Concentric Solidification for High Temperature Laser Scanning Confocal Microscopy

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A new experimental technique defined as concentric solidification has been developed to improve in-situ observations of solidification and high temperature phase transformations using laser scanning confocal microscopy (LSCM). The technique consists of applying a radial thermal gradient across a 10 mm diameter sample such that the maximum temperature is focused in the centre of the specimen. Careful control over the sample thickness, heating rate and peak temperature results in the formation of a liquid pool in the centre of the specimen. Surface tension balance between solid, liquid, gas and crucible result in minimal meniscus formation on the liquid pool, leading to a greatly enhanced in-situ observations. Examples of the range of observations possible as well as unique observations of segregation related phenomena are presented.

KEY WORDS: solidification; confocal microscope; phase transformation; in-situ.

1. Introduction

Through the efforts of many researchers a range of elegant and novel techniques has been developed for the study of solidification and solid-state transformation phenomena. Experimental techniques developed specifically to study aspects of solidification morphology include transmission X-ray observation and the Bridgman furnace. Microstructural development in continuous and strip casting has been studied using thermo-mechanical simulators, and the levitating droplet apparatus. An enhanced understanding of solid-state phase transformations has been developed through the use of dynamic techniques such as thermal analysis and dilatometry, and characterisation through the use of electron and optical microscopy. The phenomena studied cover heat transfer, surface tension, interface morphology, high temperature physical properties, and microstructural development. Before concentric solidification is discussed in more detail, it is instructive to refer very briefly to some of the techniques with specific reference to the advantages and limitations of established techniques to study microstructural development.

Several studies have been undertaken using X-ray transmission experimental techniques to observe in-situ solidification events. Sen et al.1) utilised radiography to image in-situ solidification phenomena in metallic systems. This technique provides the ability to observe the morphology of a growth front during solidification. In this study, the interaction between a solid–liquid interface and an insoluble ZrO2 particle was observed. The X-ray source was sub-micron in size, however the maximum magnification utilised was just 32 times, and the size of the particle was 500 μm diameter, indicating the limitations in resolution of the technique. Time Resolved X-ray Diffraction has been used to study the solidification in weld pools of duplex stainless steels. With a spatial resolution of 800 μm and the ability to resolve diffraction patterns every 50 ms, Elmer et al.2) were able to establish that the phase to first form on solidification was delta-ferrite, and they were able to measure in-situ all subsequent transformations. This technique generates information on the kinetics of phase transformations, but not the morphology of interfaces.

Another in-situ observational technique often used in directional solidification studies is the Bridgman furnace technique. Here, a sample is drawn through a furnace, with an imposed thermal gradient, at a controlled velocity. The combination of low temperature, high entropy analogue materials such as succinonitrile with a transparent viewing cell have enabled extensive work to be conducted into the morphology of solidification interfaces. A major advantage of this type of experimental system is that the solidification cell can be designed such that thermal gradients can effectively be considered to be two-dimensional.3) A major limitation of this technique with respect to the study of metallic systems, is that in-situ observations cannot be made on opaque materials.

Optical and electron microscopy are invaluable tools in the characterisation of microstructure resulting from phase transformation, as well as establishing the mechanism and rate by which such transformations occur. The technique typically used to study the morphology of solid-state phase transformations in iron alloys revolves around a quench–arrest–quench cycle, whereby an attempt is made to “freeze” the interface growing at high temperature so it can be subjected to microstructural analysis at room temperature. Although being a well-proven technique, its application is limited in cases where subsequent phase transformations occur. Unfortunately, a study of the important delta-ferrite
to austenite phase transformation in steel falls into this category. In low carbon steels the subsequent decomposition of austenite effectively masks the high temperature phase transformation. One approach intended to overcome this problem is the use of alloying elements such as nickel and/or chromium to stabilise the austenite. This approach in turn raises doubt as to the relevance of the experimental findings to the Fe–C system because the substitutional solutes incorporated to enable such metallographic analysis, have markedly slower diffusion rates compared to interstitial solutes such as carbon, and puts into jeopardy any conclusions about the mechanism and rate of transformations.

High temperature Transmission Electron Microscopy (TEM) and Thermionic Transmission microscopy have been used to study phase transformations in iron-based alloys. For example, Onink et al.\(^4\) used high temperature TEM to study the progression of an austenite–ferrite interphase boundary. They sought to observe nucleation and growth but were unsuccessful in observing nucleation. It was found that the velocity of the austenite–ferrite interface was not constant but exhibited periods of rapid growth and periods where a zero growth rate was approached. An inherent limitation of this technique is the extremely small volume of material that can be observed. The sample needs to be thin enough to allow the transmission of electrons, in the case of iron about one micron in thickness, and in this instance the field of view was only 5 \(\mu\)m by 5 \(\mu\)m. Nucleation of ferrite was not observed, and it was concluded that the influence of the free surface resulted in nucleation occurring in the thicker non-transparent regions of the sample and not in the thin transparent region.

Thermionic Emission microscopy uses the emission of electrons from a thermally activated surface to produce an image of the microstructure. The emission of electrons from a metal surface is dependent upon the work function and the relative energy of the electrons. In this technique the work function is reduced through the application of an activator such as barium or caesium, the energy of electrons is increased through heating, and a voltage is applied to the specimen to reduce the potential barrier. The emitted electrons are focussed onto a fluorescent screen and an image is generated, with contrast dependent upon the number of electrons hitting different areas of the screen. Due to the anisotropic nature of the work function, related to composition and crystallography, different orientations of the same phase emit varying amounts of electrons, and different lattice structures also emit differently. This then leads to a contrast between phases and grains that produce an image similar in appearance to those obtained with optical microscopy.\(^{23}\) This technique is limited in the minimum temperature at which observations can be made. As the temperature decreases the number of electrons emitted also decreases to a point, reported as 450°C, where the number striking the fluorescent screen is insufficient to resolve an image. In thermionic emission microscopy observations are made of the free surface, therefore questions arise as to the correlation of such observations to events occurring in the bulk. Grube and Rouze\(^6\) suggested that for qualitative assessment of free surface effects, serial sectioning combined with optical analysis of the microstructure in the bulk material should be conducted.

A recently developed technique that has attracted much attention is high temperature laser-scanning confocal microscopy.\(^7\)–\(^{21}\) One of the unique features of this technique is that fine scale microstructural development can be studied in-situ and at temperature. This technique can be usefully employed to study in real time solidification events, and the important solid-state transformations in carbon steels, delta-ferrite to austenite and austenite decomposition. Unfortunately the formation of a meniscus in liquid metals has, restricted the use of this technique with regard to the study of solidification due to the limited field of view resulting from the strong meniscus effect. Prior to discussing concentric solidification for overcoming this limitation it is pertinent to briefly refer to some general aspects of high temperature laser-scanning confocal microscopy.

- **Laser Scanning Confocal Microscopy (LSCM)**

  In 1961 Minski\(^{22}\) established the principles of laser scanning confocal microscopy (LSCM), and since then this technique has found widespread application in biological sciences. However, it was not until the 1990’s when Emi and co-workers combined LSCM with infrared heating that renewed interest was developed in high temperature microscopy of metals. Experimental studies conducted with this technique include the morphology of solidification and an analysis of the progress of delta-ferrite to austenite interfaces in low carbon steels\(^8\)–\(^10\); inclusion agglomeration,\(^11\),\(^12\) inclusion engulfment\(^13\)–\(^16\); crystallisation of oxide melts\(^17\); dissolution of alumina inclusions in slag\(^18\); kinetics of the peritectic transformation\(^19\) and solidification phenomena.\(^20\),\(^21\)

  In confocal microscopy, laser light is focused by an objective lens on to the object, and the reflected beam is focused onto a photo detector via a beam splitter, as shown in Fig. 1. An image is built up by scanning the focussed spot relative to the object, which is then stored in an imaging system for subsequent display. Through the use of a confocal pinhole, only light incident from the focal plane is permitted to pass through to the photo detector, represented schematically in Fig. 2. Light not returning from the specific optical plane is blocked by the pinhole. Hence, an extremely thin optical section is created, providing a high-resolution image. Because thermal radiation is also blocked by the confocal pinhole, only the polarised reflection of the high intensity laser beam reaches the imaging sensor and a sharp image is produced. The use of pinhole optics increases the resolution such that with a 0.5 \(\mu\)m diameter beam,
the effective resolution is 0.25 $\mu m$.

Magnifications up to $1350 \times$ at a resolution of 0.25 $\mu m$ can be obtained, using a He–Ne laser with a wavelength of 632.8 nm. In the system used a laser beam, 0.5 $\mu m$ diameter is reflected and scanned by an acoustic optical deflector in the horizontal direction at a rate of 15.7 kHz and a galvano-mirror in the vertical direction at 60 Hz. Specimens are placed at the focal point of a gold plated ellipsoidal cavity in an infrared furnace beneath a quartz view port, as shown in Fig. 3.

A 1.5 kW-halogen lamp located at the other focal point in the cavity heats the specimen by radiation. A quartz plate separates the specimen and lamp chambers so that the atmosphere of the specimen chamber can be controlled and the lamp can be air-cooled. The temperature measured by thermocouples incorporated in the crucible holder is displayed on a monitor and simultaneously recorded with the image on videotape and/or DVD at a rate of 30 frames per second. Hard copies of the video frames can be made or they can be subject to digital video analysis on a computer. Specimen holders consist of 5 or 10 mm diameter round holders or a 12$\times$5$\times$3 mm rectangular holder, constructed from a polymeric end-piece, alumina 2-bore tube with an outer silica support tube and a platinum holder welded to a B type thermocouple wire.

Although the shallow depth of focus in the confocal system is one of the unique features because sharp images can be obtained, it can unfortunately also be the Achilles heel of the technique. The limitation on the focal depth is of special importance when studying molten metal pools, which due to the high surface tensions typical of these systems, leads to the formation of a pronounced meniscus. The implications for conventional LSCM methodology are illustrated in Figs. 4(a) and 4(b) where the meniscus in the liquid phase results in a localized region of brightness. This leads to difficulty in resolving the liquid/solid interface across the whole field of view leading to poor imaging of the interface progression.

In Fig. 4 the morphological stability of a liquid/solid interface was being studied under a thermal gradient in a rectangular crucible. The alloy was a fully ferritic stainless steel, and the morphological stability was being assessed as a function of cooling rate. In Fig. 4(a) the interface has begun to develop perturbations, that develop into the morphology observed in Fig. 4(b). The curvature of the surface as a result of the meniscus effect is observed in Fig. 4(a) region ahead of the liquid/solid interface of high brightness. The high contrast between regions across the field of view results in only a small region being clearly delineated at any one time.

2. Experimental

The concentric solidification experimental method is defined as the formation of a centralised pool of liquid metal contained by a rim of solid of the same material under a radial thermal gradient. In our system, a specimen of 10 mm diameter is placed in an alumina crucible, which in turn is held in a platinum holder, similar to that in Fig. 3. The specimen is positioned at one focal point of an ellipsoid Infra-red heating furnace, with the heat source at the other. A schematic diagram of the liquid pool contained by a solid rim is shown in Fig. 5(a) for a Fe–0.17%C alloy at a temperature above the peritectic temperature (1,757 K). Figure 5(b) shows a section through this specimen following cooling to room temperature.

In the conventional LSCM 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, gen-

![Fig. 2. Confocal nature of the optics.](image1)

![Fig. 3. Infrared furnace and crucible holder system.](image2)

![Fig. 4. Meniscus effects on the resolution of a L/δ interface in conventional LSCM.](image3)
erally proceeds from the crucible wall up the side of the meniscus. In the melt pool configuration the liquid phase is in contact with solid material, the alumina crucible and the gas atmosphere. The resultant surface tension energy balance, in the iron–carbon alloys studied, between these interfaces leads to a significant reduction in the liquid meniscus, and hence a greatly improved image of the liquid metal, as shown and discussed in Figs. 6, 7 and 8.

The ability to establish a melt pool in the confocal microscope is dependent upon the existence of a radial thermal gradient across the 10 mm diameter crucible holder. An estimation of the temperature gradient across the crucible was obtained by observing the melting of pure metal powders scattered over the surface of the alumina crucible. For this purpose silver and nickel (99.99% pure) with melting points of 1 234 K and 1 723 K respectively were used. Silver particles at the centre melted at 20 K higher than those on the edge. Nickel powder melted over an 80°C temperature range from the centre to the edge. In both cases particles at the centre of the crucible melted first and particles at the edge were the last to melt indicating that the centre of the crucible is the hottest point. Whereas a radial temperature gradient of 5 K/mm is imposed at 1 234 K, it is clear that this gradient increases with temperature and at 1 723 K the gradient increases to 20 K/mm. The increase in temperature gradient with absolute temperature is in agreement with earlier work, using a 15 mm×5 mm rectangular crucible, measured the temperature along the crucible and also found that the temperature gradient increases with increasing absolute temperature.23)

The thickness of the sample plays a pivotal role in successfully creating a stable and sustainable liquid pool. It has been established for Fe–C alloys of a peritectic composition that the specimen thickness needs to be <250 μm. It was not possible to form a stable liquid pool in thicker samples of this steel and it seems that the formation of a stable ‘pool’ is related to the thermal distribution within the specimen. As specimen thickness increases, conductive heat flow from the centre to the edge is improved, thereby preventing the formation of a sufficiently large thermal gradient to stabilise the pool. Some evidence in support of this premise is found in the inability to produce concentric solidification conditions in pure silver, with a thermal conductivity 10 times higher than steel, even with specimens as thin as 50 μm. A beneficial consequence of using a thin sample is that the through-thickness thermal gradient approaches zero leading to the formation of a vertical solid–liquid interface,

![Fig. 6.](image)

Floating delta-ferrite particles, Fe–0.17%C, LSCM, concentric solidification technique. 4 features are of interest being (i) and (ii) floating precipitates of delta-ferrite, (iii) an agglomeration of alumina particles and (iv) a liquid oxide inclusion. The floating precipitates (i) and (ii) are of interest as they are examples of equiaxed grains forming in the melt ahead of the main solidification front as displayed in the second frame. In the 4th frame, the growth of a primary dendrite can be followed by the appearance of secondary dendrite arms on the surface.
as shown in Fig. 5(a). The significance of this is that observations made of the free surface can be more confidently attributed to events occurring in the bulk as the direction of growth will be primarily in the plane of the specimen. In thick specimens there is a thermal gradient through the sample thickness potentially leading to a disturbed liquid–solid interface, and uncertainty of the precise direction of growth.

A further factor that has a determining influence in the ability to establish a liquid pool is the sample surface finish. Primarily, a uniform surface finish is required to ensure an even radial thermal distribution. Rough spots have been observed to result in additional localised heating with an associated destabilisation of the melt pool and subsequent pool rupture or escape (melting of the solid rim).

3. Results

3.1. Solidification Experiments

The development of this experimental methodology has proven to have numerous advantages over the conventional experimental methodology and this new technique has significantly extended the use of LSCM. Most notable is the minimisation of curvature resulting from the meniscus effect, which result in the ability to focus across a solid–liquid interface, and across extended liquid pools. In comparing Fig. 6 with Fig. 4 it is clear that in the concentric solidification experiment the whole field of view is in focus and of almost equal light intensity. In this instance, solidification in steel of peritectic composition has just begun and the growth of solid delta-ferrite can be observed as floating, solid precipitates on the liquid surface.
In Figs. 7 and 8 two examples of the peritectic phase transformation are shown, with both the liquid and solid phases in focus. Because the surface of the liquid pool is flat in the concentric solidification experiment as opposed to a pronounced meniscus in the conventional technique, the field of view is much greater, leading to the capture of greatly enhanced images. The fact that both phases are in focus in the area of view, means that greater accuracy in the measurement of the velocities of the liquid/delta, liquid/austenite and austenite/delta phase boundaries can be achieved during cooling. Of equal importance is the fact that the solid/liquid and δ/γ interfaces are vertical and extend from the top to the bottom of the specimen. Hence, the mode and rate of progression of these interfaces can be determined with a high degree of accuracy.

3.2. Segregation

The segregation of solutes during the solidification process has an important impact upon the properties of cast structures and an important advantage of this particular geometry of the experimental design is that such segregation events can be studied. Again, it is the minimisation of meniscus effects that greatly improve the quality of observations made of events occurring during segregation. The first example presented here involves the precipitation of non-metallic inclusions in the inter-dendritic liquid of a steel of peritectic composition. In Fig. 9, LSCM images are reproduced following the final liquid phase transition during a concentric solidification experiment. In the first frame a liquid pool is still present, in the next frame the pool is displaced by the solid phase growing from underneath the pool, leaving thin films of the solute enriched liquid between the solid regions. In the third frame precipitation of a third phase, appearing as dark irregular structures, commences, and continues in the last frame. Analysis of this sample in a scanning electron microscope and using EDS, Fig. 10 revealed that the precipitates formed were a titanium rich phase.

3.3. Austenite Decomposition

The segregation observed during solidification in concentric solidification also have important implications for subsequent solid-state phase transformations. In Fe–C alloys austenite decomposition plays a critical role in the control of microstructure and hence the mechanical properties of a specimen. Depending on the cooling rate, prior deformation and carbon content, a range of products can form by the decomposition of austenite. These decomposition prod-

![Fig. 9. Precipitation of non-metallic precipitates in inter-dendritic liquid.](image)

![Distribution of Fe](image) ![Distribution of Ti](image)

![Fig. 10. EDS analysis of precipitates shown in Fig. 9.](image)
ucts include \(\alpha\)-ferrite, Widmanstätten and Bainitic ferrite plates, pearlite, cementite and martensite. The mechanism by which some of these products nucleate and grow is not fully understood yet and both the mechanisms of nucleation and growth are the subject of considerable debate in the literature. An example of an interesting experiment that can be performed using a concentric method involves the “quenching” of Widmanstätten ferrite plates by the growth of pearlite, shown in Fig. 11. The thermal cycle that leads to the establishment of a stable liquid pool requires long hold times, in the order of minutes, at elevated temperature. As such the grain size of the delta-ferrite is large, promoting the formation of Widmanstätten ferrite plates during austenite decomposition. The radial thermal gradient leads to the nucleation of Widmanstätten ferrite close to the outside edge of the specimen, followed by growth towards the centre of the specimen. As the plates grow into the central region of the sample, where the earlier presence of a liquid phase has resulted in the concentration of solute being high, pearlite nucleates ahead of the growing plates. The pearlite grows at a rapid rate, sweeping around the Widmanstätten ferrite plates, trapping them in-situ.

In order to further study the Widmanstätten/pearlite growth the specimen that was studied in the LSCM (Fig. 11) was prepared for optical microscopy and the same area is shown in Fig. 12. The tip structure of the Widmanstätten ferrite plates is clearly defined after being ‘frozen’ in-situ by the progression of the pearlite interface. Although a detailed discussion of Widmanstätten growth is not pertinent to the present discussion, it is apparent that a combination of LSCM and other microstructural techniques may lend itself to the study of the interfacial structure of Widmanstätten ferrite plates. Future work will be conducted using transmission electron microscopy to probe this issue further.

4. Conclusions

The melt pool technique has been developed to examine the effects of cooling rate on the stability of a solidification

Fig. 11. Pearlitic/Widmanstätten growth in 0.42C steel. (a) Widmanstätten ferrite plates (W) grow from the bottom right-hand corner, (b) a pearlite colony (P) nucleates and grows from the bottom left-hand corner, (c) the first pearlite colony sweeps across the sample consuming austenite and isolating individual Widmanstätten ferrite plates, a second pearlite colony grows from the top of the image, (d) the Widmanstätten ferrite plate colony is trapped in-situ by the pearlite which continues to grow into the austenite. The location of the pearlite has been highlighted with a broken line as while the path of the transformation can be followed easily on the video, it is poorly defined in the individual frames shown in this figure.

Fig. 12. Optical images of transformed sample (a) pearlite (P), Widmanstätten ferrite plate (W), prior austenite twin boundary (Twin). The Twin shown in these photomicrographs at ambient, correspond to the twin shown in Fig. 11.
interface as a function of cooling rate. Using this new technique it has been shown that the peritectic reaction as well as the progress of the peritectic can be studied and that the rate of the transformation as a function of cooling rate can be determined to a high degree of accuracy.

This new technique offers unique opportunities to study segregation during segregation, non-metallic precipitation and the distribution of non-metallic inclusions such as alumina. Segregation of carbon in Fe–C alloys leads to a new ways in which the fundamental structure and growth mechanism of ferrite plate morphologies during austenite decomposition can be studied.

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