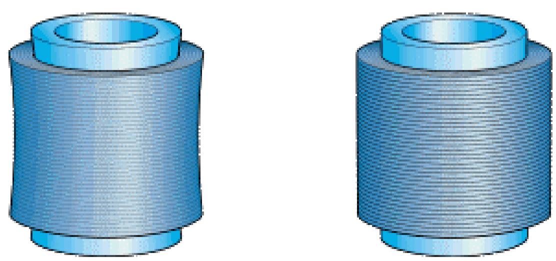

Fig. 1. Pre-stressed wire-wound vessel design. Without pressure applied (left) and with pressure applied (right).

The pre-stressed wire-wound HIP will always experience compressive stresses both on the inside and outside of the pressure vessel and the yoke frames during all phases of the HIPing process. This is the safest design and is approved by ASME per ASME Boiler and Pressure vessel code, Section VIII, Division 3. This failure mode is described as "Leak-before-burst". This means that if the pressure vessel cracks, the gas under high pressure will dissipate through the wire-wound package without any damages to the surrounding equipment and structures. For example, see Fig. 2.

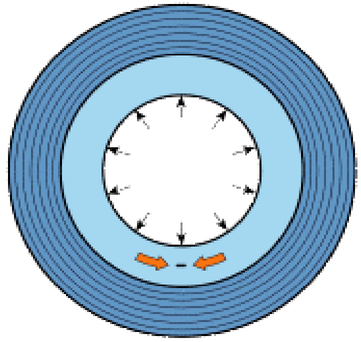

Fig. 2. Pre-stressed wire-wound vessel design showing the compressive stresses of a material fault even under high pressure.

The combination of elevated temperatures, 300-2500 °C, and high gas pressures, 50 – 300 MPa, consolidates dry metal and ceramic powders by mechanical deformation, creep and diffusion, and heal internal voids, i.e. metal castings, to substantially improve the strength of any materials. The temperature depends on the material to be HIPed, e.g., Aluminum has lower melt temperature (650 °C) than steel (1550 °C). An example of the effect of HIPing of voids in a material can be seen in Fig. 3.

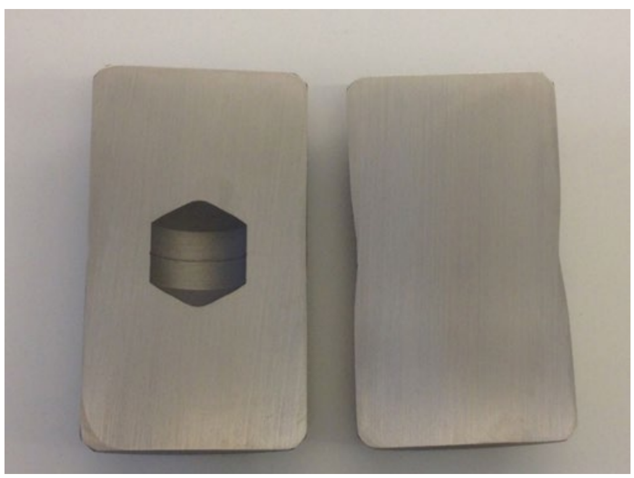

Fig. 3. Cross section of artificial pore before HIPing (left) and after HIPing (right).

The HIP cycle itself is strongly dependent, as mentioned before, on the parameters temperature and pressure. But, also time is of the essence in most applications since the material densification is depended on creep and diffusion which are time dependent mechanisms. Conventionally a HIP furnace was cooled naturally which took a lot of time of the total cycle time. A large HIP with a heavy load could take up to 12 hours to cool down before it was possible to open and start a new HIP cycle. Much efforts have been done the last decades to optimize and minimize the total cycle time. The introduction of a forced convection cooling technology called Uniform Rapid Cooling (URC®) greatly decreased the cycle time and allows the HIP operators to optimize the cycle to be most suitable for them and their materials. Cooling rates of 100 °C/min or more can easily be achieved. This increases the productivity of the HIP

Materials Research Proceedings **10** (2019) 11-17 doi: http://dx.doi.org/10.21741/9781644900031-2

unit significantly since the material throughput is increased for the same HIP size. An example of a HIP cycle without and with URC can be seen in Fig. 4.

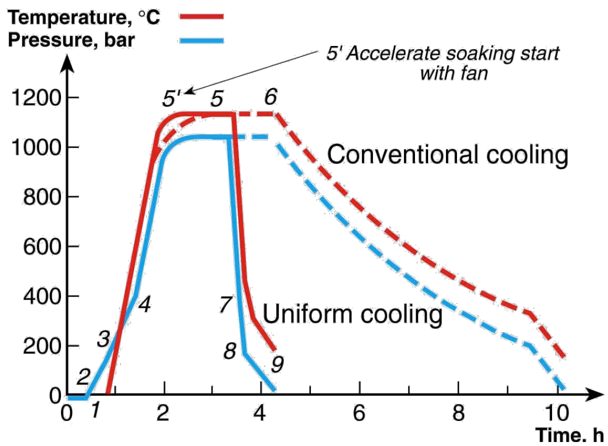

Fig. 4. Typical HIP cycle times without and with uniform rapid cooling.

The HIP system itself consists of the wire-wound yoke frames and a thin-walled cylinder, which can be considered as the back-bone of the pressure vessel since they take up the forces coming from the compressed gas. See Fig. 5.

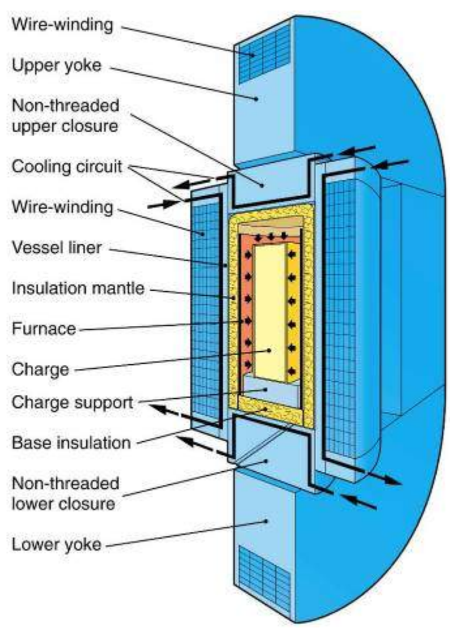

Fig. 5. Pre-stressed Wire-wound HIP machine.

The furnace, which is the heart of the HIP machine, has an elaborate design to ensure good insulation, temperature accuracy, rapid cooling and reliable and safe requirements. HIP furnaces can be supplied in Steel, Molybdenum or Graphite, depending on the operating temperature. See Fig. 6.

Materials Research Proceedings **10** (2019) 11-17 doi: http://dx.doi.org/10.21741/9781644900031-2

The charge, which can be canisters of powder or cast/forged parts, is placed on an insulated support structure. The gas flows freely around the charge for utmost temperature accuracy. The best temperature accuracy is achieved with a multi-zone convection furnace. See Fig. 6.

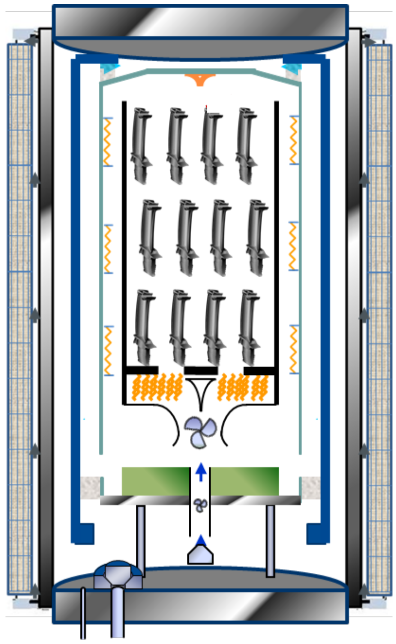

Fig. 6. Closer Look on the Furnace with multiple heating zones.

The Radioactive Waste

The Idaho Calcine is by definition a high-level waste. It is the first raffinate from the reprocessing of the spent nuclear fuel. The un-treated calcine is also classified as hazardous waste as it exhibits hazardous waste characteristics for toxicity of metals. Today, roughly 4400 m^3 of granular solid is stored in bins at the Idaho National Laboratory (INL) site. It has been shown by several studies that a calcine can be HIPed directly, if the particle size and material composition is right, or it can be mixed with an additive and then HIPed in a collapsible canister [1,2], see also Fig. 7. In both cases the result is a glassy ceramic waste form that can be packaged and disposed of in a final repository.

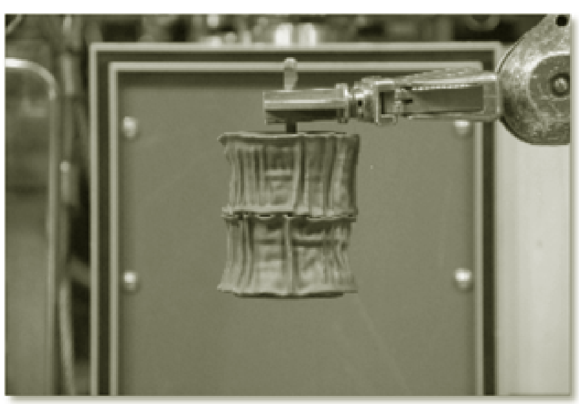

Fig. 7. HIPed treated zirconia calcine canister inside the MFC HFEF hot cell at INL.

Materials Research Proceedings **10** (2019) 11-17 doi: http://dx.doi.org/10.21741/9781644900031-2

Description

Hot Isostatic Pressing of Calcine

To make the test mixture, additives were combined with surrogate calcine to make a glass-ceramic final waste form in the HIP. The mixture was placed into a HIP can for bake out and sealed for HIP testing. Quintus Technologies, LLC assisted in development of the bake out and can sealing approach in their lab-unit in Columbus, OH as well as in the HIP-unit at INL supplied by Quintus Technologies, see Fig. 8.

Fig. 8. Quintus HIP unit installed in the HFEF hot-cell at the INL

After HIP testing, the canisters were sectioned so that samples could be collected from the HIP material. The temperature was varied to test the volumetric reduction for different pressures and temperatures. See Fig. 9 [3].

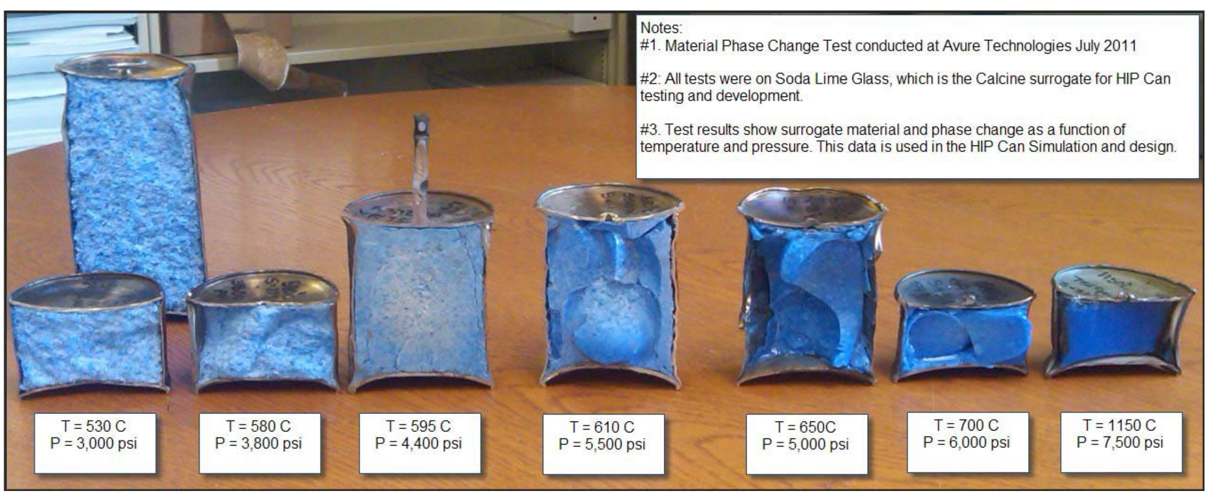

Fig. 9. Vertical cuts of all cans showing various volumetric reductions.

Materials Research Proceedings **10** (2019) 11-17 doi: http://dx.doi.org/10.21741/9781644900031-2

Discussions

Results of HIPing

From the trials mentioned above, some important conclusions could be made. First of all, that HIPing works to consolidate a ceramic like calcine. Also, that it is a versatile method for many different applications and can be used for many different possible treatment routes. See Table 1.

Secondly, that HIPing proves to be a very cost-effective method with the lowest life-cycle cost for the consolidation of the calcine, since the main cost is for storage volume and HIP shows the lowest storage volumes of all compared technologies. Today's generally established route of vitrification is shown to cost about 75% more than HIPing. See Table 2.

TABLE 1. HIP Technology Advantages over Vitrification [4].

Consolidation:	HIP	Vitrification (JHM)
Matrix:	Glass-ceramic	Borosilicate glass
Waste Loading:	60-90%	20-35%
Durability:	10-100 x EA-glass	10 x EA-glass
Final Volume: (relative to untreated calcine	20-70% reduction (treat=low, non-treat=high)	Min. 100% increase
Temperature:	1050-1200 °C	1150 °C
Pressure:	35-50 (100) MPa	Atmosphere
Off-gas/By-Product waste:	Minimal	Medium-High
Flexibility:		
- Calcine:	Treat or super-compact	Treat only
- Future mission:	Diverse/Flexible	Limited/Less flexible.

TABLE 2. Cost Comparison: HIP vs Vitrification [4].

Parameters	HIP without RCRA Treatment		HIP with RCRA Treatment		Direct Disposal		Vitrification with Separations		Vitrification without Separations	
Canisters	2 900	3 300	3 700	4 600	6 700	7 300	750	2 200	11 100	13 300
Total Life Cycle Cost (MUSD)	5 503	6 228	6 052	7 119	7 661	8 408	9 556	12 769	11 054	13 074
Cost ratio	1.00	1.13	1.10	1.29	1.39	1.53	1.74	2.32	2.01	2.38

Conclusions

Several advantages can be seen by using pre-stressed wire-wound HIP for consolidating the calcine at INL site. HIP is a proven technology since 60+ years with world-wide safe operations. Quintus HIP systems are built according to ASME Boiler and Pressure vessel code, Section VIII, Division 3, "Leak-before-burst". Technical advantages are that the waste is isolated from other process equipment, the process is scalable, it has the highest safety, there are no emissions from the consolidating process, it is flexibility to produce a range of waste forms, volume reduction of 20-70%, and it is a batch process for easy operation and diversity and reduces risk for

Materials Research Proceedings **10** (2019) 11-17 doi: http://dx.doi.org/10.21741/9781644900031-2

heterogeneous waste feeds. Economically the HIP process shows the lowest life-cycle cost compared with direct disposal and vitrification.

Acknowledgements
The authors wish to thank CH2M-WG Idaho, LLC for permission to publish the results of this work. This work was supported by U.S. Department of Energy, Idaho Cleanup Project, and Project No. 23582.

References

[1] M. Burström and R. Tegman, US patent 4642204, Feb.10, 1987.

[2] K. J. Bateman, R. H. Rigg and J. D. Wiest, Proc. ICONE10, Arlington, VA, April 14-18, 2002, 10TH International Conference on Nuclear Engineering, ASME 2002.

[3] D. C. Mecham, B. R. Helm and V. J. Balls, "Calcine Disposition Project Small Can Test Report, RPT-866, U.S. Department of Energy, Assistant Secretary for Environmental Management, Under DOE Idaho Operations Office, Contract DE-AC07-05ID14516

[4] B. D. Preussner et. al., Idaho Cleanup Project, U.S. Department of Energy, Idaho Operations Office, Project No. 23582.