Enhanced Concentric Solidification Technique for High-Temperature Laser-Scanning Confocal Microscopy

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We present further developments and enhancements of the concentric solidification technique for high-temperature laser-scanning confocal microscopy. These recent enhancements include an automated in-situ determination of phase fractions during solidification using purposely developed image processing software, more accurate temperature calibration, reproducibility tests as well as close coupling of the experimental results with computational simulations. In the present study, the newly developed capabilities of the concentric solidification technique have been applied to a study of the peritectic phase transition in the Fe–C system.

KEY WORDS: solidification; microscopy; phase transformation; experiment; steel.

1. Introduction

In 2004 an advanced experimental technique for high-temperature laser-scanning confocal microscopy (HTLSCM) has been developed by Reid et al.,1) which has been further developed and utilized for the experiments conducted in the present study. The details of HTLSCM have been described in detail in the literature1) and there is no need to repeat these details. In the concentric solidification technique, a centralized pool of liquid metal is contained by a rim of solid of the same material under a radial thermal gradient. A specimen of 9.8 mm diameter and about 250 μm thickness is placed in an alumina crucible, which in turn is held in a platinum holder, as shown in Fig. 1. A B-type thermocouple is guided through a 2-bore alumina holding rod and spot-welded on the outer edge of the holder. The specimen is positioned at the upper focal point of the furnace with the heat source being at the lower focal point of the ellipsoidal cavity within the gold-plated infer-red furnace.

The ability to establish a centralized melt pool depends upon the presence of a radial thermal gradient across the specimen, which is formed when the diameter of the radiant beam is small compared to the diameter of the specimen. A large diameter of the radiant beam results in a more uniform temperature distribution and smaller radial temperature gradient, whereas a small diameter of the radiant beam generates a hot-spot localized in the centre of the specimen, leading to a steeper radial temperature gradient. In order to measure the diameter of the radiant beam in the microscope, a disk of thermographic paper with a diameter of 9.8 mm was placed in the crucible instead of a metallic specimen and the furnace was switched on for a few seconds. The paper consists of a plain paper coated with a material that changes colour during heating. After measuring the colour intensity along the paper diameter, a plateau in the centre of the paper with a diameter of approximately 2 mm has been found indicating a focal point radius of 1 mm, as shown in Fig. 2.

The thickness of the sample plays a pivotal role in successfully creating a stable and sustainable liquid pool. Reid et al.1) established that the maximum sample thickness needs to be < 250 μm for Fe–C alloys in order to generate a thermal distribution within the specimen conducive to the formation of a stable liquid pool. A further advantage of using a thin sample is that the thermal gradient in the through-thickness direction approaches zero, leading to the formation of a vertical solid/liquid interface. The significance of the absence of a thermal gradient in the through-thickness direction is that observations made on the free surface are representative of events occurring in the bulk since the direction of growth will be primarily in the plane of the specimen. Cross- as well as planar sectioning of solidified specimens verified these assumptions and confirmed that the observations made on the surface of specimens are indeed representative of bulk behaviour. In thick specimens

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Note

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the thermal gradient established in the through-thickness of the specimen can potentially lead to a non-planar liquid/solid interface and the disturbance of the progressing interface creates uncertainty about the precise direction of growth.\textsuperscript{1)} Phelan\textsuperscript{2)} provided a detailed analysis of the general interpretation of observations made in HTLSCM and need not be repeated.

In the conventional HTLSCM technique, the presence of a meniscus makes it difficult to resolve the interface between solid and liquid phases, particularly in the early stages of solidification where growth in a fully molten sample, generally proceeds from the crucible wall up the side of the meniscus. The difficulties encountered with image quality in the presence of a meniscus are exacerbated in the course of a study of the peritectic reaction since a thin layer of \(\gamma\)-austenite grows along the liquid/\(\delta\)-ferrite interface. Hence, the presence of a meniscus in rectangular or cylindrical crucibles hampers clear observation of the position of the phase interfaces, as illustrated in Figs. 3(a)–3(b). In the concentric configuration (Fig. 3(c)), the liquid phase is in contact with a solid rim of the same material, the alumina crucible and the gas atmosphere. The resultant surface tension energy balance, in the alloys studied, between solid and liquid phases, particularly in the early stages of solidification where growth in a fully molten sample generally proceeds from the crucible wall up the side of the meniscus, resulting in a larger area that is in sharp focus across the phase interfaces without the need of a constant refrocusing.\textsuperscript{3)}

2. Determination of Phase Fractions

Another benefit of the vertical solid/liquid interface in conjunction with the concentric geometry of the phases is that an accurate measurement of the phase fractions of liquid and solid can be carried out \textit{in-situ} throughout the experiment. For a known sample radius \(R_S\) and liquid pool radius \(R_L\), the fraction of liquid and solid can be calculated as:

\[
f_L = \frac{(R_p^2 \cdot \pi) / (R_S^2 \cdot \pi)}{1} \quad \text{........................ (1)}
\]

\[
f_S = 1 - f_L \quad \text{........................ (2)}
\]

These phase fractions are strongly dependent on the occurrence of micro-segregation during solidification and it is vitally important to quantify the extent of segregation in order to gain understanding of and to control microstructural development. The traditional approach to determine such segregation consists of a variety of mathematical models and quenching experiments, but does not include \textit{in-situ} measurements during actual solidification due to experimental difficulties. Therefore, an attempt was made to quantify the extent of micro-segregation \textit{in-situ} during solidification experiments utilizing HTLSCM in combination with the concentric solidification technique. By accurately measuring the phase fractions and interface temperatures during solidification, a simple mass balance can be used for binary alloys in order to calculate the average composition of each phase and hence, the amount of micro-segregation can be quantified.\textsuperscript{3,4)} A detailed description of the temperature calibration is has been given by Griesser et al.\textsuperscript{3,4)} For the determination of concentration gradients across the specimen the commercially available software package DICTRA\textsuperscript{5–7)} has been successfully utilized to model the concentric solidification experiment.\textsuperscript{4,8)}

When the temperature of the liquid/\(\delta\)-ferrite interface drops below the peritectic temperature, a layer of \(\gamma\)-austenite forms and grows along the interface, separating the liquid from the \(\delta\)-ferrite (\textit{i.e.} the peritectic reaction). During the subsequent peritectic transformations, at cooling rates approaching equilibrium cooling, one interface of the \(\gamma\)-austenite platelet grows into the remaining liquid and the other interface grows into \(\delta\)-ferrite, maintaining the concentric arrangement of the phases as schematically illustrated in Fig. 4.

By measuring the liquid pool radius and the thickness of the \(\gamma\)-austenite phase, the fraction of \(\gamma\)-austenite can be calculated. Figure 4 shows the fraction of \(\gamma\)-austenite after isothermal holding at several temperatures below the equilibrium peritectic temperature. The phase fractions were measured after the transformation interfaces came to a standstill at a fixed position in the specimen (\textit{i.e.} equilibrium conditions). According to the principles of thermodynamics, no \(\gamma\)-austenite will be thermodynamically stable at the equilibrium peritectic temperature (1 768.76 K\textsuperscript{9)}), which is in excellent agreement with the measurement. However, note...
that these measurements are determined in the presence of a radial temperature gradient and are not to be compared with isothermal conditions.

3. Automated Video Analysis

Automated video processing software has been developed to track the progression of the phase interfaces as a function of time and temperature as well as to automatically measure the radius of the liquid melt pool for the determination of the phase fractions. A detailed description of the method has been provided by Griesser et al., but it is instructive to provide a brief summary pertinent to the present discussion. By detecting the position of the solidification interface as a peak in the gradient intensity profile along a user-defined tracking path, the software is able to automatically determine the radius of the centralized liquid melt pool. The results of this analysis are the interface position and radius over time and temperature, which are further used to calculate the phase fractions of solid and liquid respectively. A module to automatically measure the rate of the peritectic reaction is also incorporated in the software. By the use of this software the analysis of the recorded experimental observations can be conducted with increased accuracy, reproducibility and efficiency compared to a manual evaluation.

4. Reproducibility

In order to ensure the reproducibility of the experiments, different specimens of a Fe-0.18C alloy have been solidified with applied cooling rates of 2 and 10 K/min, respectively, starting from the same initial liquid pool radius in each experiment. The progression of the liquid/δ-ferrite interface over time for both cooling rates is shown in Fig. 5. All curves are plotted until the peritectic phase transition occurred. The repetition of the experiments shows high accuracy and similarity of the measured solidification progress, indicating reproducible thermal and diffusional conditions. The small deviations at the end of the experiments are believed to result from different numbers of δ-ferrite grain boundaries in a particular specimen, which have a small effect on the diffusivity of carbon and therefore slightly affect the progression of the liquid/δ-ferrite interface. However, this difference is insignificant. Also, the morphology and kinetics of the observed peritectic phase transition, as well as the measured undercooling below the equilibrium peritectic temperature, were reproducible.

5. Summary and Conclusion

New developments and enhancements of the concentric solidification technique that is used in high-temperature laser-scanning confocal microscopy have been presented. These newly developed capabilities have been demonstrated with reference to the peritectic phase transition in the Fe–C system. The use of purposely developed image processing software enables the automatic determination of phase fractions as a function of time and temperature during solidification. The experimental results were reproducible in terms of solidification interface progression and transformation kinetics of the peritectic phase transition when different cooling cycles were used.

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