

# High-Pressure Laser Diode Floating Zone and the Synthesis of NbTiSi<sub>x</sub>

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Muchiri Mbugua

Department of Mechanical Engineering, University of Maryland, Baltimore County (UMBC)

PI: Professor Tyrel McQueen, Director, PARADIM

Mentor: Ben Redemann, Graduate Student, Department of Chemistry, Johns Hopkins University

Contact: [jmbugua1@umbc.edu](mailto:jmbugua1@umbc.edu), [mcqueen@jhu.edu](mailto:mcqueen@jhu.edu), [bredema1@jhu.edu](mailto:bredema1@jhu.edu)

Website: [https://www.paradim.org/REU\\_participants/2021](https://www.paradim.org/REU_participants/2021)

We present the design of the High-Pressure Laser Diode Floating Zone Furnace (HPLDFZ) and the various custom designed parts to allow for the safe operation of the instrument. This was accomplished using 3D modeling software to properly secure the inner components of the instrument. We also present how the addition of silicon to niobium alloys improve their oxidation resistance at temperatures up to 1000°C. Materials were characterized using x-ray diffraction to properly determine the purity of the samples. The diffraction patterns show as more silicon is added to the pure NbTi phase, binary Nb<sub>5</sub>Si<sub>3</sub>, Nb<sub>3</sub>Si, and TiSi<sub>2</sub> phases start to become the majority phases in the diffraction pattern. The oxidation trend is supported by micron measurements on a ZIESS Stereo Microscope Stemi 508.

### Introduction:

The floating zone technique is a common crystal growth method. Typical floating zone growths are done without a crucible, which allows for no contamination from the containers and allows the material to solely react with the desired reactants, producing purer samples. This makes the technique a favorite for synthesis of highly reactive materials, intermetallic compounds, and refractory metals.<sup>[1]</sup>

The High Pressure Optical Floating Zone (HPFZ) in the PARADIM facility at Johns Hopkins University utilizes a halogen bulb to heat samples up to 3000°C and can achieve extremely high pressures of up to 300 bar. This allows for synthesis of materials that may not be possible under normal solid state synthesis techniques.

Located in the same facility, the Laser Diode Floating Zone (LDFZ) is composed of five class four lasers, which are the highest class of lasers due to the fact they are the most powerful and dangerous. The LDFZ also allows for the synthesis of single crystalline materials where the melting temperature of the materials is not achievable using standard furnaces. The five lasers housed inside the LDFZ are 200 W lasers that can be adjusted up to 35° to ensure the materials are properly melted.

The new HPLDFZ will contain seven 400 W lasers compared to the LDFZ's five 200 W lasers and greater than 3x the pressure capabilities of the HPFZ at 1000 bar. The HPLDFZ amplifies the traits of each instrument and combine them into a more powerful floating zone furnace. Several parts were designed to ensure the inner components are properly secured within the instrument.

Refractory materials are crucial to enable increased capabilities, improved efficiency, and decreased total life cycle costs.<sup>[3]</sup> High oxidation resistant materials are now highly sought after for their usage in high temperature environments. Oxidation in high-temperature environments can damage the materials and weaken their strength over time. Making materials that can be

highly resistant to oxidation as well as being mechanically tough is paramount for reducing costs as well as improving efficiencies over time.

### Method:

Autodesk Inventor was the CAD software used to design the parts for this project. The CAD models of the HPLDFZ enclosure and pulling drives were designed by Sci-Dre, the high-pressure chamber was designed by the IDG group at Johns Hopkins, and the high-pressure gas compressor was designed by Nova Swiss. Once the models were received, an adapter needed to be designed to allow the pulling drives to connect with the high-pressure chamber properly and securely. The HPLDFZ adapter was modeled from previous renders<sup>[2]</sup> as shown in figure 1. The model was then designed specifically for the HPLDFZ and the dimensions of the high-pressure chamber and pulling drives.

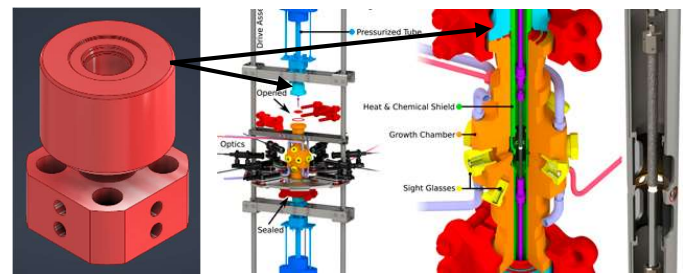
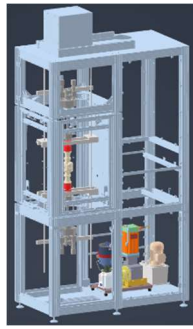


Figure 1: HPLDFZ Adapter and Reference Model

An exploded view of the entire assembly of the HPLDFZ is highlighted in figure 2, where you can see HPLDFZ bomb-proof enclosure (panels removed), pulling drives, high-pressure chamber, and high-pressure gas compressor.

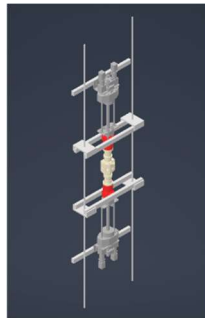
Different stoichiometric amounts of Nb pellets, Ti pellets, and Si crystals were massed out for alloys of  $\text{NbTiSi}_x$ , with  $x$  ranging from 0 to 1. Each sample was then prepared for arc melting, where the arc melter was purged with Ar gas for three cycles, each lasting five minutes. Zirconium was then melted in the center of the copper hearth to test for any residual oxygen. Each sample was melted until a bead formed and homogenized for three seconds. After cooling, the samples were then transported to the water jet cutter. Using the finest setting, the water jet cutter sliced each spherical sample in half into 1 cm samples to be used for XRD. Using a Bruker D8 Advance  $\text{Cu K}\alpha$  x-ray diffractometer in Bragg-Brentano geometry, samples were measured to determine purity. Some samples were also characterized using a scanning electron microscope (SEM) to determine the composition of the alloys. After characterization of the alloys was finished, the samples were then placed in an alumina crucible inside a box furnace to simulate the oxidation process. The box furnace was stepped to  $1000^\circ\text{C}$  for 24 hours then stepped down to room temperature. After the heating the samples were removed and examined under a ZIESS Stereo Microscope Stemi 508 with micron measurement capabilities.



**Figure 2:**  
Exploded View of the HPLDFZ

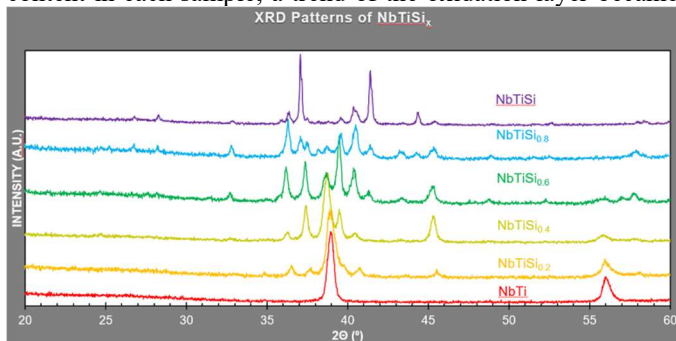
## Results:

After extensive design work, the HPLDFZ adapter successfully connected both the upper and lower pulling drives to the high-pressure chamber. This ensures the chamber will be properly secured inside the framework as well as providing an airtight seal for the enclosure. The frame was designed to secure the pulling drives and the high-pressure chamber of the HPLDFZ to the outer framework of the instrument as shown in figure 3.

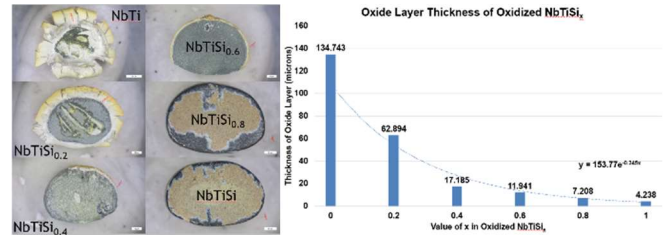


**Figure 3:** HPLDFZ Adapter, High-Pressure Chamber, and Pulling Drives Connected to Frame

After pure NbTi was synthesized, stoichiometric amounts of silicon were added. During the addition of silicon, new peaks started arising in the XRD patterns, these peaks were from the emergence of various  $\text{Nb}_5\text{Si}_3$ ,  $\text{Nb}_3\text{Si}$ , and  $\text{TiSi}_2$  phases shown in figure 4. Starting from NbTi and increasing the silicon content in each sample, a trend of the oxidation layer became



**Figure 4:** XRD Pattern of  $\text{NbTiSi}_x$



**Figure 5:** Oxide Layer Trend of Six  $\text{NbTiSi}_x$  Samples

apparent. As the silicon content was increased in the alloy samples, the oxidation layer decreased in size as shown in Figure 5. The width of the oxidation layer from NbTi was 134.743 microns compared to only 4.238 microns from NbTiSi, the sample with the highest silicon content.

## Conclusions:

We have shown the inner components successfully connect to each other via the HPLDFZ Adapter, but more designs will need to be designed to properly secure the frame for safe usage of the instrument.

We have also shown the addition of silicon to niobium titanium alloys drastically increases their oxidation resistance at temperatures up to  $1000^\circ\text{C}$  in addition to forming  $\text{Nb}_5\text{Si}_3$ ,  $\text{Nb}_3\text{Si}$ , and  $\text{TiSi}_2$  phases.

## Future Work:

The next step for the HPLDFZ is to add safety interlocks that further ensure the instrument operates in a safe manner. In addition, more parts will be designed and added to the supporting frame holding the pulling drives, high-pressure chamber, and HPLDFZ adapters.

As for the  $\text{NbTiSi}_x$ , the next step is to utilize SEM to verify the chemical composition of the alloy samples post box furnace oxidation. Nanoindentation will also be used to test the mechanical strength of the materials.

## Acknowledgments:

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