ISBN: 978-3-95450-229-5 ISSN: 2673-5520 doi:10.18429/JACoW-MEDSI2020-WEPC05

AN IMPROVED, COMPACT HIGH TEMPERATURE SAMPLE FURNACE FOR X-RAY POWDER DIFFRACTION

E. Haas, E. Cardenas, A.P. Sirna
National Synchrotron Light Source II, Brookhaven National Laboratory,
Upton, NY 11973-5000, USA

Abstract

A compact sample furnace was designed to heat samples to temperatures of 2000 - 2300°C at the X-ray Powder Diffraction (XPD) beamline at NSLS-II. This furnace allows the XPD photon beam to pass through with adequate downstream opening to collect diffraction data for high-temperature materials research. Since the XPD samples did not reach the desired temperatures, engineering studies, tests, and incremental improvements were undertaken to improve performance. Several approaches were considered, and the sample holder design was improved, and high-temperature coatings were used. The engineering work undertaken to improve furnace performance is included herein.

INTRODUCTION

The XPD Beamline at NSLS-II does materials research at temperatures ranging from cryogenic to ~1700°C, and efforts to conduct materials research at higher temperatures are ongoing. The present XPD sample furnace uses infrared lamps with ellipsoidal reflectors to focus infrared rays at a single sample. This furnace has been operating in air, and high-temperature oxidation has been destroying thermocouples. A plan therefore was developed to investigate potential areas of improvement:

a. The use of IR lenses to focus forward-directed IR rays otherwise lost as halo.
b. Improved sample tube holders.
c. The use of inert gas shielding to prevent oxidation.
d. The use of high-temperature coatings.
e. The use of a custom-designed secondary reflector to redirect forward-directed IR halo.
f. The use of a CO2 laser as a supplementary (or primary) sample heat source
g. Optimization of heat lamps and focal distance.

This paper briefly discusses each of the above options and explains why some of the above items were pursued further and others rejected. Considerations of material properties at high temperatures are also included.

SAMPLE AND FURNACE CONDITIONS

Powder samples contained in sapphire tubes are held by ceramic holders for X-Ray Powder Diffraction research in air at atmospheric pressure. Sapphire is used for its' high-temperature and optical properties. A computer graphic (CG) image and photo of a sapphire tube in its' ceramic tube holder is shown in Fig. 1 along with an image of one infrared heat flux pattern. This heat flux for a single ellipsoidal reflector indicates loss of some flux past the sample holder tube. Infrared lamps direct 150W each of heat flux at a ceramic sample tube holder in the bandwidth shown in Fig. 2. Six infrared lamps each have ellipsoidal reflectors and water-cooled jackets to minimize external temperatures for safety and to extend lamp life. Cooling water at 17-20°C from a chiller is circulated in parallel paths through all lamp cooling jackets and through two water-cooled outer shells.

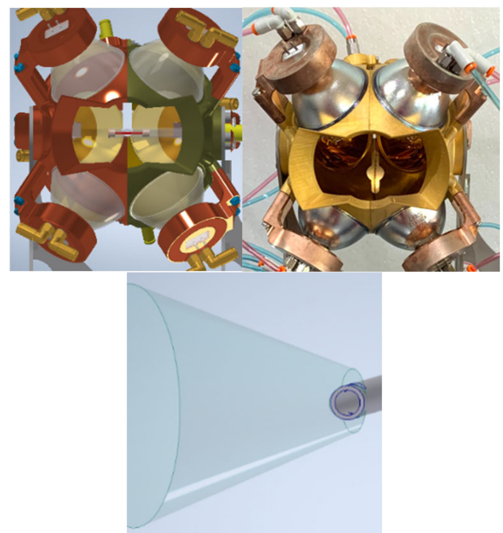

Figure 1: XPD Sample Furnace looking upstream: (a) CG image, (b) photo, and (c) heat flux pattern.

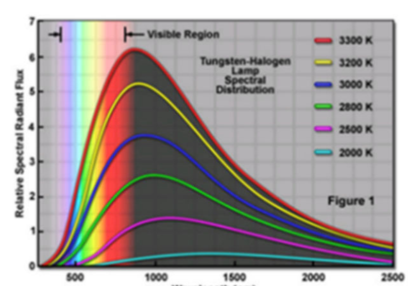

Figure 2: IR lamp flux spectrum.

ENGINEERING INVESTIGATIONS

a. The use of plano-convex infrared lenses as a primary means to focus forward-directed rays was considered, but not pursued as available infrared lens geometries and IR source distance limitations meant only a small portion of forward-directed rays were properly focused. When IR lenses were used to supplement ellipsoidal reflectors, too many reflected rays that would have been focused at the sample center were misdirected. IR lenses therefore were ruled out as a viable solution. Two IR lens configurations are shown in Fig. 3.

ISBN: 978-3-95450-229-5 ISSN: 2673-5520 doi:10.18429/JACoW-MEDSI2020-WEPC05

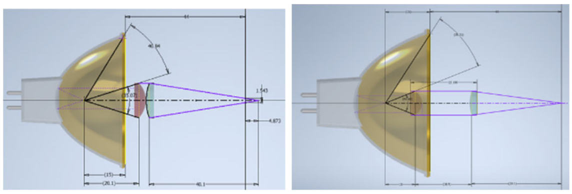

Figure 3: IR lens designs; focal point (a) behind sample, and (b) at sample center

b. Iterative design and thermal analyses were used to optimize the design of the sample tube holder. The external surface area was increased to capture more infrared rays while eliminating an internal cavity, thus convective cooling was reduced, and the conduction path to the sample itself shortened to transfer more heat to the sample by direct conduction to the sapphire tube. Figure 4 below shows the improved sample tube holder. Its features include a small hole in the front to allow the XPD beamline in, and a ± 45° downstream opening to allow X-ray diffraction data to be collected.

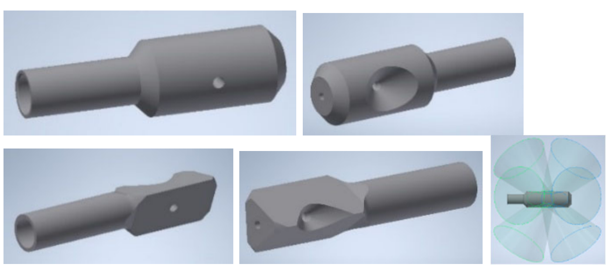

Figure 4: New sample holder designs and heat flux pattern impinging on a redesigned sample holder

c. Inert gas shielding - oxidation within the furnace increases non-linearly as the temperature inside the furnace increases. This may be mitigated by introducing an inert gas to shield materials from oxidation. Three inert gases and their specific heat capacities are shown in Table 1. The lowest specific heat will absorb the least amount of heat. This concept is demonstrated in the equation below. Based on gas specific heat capacity and cost effectiveness, Argon is the best inert gas for this application as it will carry away the least heat via convection.

$$Q = mc\Delta T \quad (1)$$

Table 1: Specific Heat Capacities for Helium Argon, and Nitrogen Gases [1]

Gas	Specific Heat Capacity (KJ/kg K)
Helium	5.1926
Argon	0.5203
Nitrogen	1.039
Air	1.005

d. Two high-temperature coatings were tested to improve heat transfer to high heat-flux areas of the sample holder and insulate areas outside the heat flux area. Important properties include coating adherence and high-temperature resistance in air. YSZ coating from Zypcoatings is a zirconia-based coating that can reach temperatures >2000°C without oxidizing per the manufacturer's specifications. This coating reduced the maximum internal temperature in initial tests and is being considered as an insulator outside the sample holder high-heat flux area. In other tests, a silicon carbide high-temperature coating significantly increased heat transfer to increase heat absorption are ongoing. Thermal properties of three sample holder materials are presented in Table 2.

Table 2: Thermal Properties of Tubing Materials [2, 3]

Properties	Alumina	Zirconia	Sapphire
Thermal Conductivity (W/m K)	2.2-4.3	14-30	34.6
Thermal Expansion (µm/m K)	8.8-9.5	6.7-8.2	4.5
Max. Temp (˚C)	1330	1730	2053
Specific Heat Capacity	400-510	870-940	750

e. The use of a custom designed secondary reflector was investigated to recover some of the forward directed IR rays otherwise lost as halo. The shape and overall design of the reflector is shown in the Fig. 5. This shape and location were determined by radiation heat transfer view-factor calculations. The reflector needs the lowest view factor possible to reduce the number of rays blocked. Fig. 5 (a) represents the View Factor Geometry. Spherical reflectors were not pursued further due to decreased lamp life and loss of flux reflected by the ellipsoidal reflector.

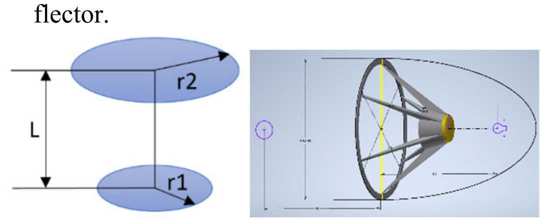

Figure 5: (a) Reflector View Factor Geometry and (b) Proposed Spherical Reflector Design

f. Small cross-sectional beam (i.e., ≤ 1.5 mm diameter) CO2 lasers operating at a wavelength between 9.5 and 10.5µm can apply IR heat energy directly to the sample at a wavelength where the heat energy will be absorbed more efficiently by the sapphire sample tube. Due to a limited budget, this area was not pursued yet, but future laser power versus sample temperature tests are being considered.

g. IR heat lamps tests indicated as much as 28°C difference when moving a single heat lamp 1 mm away from its' maximum temperature point. Further tests using shims to optimize the focus of heat lamps are ongoing.

RESULTS

The following two graphs (Figs. 6 and 7) were produced by focusing an ILT (model L6409G) lamp at ¼" diameter Alumina and Zirconia ceramic tubes with, and without silicon carbide and type YSZ high-temperature coatings. In all tests, lamp voltage, tube position, and thermocouple position were held constant, and the same size tube was used. The heating and cooling curves in Fig. 7 show the results of the tube materials and the effects of the coatings. The highest temperature was reached by a silicon carbide coated zirconium tube, and the YSZ coating showed some value as an insulator. The effects of various gases where also explored and tabulated in Table 3. It was found that the use of an inert gas can improve the efficiency of radiation transfer from lamp to inside ceramic tubing with Argon gas proving the best performance.

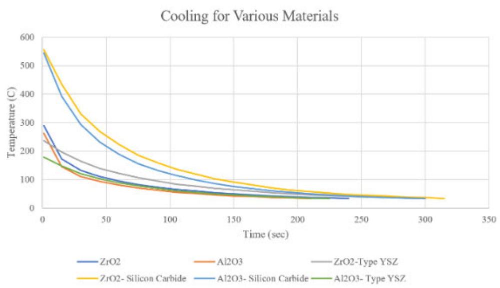

Figure 6: Cooling rate for selected ceramic materials and coatings.

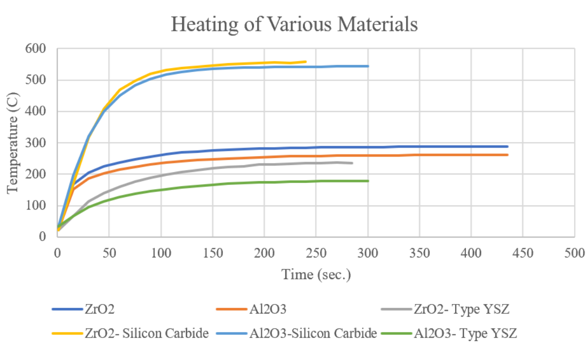

Figure 7: Heating rate for selected ceramic materials and coatings.

Table 3: Single Lamp Max Temps for Noble Gases

Gas	Max Temp Achieved (˚C)
Helium	185
Argon	258
Air	242
Nitrogen	243

CONCLUSIONS

Materials research at temperatures above 1750°C in air requires careful sample holder material selection and an understanding of material properties at high temperatures. Refractory metals undergo oxidation, and high-temperature ceramics are very difficult to machine. Additionally, thermal absorptivity and emissivity are wavelength dependent when using infrared heat sources. Some materials (e.g., sapphire) are mostly transparent to near infrared rays, and many ceramics are poor heat conductors. Cost is also a consideration. The solutions chosen to improve the performance of the XPD Infrared Sample Furnace include the selective use of infrared lamp position adjustments and an improved sample holder. The new sample holder maximizes heat absorption, provides a short heat conduction path to samples, and optimizes external size to reduce convective heat transfer losses. The use of high-temperature coatings is promising, but coating adherence and maximum coating temperature need further improvement. The authors were not able to improve performance with infrared lenses, and secondary reflectors will diminish lamp life with insufficient thermal gain. Lastly, CO2 lasers (~10µm wavelength) hold significant promise as a primary or secondary heat source, but tests were not undertaken in time for publication of this paper.

ACKNOWLEDGEMENTS

The authors thank Jianming Bai, PhD (XPD scientist and project leader) and Sanjit Ghose, PhD (XPD Lead Beamline Scientist) for project guidance, Hui Zhong (SUNY SB liaison engineer) for thermal data, G. Lawrence ('Larry') Carr, PhD (FIS/MET Lead Beamline Scientist) for infrared guidance, Leo Reffi for design drawings, and John Trunk (HXSS program chief technician) for thermal experiment support. Also, thanks go to Eric Dooryhee, PhD (HXSS program manager), Lisa Miller, PhD (NSLS-II educational program coordinator), and Noel Blackburn (BNL SULI program coordinator); without their support (and DOE-SULI program funding), this work would not have been possible.

REFERENCES

[1] Properties of Various Ideal Gases (at 300 K), www.ohio.edu/mechanical/thermo/property_tables/gas/idealGas.html

[2] Oxide-Based Engineering Ceramic, MakeItFrom.com, www.makeitfrom.com/compare/Alumina-Aluminum-Oxide-Al2O3/Zirconia-Zirconium-Dioxide-ZrO2

[3] Supplier Data - Sapphire Single Crystal (Alumina 99.9%) - (Goodfellow), www.azom.com/properties.aspx?ArticleID=1721