Infrared Imaging of Temperature Distribution in a High Temperature X-ray Diffraction Furnace

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ABSTRACT

High Temperature X-ray Diffraction (HTXRD) is a very powerful tool for studies of reaction kinetics, phase transformations, and lattice thermal expansion of advanced materials. Accurate temperature measurement is a critical part of the technique. Traditionally, thermocouples, thermistors, and optical pyrometers have been used for temperature control and measurement, and temperature could only be measured at a single point. Infrared imaging was utilized in this study to characterize the thermal gradients resulting from various sample and furnace configurations in a commercial strip heater furnace. Furnace configurations include a metallic strip heater, with and without a secondary surround heater, or a surround heater alone. Sample configurations include low and high thermal conductivity powders and solids. The IR imaging results have been used to calibrate sample temperatures in the HTXRD furnace.

Keywords: Infrared imaging, x-ray diffraction, HTXRD, strip heater

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1. INTRODUCTION

High temperature x-ray diffraction (HTXRD) has been used in materials science to identify crystallographic changes and phase transformations as a function of temperature¹⁻³. Obtaining x-ray diffraction patterns in this dynamic fashion has allowed researchers to learn more about how and under what conditions a crystal or a certain phase in a complex material is formed. One of the key factors in HTXRD is accurate measurement of specimen temperature and temperature distribution in the heater/sample holder.

Many laboratory HTXRD furnace designs are based on resistance heating of a metallic strip. A specimen either in powder or bulk form is placed on top of the heater strip during the measurement. Ideally, a large uniform temperature zone is expected in the heater strip and the specimen temperature should be the same as the thermocouple(s) attached to the heater. However, thermal gradients along the heater strip are inherent in the design, since both ends of the strip heater are usually water cooled in some manner. In addition to the thermal gradients along the length of the heater strip, thermal gradients between the sample and the strip may exist depending upon the interfacial thermal contact resistance. One method of studying these thermal gradients quantitatively is to attach numerous thermocouples to the heater strip and/or sample. Several difficulties arise when using this method: (1) attaching thermocouples to the heater strip and the sample is difficult and tedious; (2) heat loss through thermocouple contacts can cause errors in the measurements; and (3) only a limited number of thermocouples can be put on a heater strip and the measurement can easily miss thermal non-uniformity in a larger area. Thermistors and single point optical pyrometers have also been used in temperature calibrations of HTXRD furnaces. However, none of the above techniques has imaging capability. Thus, the true temperature distribution of the heater strip cannot be obtained.

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Recent developments in infrared imaging technology permit a real time digital thermal map to be made with high spatial and temporal resolutions^{4,6}. For the first time, IR imaging was used to take temperature maps of the heater strip in a HTXRD furnace. The uniformity of the heating system was studied by varying the set temperatures. The effectiveness of a surround heater was also studied. Temperature maps with specimens in bulk and powder forms were obtained. The results provided important information about temperature distribution of the strip heater as well as calibration for temperature dependency of the x-ray diffraction patterns. This information can be used directly in designing the HTXRD furnace and improving measurement accuracy.

2. EXPERIMENTAL

The high temperature system consisted of a furnace [Bühler HDK-2] with two programmable DC power supplies [Lambda] and a dual input dual output programmable controller [Micristar - Research Instruments]. The furnace is shown in Figure 1. The sample holder/heater is a Pt - 30%Rh alloy strip, clamped to water-cooled electrodes at both ends. A Type S thermocouple was spot welded at the bottom center of the strip. It was used for both the temperature measurement and heating control. Another thermocouple (Type K) was attached to the water-cooled end. Readings of these two thermocouples were used in temperature calibration for the IR camera. A surround heater assembly consisting of a Pt-Rh strip wrapped around the main heater strip, open at the top to pass the x-rays, was used to test its efficacy in providing a more uniform heating of the strip. Both the strip and the surround heater were controlled via a desktop PC using commercial diffractometer software [DMSNT version 1.34 - Scintag, Inc.]. When the heating system is inside the chamber, only a tiny spot about a few square millimeters could be observed through a small window in the furnace. Therefore, all the experiments were carried out in air without radiation heat shields or water-cooled vacuum chamber so that the entire heater could be imaged. Because of the absence of heat shields we confined the study to temperatures up to 300°C. At a set point of 300°C, heating current going through the strip heater was 31 A and heating current of the surround heater was 30 A.

Figure 1. Heating system in a HTXRD furnace (50 mm between cooled electrodes).

A Raytheon Amber infrared camera, model Radiance HS, was used in this study. A full image has 256 x 256 pixels. With a 25 mm lens, a spatial resolution between 300 – 400 μm per pixel was achieved in this study. Snap shots were taken after the heater had reached thermal equilibrium. At temperatures above 100°C, the IR intensities were reduced by using faster integration times and a neutral density filter (ND1 or ND2), attached between the camera and the lens. An Areodag graphite spray was used to make all the surfaces black with the same emissivity. All temperature maps presented in this paper were taken at 300°C. Line profiles between the two cooled electrodes were obtained from the Image Desk II software. Temperatures at the center and the end of the heater were recorded before each snap shot was taken. The IR intensity data were transferred into an Excel spreadsheet where temperature and distance conversions were applied to the data.

3. RESULTS AND DISCUSSION

3.1 Using strip or surround heater alone

Figure 2(a) shows the temperature map using the strip heater alone at a set point of 300°C. After temperature calibration, using the readings from the center and end thermocouples, the temperature profile, shown in Figure 2(b), reveals a temperature gradient of nearly 100°C/cm. The distribution indicates an area of 4 - 5 mm wide was near the set point temperature. Temperature dropped quickly further away from the center. The uniform temperature region becomes even smaller when the set point temperature is increased. This is due to the electrodes being water cooled causing the temperature gradient to become steeper when the set temperature at the center of the heater strip gets higher. The temperature map in Figure 2(a) also shows some non-uniformity across the heater, i.e. perpendicular to the heating current. One edge of the heater (at the top of the figure) was hotter than the opposite edge by about 20°C. A temperature gradient in this direction may pose more of a problem than temperature gradient along the length of the heater strip. This temperature gradient is not yet fully understood, but could be a result of uneven contact of the strip with the electrodes. The edge effect and other boundary conditions of the heater could also influence the temperature distribution in the heater strip.

A surround heater is frequently used in this HTXRD furnace to improve the temperature uniformity of the heater strip. Figure 3(a) shows the temperature map using only the surround heater, with the set point at 300°C. Note the surround heater had to be set to a much higher temperature in order to heat the strip heater to the set point via radiation. A temperature profile along the strip heater, shown in Figure 3(b), has a different shape than the temperature profile shown in Figure 2(b). The profile is noisy and the temperature gradient along the length of the strip appears to be steeper. However, the 20°C temperature gradient across the edges of the strip was not observed which suggests that using the surround heater alone can provide more uniform heating across the heater strip. The surround heater was also controlled by the thermocouple welded at the center of the strip heater. Because both electrodes of the strip heater were water-cooled, temperature gradients similar to the case of using the strip heater alone were observed.

3.2 Using both strip and surround heaters

Figure 4(a) is a temperature map using both the strip and surround heaters. This was the suggested operating mode by the manufacturer of the HTXRD furnace. The temperature distribution looked similar to the one shown in Figure 2(a) with the strip heater alone. In fact, the temperature profiles shown in Figure 4(b) and Figure 2(b) give exactly the same temperature gradient. The same temperature gradient across the heater strip was also observed. These results indicate the surround heater as operated is not reducing the thermal gradient in the present arrangement in which nearly equal DC voltages are applied to the strip and surround heater. A much larger surround heater voltage is clearly needed. For this particular HTXRD system, using the surround heater as the main heater can help to eliminate temperature gradient across the strip. However, this operating mode is not practical at high temperatures, i.e. above 1000°C, since the surround heater must be at a much higher temperature than the set point and the melting point of the surround heater material must be considered.

3.3 Temperature distribution of heated powder specimen

LaB₆ powder was used to study the temperature uniformity of the heater strip with a powder specimen. A LaB₆ – ethanol paste or slurry was applied as a strip covering half the heater length. The LaB₆ powder only covered half the width of the heater strip leaving the surface of the heater strip exposed to compare with the specimen temperature. Only LaB₆ powder was left on the heater when the solvent was evaporated. Figure 5(a) is the temperature distribution of the powder and the heater with the strip heater at 300°C. The heater and the specimen showed similar temperature gradients. The heater appears to have a higher temperature than the specimen. This observation was confirmed in Figure 5(b) where the two line profiles, measured from the heater and the powder, are shown together. The heater strip was found to be about 20°C hotter than the specimen. Such temperature differences were significant enough to cause concerns in HTXRD measurements. Thus, it is necessary to calibrate sample temperature in the measurements, ideally as a function of temperature, because the temperature

measured by the thermocouple spot welded below the heater strip can show much higher temperature than the real sample surface temperature. These effects are likely larger for low thermal conductivity materials, thicker specimens, larger particle sizes, and low packing densities.

3.4 Temperature distribution of a solid specimen

The effect of heat transfer between the heater strip and the sample became more obvious when a bulk Al_2O_3 specimen was clipped onto the strip heater. The surface of the specimen was sprayed with graphite in order to compare the temperature with the heater strip which was also sprayed with graphite. Although Al_2O_3 is a fairly good thermal conductor among oxides, poor thermal contact between the heater and the specimen caused a temperature drop as shown in Figure 6(a). A temperature difference of nearly 40°C was measured according to the temperature profiles in Figure 6(b). At the edges of the specimen the sample temperatures were slightly higher than the heater because of the cavity effect at the edges which resulted in temperature readings being higher than the real values. This result showed that bulk specimens in HTXRD measurements can have much lower temperature than the thermocouple spot welded to the bottom of the heater strip. Necessary steps need to be taken to improve the thermal contact between the sample and the heater and ideally locate a thermocouple on the surface of the specimen. A temperature calibration using a sample with the same dimensions is thus strongly recommended.

4. CONCLUSIONS

Infrared imaging of the HTXRD furnace showed significant temperature variations of the heater strip. The line profile along the length of the strip indicated only a 4-5 mm region at the center of the heater was at the set point, and temperature dropped quickly away from the center. Significant temperature gradients across the width of the heater strip were also observed. The use of the surround heater does not change the temperature profile of the heating strip unless the power is controlled independently and a much higher power is applied to the surround heater assembly. Similar temperature profiles were found when LaB_6 powder was used as the test specimen. A temperature difference of 20°C was measured between the powder and the heater strip. A more significant temperature difference of 40°C was measured when a bulk Al_2O_3 specimen was clipped onto the heater strip. Poor thermal contact between the specimen and the heater was believed to be the cause of the temperature difference. This study suggests that temperature calibrations must be taken when performing HTXRD experiments, especially when bulk samples are used, in order to minimize the errors in estimating the sample temperature. One of the objectives of this study is to provide temperature distribution information to the designers of HTXRD furnaces. The results suggest a more complex heating system is needed to achieve a larger uniform temperature zone for x-ray diffraction measurements

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REFERENCES

1. R. Srinivasan, T.R. Watkins, C.R. Hubbard, B.H. Davis, "Sulfated zirconia catalysts: the crystal phases and their transformations", *Chem. Mater.*, **7** (1995) 725-730
2. E.A. Payzant, W.D. Porter, C.R. Hubbard, "High temperature phase transformation in rhombohedral bismuth strontium oxide", *Thermochimica Acta*, **318** (1998) 45-50
3. R.M. Stroud, K.F. Kelton, S.T. Misture, "High temperature X-ray and calorimetric studies of phase transformations in quasicrystalline Ti-Zr-Ni alloys", *J. Mater. Res.*, **12** (1997) 434-438
4. H. Wang, M. Bartkowiak, F.A. Modine, R.B. Dinwiddie, L.A. Boatner and G.D. Mahan, "Nonuniform heating in zinc oxide varistors studied by infrared imaging and computer simulation," *J. Am. Ceram. Soc.* **81**(8) pp. 2013-2022, 1998
5. H. Wang and R.B. Dinwiddie, "Thermal diffusivity mapping of 4D carbon-carbon composites," *Ceramic Engineering and Science Proceedings*. Vol. 18. No. 4, pp. 755-762, 1997
6. H. Wang and R. B. Dinwiddie, Advanced thermal imaging of composites," *Advances in Ceramic-Matrix Composites*, III, *Ceramic Transaction* Vol. 74 pp. 609-618, 1996

Figure 2(a) Temperature map using the strip heater alone.
Dimension: 0.12 x 10 x 80 mm; 50 mm between cooled ends

Figure 2(b) Temperature profile along the long axis of the heater strip showing a 4-5 mm uniform zone at the set temperature.

Figure 3(a) Temperature map of the heater strip using the surround heater only.

Figure 3(b) Temperature profile of the heater strip with surround heater only.

Figure 4(a) Temperature map of heater strip with both strip and surround heating.

Figure 4(b) Temperature profile of the heater strip with both strip and surround heating.

Figure 5(a) Temperature map of LaB_6 powder on the heater strip at 300°C .

Figure 5(b) Temperature profiles of LaB_6 powder and strip heater at 300°C .

Figure 6(a) Temperature map of a bulk Al_2O_3 specimen on the heater strip.

Figure 6(b) Temperature profiles of a bulk Al_2O_3 specimen and the heater strip.