HIGH ENERGY X-RAY DIFFRACTION TECHNIQUE FOR MONITORING SOLIDIFICATION OF SINGLE CRYSTAL CASTINGS

Dale W. Fitting, William P. Dube', and Thomas A. Siewert

Materials Reliability Division
Materials Science and Engineering Laboratory
National Institute of Standards and Technology
Boulder, CO 80303

INTRODUCTION

X-ray diffraction has been used successfully to study metal solidification and temperature-dependent phase changes[1-3]. However, this research used very thin specimens (a few mm at most), furnaces with low attenuation x-ray windows (beryllium, graphite, or polyimide), and low x-ray energies (< 50 keV). Since the penetration depth of low-energy x-rays is shallow, traditional x-ray diffraction is thus unable to probe the interior of thicker structures. Others have used higher energies to study thicker samples. Work, including that by Green[4] and by Kopinek, et al[5], extended x-ray diffraction investigations to energies exceeding 150 keV.

In this paper, we show that by using higher x-ray energies (up to 320 keV) and a transmission configuration, solidification of a metal casting within a mold can be studied. The liquid-solid boundary in the casting can be precisely located from the spatial distribution of x-rays transmitted through the casting and diffracted from the liquid or solid. Diffraction spots arise when the probing x-ray beam encounters an crystalline solid. X-ray diffraction from the liquid metal is characterized by a diffuse ring. The high-energy x-rays used have sufficient energy to penetrate the furnace walls, mold, and specimen, while retaining a moderate cross-section for coherent scattering (which can produce diffraction).

PHYSICS OF TRANSMISSION DIFFRACTION

Probing the interior of a casting requires a transmission configuration. X-rays energies of over 100 keV are needed to penetrate the refractory oxide mold (5-10 mm wall thickness) and casting specimen (1 -20 mm thick). Below, we explore the physics of using high energy x-rays to produce transmission x-ray diffraction.

Nondestructive Characterization of Material VIII
Edited by Robert E. Green Jr., Plenum Press, New York, 1998

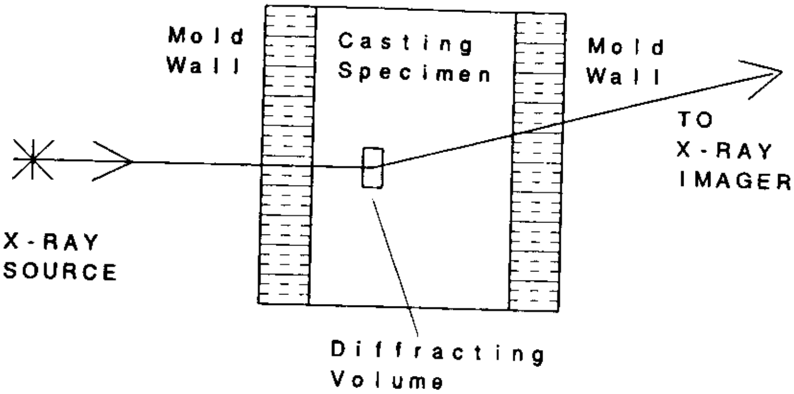

Figure 1. Geometry of transmission diffraction from a crystalline specimen surrounded by a casting mold.

Consider the situation shown in Figure 1, of an x-ray beam incident on a crystalline specimen contained within a casting mold. The primary x-ray beam is attenuated as it passes through the mold wall and a portion of the specimen to a location where coherent (elastic) scattering occurs. The scattered x-ray is attenuated along the exit path through the remaining specimen and exit mold wall. Losses along entrance and exit paths are minimized by raising the energy substantially above that used in conventional x-ray diffraction systems (5-20 keV) to 150-300 keV.

The amplitude of elastic scattering from a crystalline solid is given by the product of the structure factor (F) of the crystal and the Thomson scattering amplitude from an isolated electron[6]. F is a function of the types of atoms in the crystal (through the atomic scattering factor, f), the configuration of the unit cell of the crystal , and the particular lattice planes which are involved in the scattering. For a face-centered cubic (fcc) crystal, such as nickel, the structure factor is equal to $4f$ if the Miller indices (hkl) are unmixed (all odd or all even) and is equal to zero if the indices are mixed. The intensity of the scattering is the square of the amplitude. The structure factor is thus squared, to give $16f^2$ for an fcc material with lattice planes defined by unmixed Miller indices.

The dependence of the atomic scattering factor for nickel is plotted in Figure 2, as a function of the scattering angle for several x-ray energies. The empirical method proposed by Waasmaier and Kirfel[7] was used to calculate f. In the forward direction the scattering from all electrons in the atom is in phase and so f is equal to the number of electrons which scatter the

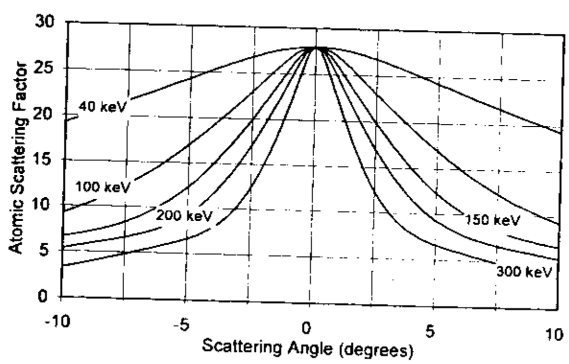

Figure 2. Atomic scattering factor calculated for nickel, plotted as a function of scattering angle for several x-ray energies.

x-rays, that is, the atomic number of the atom. At low x-ray energies, the atomic scattering factor is quite large, even for large scattering angles. However, with high x-ray energies, f is significant only for small scattering angles, that is for a transmission geometry. For example, the scattering factor for nickel is 16.5 for 150 keV x-rays scattered at 3.5-degrees from the direction of the incident x-ray beam (typical of our experimental geometry). The structure factor squared for this example is 16×16.5^2, or 4356.

The large structure factor, the enormous number of atoms along the path of the primary beam (all with the same crystalline structure), in addition to the substantial intensity of high-energy x-rays which penetrate through a mold and specimen, account for the high efficiency of transmission diffraction. Figure 3 plots the transmission energy spectra recorded for a mold encased nickel alloy single crystal casting. The peak in the spectrum of the diffraction spot at 185 keV, confirms that the high energies required to penetrate the mold and casting can indeed produce intense transmission x-ray diffraction.

SENSING SOLIDIFICATION DURING CASTING

A directional solidification furnace, used for prototype single crystal turbine blade castings, was fitted with an x-ray source and real time x-ray imager as shown in Figure 4. The x-ray tube was a 320 kV tungsten target tubehead normally used for radiographic imaging. The emission from its 1.2 mm focal spot was collimated with lead apertures to produce a 1 mm diameter beam. The primary x-ray beam entered the vacuum furnace through a 10 mm thick borosilicate glass port. After transmission through the 9.5 mm thick alumina hot zone coil support, the beam entered the mold and specimen. Diffraction from the specimen emerged from the casting, and passed through the alumina coil support and furnace exit port. A real time x-ray imager (fiber optic glass scintillator coupled through an image intensifier to a CCD camera) was used to observe the diffraction pattern. Two-axis linear motion stages on the x-ray tube and imager were used to scan the probing x-ray beam over the casting. The distance from the x-ray focal spot to the casting specimen was 835 mm. The imager was 505 mm from the specimen.

In a series of casting experiments, the x-ray beam was first directed into the lower section of the casting, where the water-cooled ram of the DS furnace had caused the alloy to solidify. Generally, only one or two diffraction spots were observed in the XRD image; although others

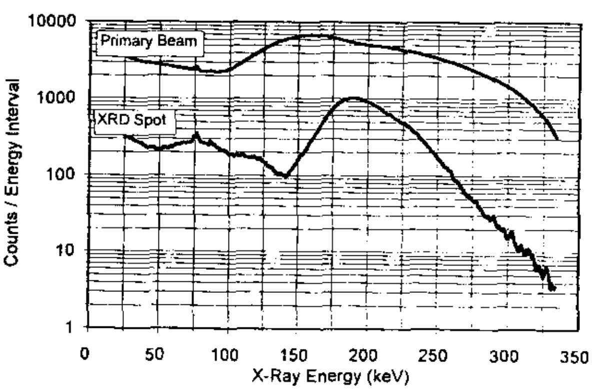

Figure 3. Energy spectrum recorded during a transmission diffraction experiment on a 6 mm thick nickel alloy (N5) single crystal casting in a mold (5 mm wall thickness). X-ray parameters were 320 kV, 0.75 mA. A 25 mm thick lead collimator, with a 2 mm clear aperture was placed on the 37 mm diameter x 13 mm thick intrinsic Ge detector to separately sample the two spectra by centering the aperture on the primary beam or the diffraction spot.

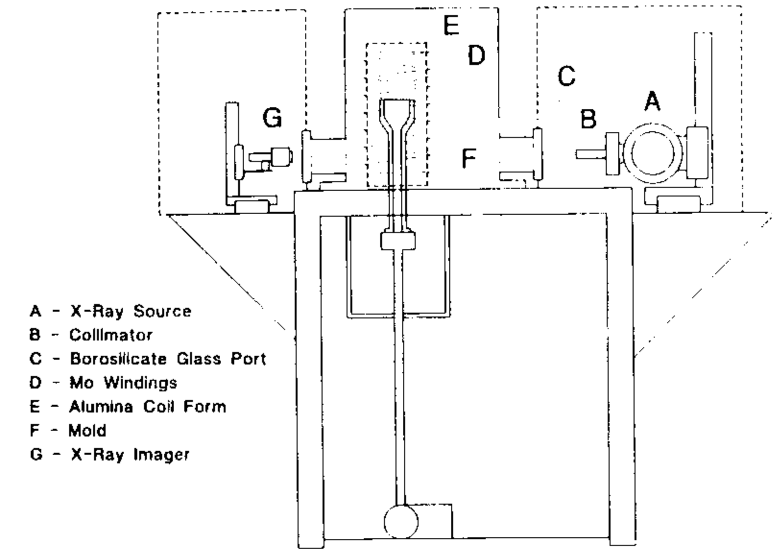

Figure 4. Resistively heated directional solidification furnace, fitted with a collimated x-ray source and real time x-ray imager for transmission x-ray diffraction studies during casting of single crystal nickel superalloys.

could be brought into the small field-of-view (50 mm diameter) of the x-ray imager by rotating the casting. We optimized (by rotating the specimen) the intensity of a particular diffraction spot and used the motion stages on the x-ray source and x-ray imager to scan the sensed area vertically.

Figure 5 plots the mean XRD spot gray level (spot brightness) of an intense transmission diffraction spot as the x-ray source and imager were scanned vertically with respect to the casting. A high spot intensity was observed when the x-ray beam probed a solid region of the casting. The diffraction spot intensity diminished as the x-ray beam entered the region of dendritic solidification (x-ray path traversing a region which is part liquid and part solid). When the x-ray beam was directed through a region of the casting containing only molten alloy, the diffraction spot vanished and the intensity in the region of interest dropped to a low level. Repeated vertical scans, both up and down, showed the same behavior of diffraction spot

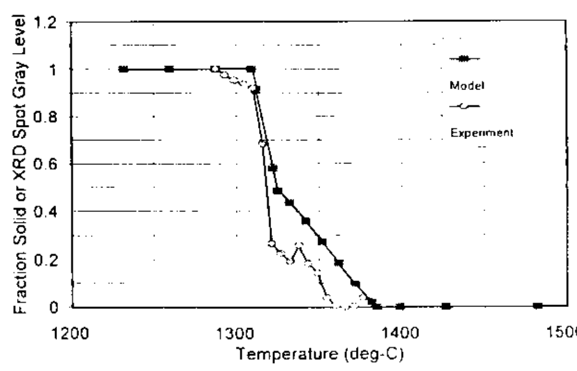

Figure 5. Transmission diffraction spot gray level plotted along with model (Lever) predictions of the fraction solid versus temperature for N5 alloy. XRD spot gray level (intensity) is plotted versus position as the XRD sensor is scannedvertically from a solid region of N5 upward into areas containing less solid and more liquid.

intensity increase (more solid) and decrease (more liquid). Other diffraction spots were rotated into view and additional vertical scans performed. The diffraction spot intensity varied in a similar manner.

A model (Lever) prediction of the fraction solid versus temperature for N5 alloy is also plottted in Figure 5 along with the measured transmission diffraction spot gray level. Since the temperature profile in the casting was not measurable in this experiment , we have adjusted the vertical and horizontal scale of the XRD data to coincide with the solidification model predictions. It is encouraging that the shape of the modeled and experimental curves are quite similar. The experimental data also appears to show formation of the predicted second phase (break in the curve).

SUMMARY

A method has been developed for sensing the physical state (liquid or solid) of a single crystal casting during withdrawal in a directional solidification furnace. A collimated high energy (320 kV) x-ray beam is directed, through the mold, into the metal alloy casting. X-rays diffracted from the crystalline solid produce an ordered array of high intensity transmission diffraction spots. Diffraction from the molten alloy produces a transmission image characterized by a diffuse ring of scattering. The vastly different diffraction patterns provide a means for locating areas of liquid and of solid during the casting process. We have also found that the intensity of a single diffraction spot varies with the fraction of the specimen volume probed by the x-ray beam which is solid. Plots of diffraction spot intensity versus vertical position in the casting are in good agreement with the modeled fraction solid versus temperature curve.

REFERENCES

1. S.E. Doyle, A.R. Gerson, K.J. Roberts, and J.N. Sherwood, Probing the structure of solids in a liquid environment: a recent in-situ crystallization experiment using high energy wavelength scanning, *J. Crys. Growth*, 112:302-307 (1991)

2. K.G. Abdulvakhidov and M.F. Kupriyanov, High-temperature unit for x-ray diffraction studies of single crystals, *Instr. & Exptl. Tech.*, 35(5): 942-943 (1992)

3. H.L. Bhat, S.M. Clark, A. Elkorashy, and K.J. Roberts, A furnace for in-situ synchrotron Laue diffraction and its application to studies of solid-state phase transformations, *J. Appl. Cryst.*, 23:545-549 (1990)

4. R.E. Green, Jr., Applications of flash x-ray diffraction systems to materials testing, *Proc Flash Rad. Symp., ASNT Fall Conf.*, 151-164 (1977)