Time-resolved X-ray diffraction studies of solidification microstructure evolution in welding

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Abstract

High-brilliance and high-energy polychromatic X-ray diffraction (XRD) has been used for time-resolved studies of rapid solidification microstructure evolution in situ during simulated spot welding in austenitic steels. Weld pools of 5.0 mm diameter and 0.6–1.5 mm depth were formed at the steel plate surfaces by radiation heating from halogen lamps. Solidification was initiated by powering off the lamps, and completed within 1.5 s while measuring diffraction from the solidifying grains of the pool at a 1 kHz frame rate. The data contain time-resolved information on individual grain growth and overall solid fraction evolution, and furthermore reveal prominent individual and collective motion of grains during early stages of solidification, presumably caused by convective currents in the pool. Ultrafast polychromatic high-energy XRD is novel in studies of microstructure evolution during welding. The experimental technique could quite readily be used for similar studies in real welding.

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1. Introduction

In fusion welding, metals coalesce via extremely rapid solidification where liquid → solid phase transformations controlled by mass and heat transport occur under conditions far from equilibrium. The transport conditions are decisive to the microstructure pattern formation and morphological evolution of the advancing solidification fronts, as well as to chemical segregation that may develop in the weld pool. As a result, the final microstructure and mechanical properties of the weld, especially in critical regions where the terminal solidification fronts meet and weld failures often occur, will be dictated by the full dynamic history of the growth processes. In most approaches to welding, the weld fusion zone (FZ) is considered as a mini-casting and its solidification behaviour is handled by extrapolation of simulation models developed for solidification processes\cite{1,2} at cooling rates that could be orders of magnitude lower than those that apply to real welds\cite{3–6}. Improvements in computational capabilities have allowed for numerical simulations of various advanced solidification processes and are often guided by or verified via in situ experiments\cite{1,2}.

Experimental studies have also been reported for weld solidification\cite{7–9} or welding-related solid-state transformations\cite{10,11}, employing X-ray diffraction (XRD) techniques. However, the high cooling rates, corresponding
rapidness and complexity of welding processes have made direct in situ experimental studies of the solidification part of the process extremely demanding. In a recent paper, Yonemura et al. [9] report an in situ XRD study of microstructure evolution in arc welds, but mainly with a focus on post-solidification transformations. Nevertheless, the first 2 s or so of observations are associated with the actual solidification part of the process. Yet, besides a few rather vague assumptions, the observations corresponding to solidification are not analysed in any greater detail to yield results on individual grain growth. The data are also concerned with features, such as prominent diffuse X-ray scattering, which are not fully accounted for. Thus, solidification dynamics or individual grain growth in actual metallic welds has not yet been studied in any detail.

The present work provides novel time-resolved data on individual dendrite evolution during spot welding of an austenitic steel, studied in situ by polychromatic synchrotron ultrafast XRD.

2. Experimental

Samples were cut from 2 mm thick rolled and annealed Fe–Cr–17.3% Ni–11.1% Mo–2.1% C < 0.1% austenitic steel plates into 30 mm × 20 mm rectangular pieces.

The primary heat source employed to simulate a real spot-welding process comprised seven 1 kW halogen lamps equipped with elliptical reflectors, each yielding a circular focal spot ~4 mm in diameter. The lamps were mounted in a custom-built rig that allowed for individual adjustments to superpose all lamps into a common focal spot at the centre of the sample surface with closely circular cross-sections ~5.0 ± 0.5 mm diameter, at a controlled position from one sample to the other to a precision ±1.0 mm. The sample was placed on top of a boron nitride plate heated from below by a resistance heater, allowing for cooling rates to be adjusted during pool freezing. A controlled argon flow could be brought in at the sample surface just adjacent to the weld spot to reduce oxidation rates at the pool surface. Weld pool depths varied between 0.6 and 1.5 mm, depending on the setting temperature of the resistive heater and on the Ar flow rate. During each in situ welding experiment, the resistive heater was kept at a constant temperature and the Ar flow set at a fixed rate, before switching on the halogen lamps to full power.

Typically melting initiated after a few seconds, followed by stabilization of a molten pool within 1–3 s. As soon as complete melting in the part of the pool illuminated by the incident X-ray beam was verified, pool freezing was initiated by powering off the halogen heaters (the power-off time is referred to as $t = 0$ s hereafter). Typically, the observed region of the weld pool re-solidified completely within 1.0–1.5 s.

The experiments were carried out at the ID15A beamline at the European Synchrotron Radiation Facility. For each welding experiment, a sample was placed on top of the resistive heater with the surface normal tilted 0.035 rad towards the incident beam (see Figs. 1 and 2). The incident X-rays were polychromatic, i.e. a white beam as delivered at the beam line by the U22 undulator, with photon energies in the range 50–150 keV, and defined by apertures upstream of the sample to a cross-section of $1 \times 0.1 \text{ mm}^2$ (H × V). The incident beam was positioned concentric to the pool surface centre, and with the sample tilt and the incident beam geometry, the region monitored throughout the experiments corresponded to a 1 mm wide region stretching diametrically across the pool surface, with maximum penetration into the pool of ~140 µm towards the exit beam side (see Fig. 2). Due to the low sample tilt angle with respect to the incident beam and the angular coverage of the detector, only X-ray photons diffracted at angles $<\sim 0.15$ rad relative to the sample surface are registered in the two-dimensional (2-D) detector. In a typical spot weld, columnar grains grow towards the centre of the weld from the FZ periphery, opposite to the heat flow. For the experiments present here, the X-ray beam width is small compared to weld pool diameter, and accordingly, columnar grains under X-ray illumination would be expected to grow nearly parallel or anti-parallel to the beam direction in the surface-near regions of the weld, as indicated schematically in Fig. 2.

During experiments, temperature was monitored by a thermocouple placed inside the boron nitride plate on top of the resistance wires. XRD was collected in reflection geometry in one quadrant of the upper hemisphere in the forward scattering direction, and at a distance, $d = 920$ mm from the sample, spanning an angular range $2\theta \in [0.06, 0.3]$ rad. Diffraction data were collected with a PCO.dimax® complementary metal oxide semiconductor (CMOS) equipped with an image intensifier. The CMOS spans a $2016 \times 2016$ pixel array, and was read at a 1.0 kHz frame rate. In total, 24 XRD data sequences were collected in situ during simulated spot welding.

3. Polychromatic XRD and data processing

The diffraction condition with polychromatic X-rays is best illustrated in reciprocal space. In the monochromatic case, reciprocal space illustration of the diffraction condition is by the so-called Ewald sphere construction [12]. By selecting an arbitrary reciprocal lattice point as the origin, and letting the incident wave vector, $k_0$, terminate at this lattice point, the diffraction condition is constructed by drawing an Ewald sphere with radius $|k_0|$ about the position from which $k_0$ originates. Any other reciprocal lattice points, with reciprocal lattice vector $h$ relative to the origin, are said to be in diffraction condition if they intersect with the surface of the sphere, with a diffracted wave propagating along $k_h$, as can be seen in Fig. 3A. Rotation of the crystal is equivalent to a rotation of the reciprocal lattice about the origin relative to a fixed sphere, and brings new reciprocal lattice points into diffracting position, as shown in Fig. 3b. If the material is polycrystalline (or multiphase), there will simply be one set of reciprocal lattice
points associated with each crystallite (or phase), all with coinciding origins where \( k_0 \) terminates.

The polychromatic situation is quite readily illustrated by imagining a series of Ewald spheres, one for each discrete wavelength component. If the X-ray source is continuous over a range of X-ray photon energies, it suffices to draw the Ewald spheres corresponding to maximum and minimum photon energies, as shown in Fig. 3c. Now, every reciprocal lattice point which falls inside the volume spanned between the delimiting upper and lower energy spheres is in diffraction condition for some given wavelength component of the source spectrum.
There may be several aspects that distinguish a typical monochromatic X-ray diffraction experiment from a polychromatic one, but here the focus will be only on the points most crucial to this particular study. In order to resolve individual grain growth dynamics taking place in a weld pool during freezing, it is necessary to track and monitor a set of reflections as a function of time throughout the experiment. For the monochromatic case this may be extremely difficult or even impossible, as the diffraction condition by intersection of the lattice node with a discrete stationary surface is highly sensitive to any changes in the reciprocal lattice during the process, such as might be caused by e.g. grain rotation or thermal contraction of the crystal lattice. Thus, in the monochromatic situation individual reflections may only be followed through a few time frames, and as a result the experiment would fail to bring about thorough-process information on the dynamics and kinetics of individual grains. In the polychromatic case, however, tracking of specific reflections is robust towards changes in the reciprocal lattice as long as the lattice node remains inside the diffraction volume. Thus, extraction of through-process data for individual grains is possible and robust towards dynamic effects like crystal re-orientation, thermal contraction, lattice parameter changes by stresses, solid solutions, etc. On the other hand, indexing of reflections becomes more cumbersome in the changes by stresses, solid solutions, etc. On the other hand, indexing of reflections becomes more cumbersome in the polychromatic case compared to a monochromatic one, in particular when the sample is polycrystalline.

As mentioned above, XRD data were collected frame by frame at a fixed rate of 1.0 kHz. Each frame comprises a set of raw pixel intensities that must undergo a few processing steps to arrive at useful quantities. Background intensity caused by diffuse scattering from air or the liquid metal was assessed from frames captured with the weld pool complete-ly molten, and found to be weak and fairly constant across the solid angle covered by the detector. Accordingly, corrections could be performed simply by subtracting the mean background intensity value \( J_{\text{bck}} \) from all pixels to arrive at a set of net pixel intensities for each frame. After background intensity subtraction, separable diffraction peaks emerged as islands of interconnected pixels with net intensities \( J_{\text{net}}(r_h, \psi_h, t) \), delimited by the \( J_{\text{net}}(r_h, \psi_h, t) > 2 \sigma(J_{\text{bck}}) \) boundary, with \( t \) as the time stamp of the frame, \( \sigma(J_{\text{bck}}) \) the standard deviation of pixel intensities calculated from the background frames and \( r_h \) and \( \psi_h \) representing radial and azimuthal polar coordinates in the detector plane, respectively. The situation of having net intensities superimposed on a flat background is favourable for data processing since it allows morphological rendering of diffraction peaks by a simple \( J_{\text{net}} > 2 \sigma(J_{\text{bck}}) \) 2-D threshold masking of the background-corrected data. Due to high X-ray energies involved and small angular range covered, relative corrections for beam polarization and X-ray absorption is reasonably uniform and modest for all data and may be neglected. The scattering power itself also includes X-ray-energy-dependent terms and can be shown to scale with \( k^{-3} J_0(k) \), where \( k = |\mathbf{k}_d| = |\mathbf{k}_h| = 1/\lambda \) is the wave vector magnitude, \( \lambda \) the X-ray wavelength and \( J_0(k) \) the photon-energy-specific power density of the incident beam at the sample position. A prominent dynamic feature of the experiment(s) was substantial grain motions, observed as angular motions of the diffraction peaks, in particular during the earlier stages of freezing. These made it necessary to track and follow reflections along their angular trajectories, requiring corrections to be made as a reciprocal lattice node move and the X-ray energy at which it diffracts is shifted. The ID15 U22 undulator source delivers a \( J_0(k) \) which varies by orders of magnitude over the relevant energy range. This needs to be corrected for, and may not be entirely straightforward, since it requires knowledge of the exact length of the reciprocal lattice vector and its orientation at any time to precisely assign the correct diffracting position and X-ray energy. The angular rotations encountered here were three to four orders of magnitude larger than those that would result from thermal contraction \([13,14]\), stresses or any other change of the lattice parameter, and could accordingly be ascribed mainly to rigid body rotation of the reciprocal lattice. Therefore, neglecting to a first approximation changes of the lattice parameters during grain growth, the intensity data may be corrected under the constraint of \( |\mathbf{h}| = \text{constant} \), for which it becomes possible to determine the exact diffraction geometry \( \mathbf{k}_d(t) \), and thereby also \( J_0(k(t)) = J_0(t) \) uniquely for any spot in every frame. After corrections, every frame consists of a set of scaled pixel intensities, \( J_{\text{sc}}(r_h, \psi_h, t) = k(t)^3 J_{\text{net}}(r_h, \psi_h, t)/J_0(t) \).

After scaling the next processing step is intensity integration for individual diffraction peaks or reflections. Let \( I_h(t) \) denote the integrated intensity associated with reflection \( \mathbf{h} \). \( I_h(t) \) is computed directly from the scaled pixel intensities of the corrected images as

\[
I_h(t) = \sum_{r, \phi} J_{\text{sc}}(r, \psi, t)
\]

where the sum is taken over all interconnected pixels inside the \( J_{\text{net}} > 2 \sigma(J_{\text{bck,sc}}) \) bounded region. Another useful quantity to track in order to analyse lattice parameter changes or rotation of the grain is the peak centre of mass (c.o.m.) \( \{r_h(t), \psi_h(t)\} \), given by

\[
r_h(t) = \frac{\sum_{r, \phi} J_{\text{sc}}(r, \psi, t)r}{I_h(t)}
\]

\[
\psi_h(t) = \frac{\sum_{r, \phi} J_{\text{sc}}(r, \psi, t)\psi}{I_h(t)}
\]

where the sums are taken over every pixel \( \{r, \psi\} \) inside the \( 2 \sigma(J_{\text{bck,sc}}) \) bounded regions. It was also found convenient to keep track of the time evolution of the peak area, represented by the peak full widths, \( \{\sigma_r(t), \sigma_\phi(t)\} \), along the radial \( r_h(t) \) (Eq. (2)) and azimuth \( \psi_h(t) \) directions (Eq. (3)), respectively.

It follows from the theory of kinematical diffraction that \( I_h \) from a single grain volume bathed in the incident beam can be expressed as \([12]\)
\[ I_h = V_c^{-2} K_h |F_h|^2 \cdot V_g \]  

(4)

where \( V_c \) is the volume of the unit cell and \( V_g \) the X-ray illuminated grain volume. \( K_h \) is a factor that collects reflection-dependent experimental corrections and universal physical constants, and the former may be neglected here as all required corrections have been done on the pixel intensities. Finally, \( |F_h| \) is the amplitude of the structure factor. As long as crystal growth is analysed on a relative scale, and under the assumption that the effect of thermal contraction can be neglected, the time evolution of the crystal volume can be expressed in terms of a relative fraction:

\[ f_g(t) = \frac{V_g(t)}{V_\infty} = \frac{I_h(t)}{I_\infty} \]  

(5)

where \( V_\infty \) and \( I_\infty \) represent the final X-ray illuminated grain volume and the corresponding intensity scattered in direction \( k_h \) at sequence termination when the weld pool is completely solidified.

4. Results and discussion

The 24 sequences collected for the weld simulations were found to be quite similar in a statistical sense in terms of grain motion, individual and overall grain growth. Typically, the first diffraction spots appeared at about \( t \sim 100 \text{ ms} \) relative to power-down of the primary heat source at \( t = 0 \). Most diffraction spots start to appear within \( t \sim 100-400 \text{ ms} \), although in some sequences a few new spots are found to appear in the images at later times. The geometry of the experiment does not allow us to conclude directly from the X-ray images whether the spots that appear correspond to nucleation events or to situations where crystals grow from the outside and into the X-ray illuminated region. Yet, from post-experimental metallography on some of the weld pools, as shown in Fig. 4, it seems likely that the grains form predominantly at the melt–solid interface at \( t \sim 0 \) and grow into the X-ray illuminated region, as illustrated schematically in Fig. 2, within the first 300–400 ms or so. Post-experimental metallography shows evidence of substantial convection also being present in the pool, with a depletion at the weld centre and a protruded ring on top of the original sample surface extending outwards from the periphery of the original melt interface.

In the early stages of pool freezing, \( t < 400 \text{ ms} \), individual diffraction spots move along closely fixed trajectories with quite substantial angular velocities \( \omega_x = \frac{dx}{dt}, \omega_\phi = \frac{dy}{dt} \), where \( \phi_x = \tan^{-1}(r_y/d) \). Fig. 5 shows \( \phi_x(t) \) and \( \psi_y(t) \) evolution for 10 representative grains, selected from two different welding sequences. The first peaks that appeared before \( t < 250 \text{ ms} \) showed very high \( \omega_x \), typically \( > \sim 1 \text{ rad s}^{-1} \), and disappeared from the angular field of view covered by the detector within some tens of milliseconds or less. Around \( t \sim 250 \text{ ms} \), the angular motion reduced by at least one order of magnitude, however the motion still remained prominent for most peaks. At around \( t \sim 700 \text{ ms} \) and later, angular motion reduced to rates \(< 0.001 \text{ rad s}^{-1} \), which is proximetrically consistent with thermal contraction at 200–400 K s\(^{-1}\) cooling rate. Accordingly, angular motions at \( t < 700 \text{ ms} \) are assumed to relate exclusively to re-orientation/rotation of the diffracting grains.

Generally, spots appearing before \( t < 250 \text{ ms} \) pop up somewhere inside the 2-D detector area, move some distance across the field of view and then disappear suddenly, well before reaching the edges of the detector. Together with the post-welding metallography, this supports a conclusion that the initial spots originate from free crystals that eventually move completely out of the X-ray illuminated region. We cannot firmly conclude whether these are fragments that detach from a columnar growth front or equiaxed dendrites that form in the melt. Their sudden appearance may indicate that the free grains form predominantly outside the X-ray illuminated region. As seen in Fig. 4, a particular region with equiaxed dendrites fairly close to the original edge of the melt pool coincide with the area where grains would enter into the incident X-ray beam. The peaks that start to appear at \( t > 250 \text{ ms} \) are clearly distinguishable from the initial free crystals not only by their maximum angular velocities, but also by remaining within the X-ray illuminated volume throughout the experiment.

The archetype grain re-orientation/rotation dynamics are further illustrated in Fig. 6, and arrangement of the observations in this manner allowed individual crystals to be ascribed into: (i) ultrafast moving free crystals (f) appearing at \( t < 250 \text{ ms} \) (typical example are illustrated by f1, f2 and f3) and (ii) columnar crystals (c), appearing at \( t > 250 \text{ ms} \) (typical examples are illustrated by c1, c2 and c3). It can be seen that c-crystals show reduced motion at early stages, within one order of magnitude slower than those of the f-crystals at the same time. Crystals of the

Fig. 4. Typical experimental weld cross-section micrographs sectioned along the beam direction. (a) Full weld cross-section from a post-experimental sample. (b) Half-cross section detailing an experimental weld pool. A depleted centre zone (i) and a significant bulging at the weld pool periphery (ii) is observed in the post-solidified samples.
$f$-category show consistent and very high angular peak velocities before disappearing from the illuminated region within $\sim$80–150 ms. Assuming equiaxed grain sizes as those found in the post solidified metallographic samples and the diffraction peak motion rates, we accounted for metric rotation velocities at the free-crystal periphery up to $\sim 5$ mm s$^{-1}$. Motion of $c$-type crystals reduces exponentially until $t \sim 450$ ms, where an abrupt decelerating transient by 2.5–3 orders of magnitude to more or less fixed orientations occurs. After this transient, the crystals remain stationary with respect to diffracting positions within the limits that could be ascribed to thermal contraction effects.

For each sequence, the time evolution of a set of individual $c$-type grain volume fractions, $f_g(t)$, was extracted as outlined in Section 3 (see Eq. (5)). Typical growth curves are presented in Fig. 7, by data selected from two sequences. Note that no smoothing has been carried out with the $f_g(t)$ data, such that the curve smoothness encountered serves to underline high quality in terms of diffraction signal statistics, as well as soundness in the correction scheme applied. These crystals would continue to grow within the X-ray illuminated region throughout the remaining pool freezing time, as illustrated for the cases $c_1$ and $c_2$ in Fig. 7a, as well as for all four $c$-crystals in Fig. 7b. $c_3$ in Fig. 7a grows in a direction at a much higher angle to the incident beam and therefore relatively rapidly extends outside the X-ray illuminated region. $c_4$ in Fig. 7a is a late arriver, assumed to grow semi-parallel to the surface plane, but directed at a higher angle with respect to $k_0$.

The hypothesis of convection-driven crystal motion at early stages is further supported by observations for the $c$-type crystals. $c$-Crystals that can be tracked throughout

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Fig. 5. Observed $\phi_u(t)$ and $\phi_w(t)$ evolution. (a and b) Evolution in the first 500 ms of solidification from two experiments, referred here as $a$ and $b$, respectively. (c) Time evolution of $\phi_u(t)$ and $\phi_w(t)$ from experiment $a$, for complete solidification time 1500 ms. $f_1$, $f_2$ denote two peaks from each sequence representative of the free grain motion, while $c_1$, $c_2$ and $c_3$ denote three peaks representative of the columnar grains.

Fig. 6. Time evolution of angular velocities, including standard errors, for six re-orienting crystals. A logarithmic scale is used for better illustration of the velocity range accounted for during grain motion. Velocities are binned every 5 ms to enhance data readability.
the full pool freezing experiment are grains that grow with their columnar tip directions at relatively modest angles to \( \mathbf{k}_0 \). In each experiment, the simultaneous appearance of some of the diffraction peaks allowed us to identify possible reflection pairs or triplets, likely to belong to the same columnar crystal. If one assumes \( h_{100} \) as the preferred tip-growth direction for austenitic steel dendrites [6,9], it allowed peak-pairs or triplets from classes \([002], [020] \) and \([022] \) to be indexed, corresponding to \( h_{100} \) columnar crystals growing at angles <\( \frac{\pi}{24} \) rad to \( \mathbf{k}_0 \). As the grains detected and tracked have similar orientations, and are somewhat constrained in motion by being semi-parallel to nearby primary branches, the associated diffraction spot motions follow along relatively straight lines in reciprocal space. To understand more of the nature of this motion, one has to consider several possibilities. If the motion results in a shift of \( r(t) \) and/or \( w(t) \), it would correspond to re-orientation of the whole crystal, i.e. rigid body rotation. This would be consistent with tilting of the whole columnar branch, probably by bending at its attaching root to the original solid [15]. If the motion results in an increase in the profile width along the same direction as \( \omega_r + \omega_w \), it would relate to local re-orientation along the branch, i.e. peak-broadening effects associated with lattice strain. This would be observed if the liquid–solid momentum transfer caused the dendrites to bend along the primary stalk. It is, however, necessary to keep in mind that peak broadening may also result from other phenomena, such as crystal growth, as the diffraction spot area is basically a projection along \( \mathbf{k}_0 \) of the diffracting grain. Fig. 8 presents the time evolution of the peak widths, \( \sigma_r \) in the radial direction (along \( \mathbf{k}_0 \)) for the intensity–grain volume curves presented in Fig. 7. It was found that the change in peak area was more or less entirely directed along \( \mathbf{k}_0 \), with the increase along \( \Psi_h \) being very modest or even negligible.

Thus, to distinguish these two possible sources it is convenient to compute the average time correlations, \( \rho(\sigma_r(t), x(t)) \), between peak broadening \( \sigma_r(t) \), and the corresponding crystal fraction, \( x(t) = f_g(t) \), and crystal rotation, \( x(t) = \omega(t) \), respectively. The results, as extracted from 36 crystals taken from three sequences, yield correlation \( \rho(\sigma_r(t), f_g(t)) = 0.79 \pm 0.12/–0.09 \), whereas \( \rho(\sigma_r(t), \omega(t)) = 0.37 \pm 0.14/–0.16 \). Thus, peak-broadening effects are rather strongly correlated with growth, and hence the predominant rotation for \( c \)-crystals seems to be tilting, i.e. rigid body motion of the primary stalks by bending at their attaching roots [15]. Liquid–solid momentum transfer from convection implies that the columnar dendrites tend to move with the shear flow direction at the solidification front. It should be noted that the neck (base) of rapidly growing dendrites are generally observed to be much narrower than the primary dendrite trunk. Furthermore, as
the temperature approaches the melting point, the elastic moduli of γ-iron, as with many other metals, is expected to reduce drastically [16]; which may also be supported by rather crude extrapolation from low temperature data [17]. The presence of shear flow also means that solute-enriched liquid ahead of the tips is constantly taken away, at least in the early stages of growth, which again should have a prominent effect on the growth rates.

Compared to the previous experimental attempts on in situ characterization of phase transformations in welding by XRD [7–11], the main technical difference of the current experiment is the use of polychromatic X-rays. Even though data processing is more cumbersome for the latter, it is pivotal for successful intensity integration and peak position tracking during solidification if grains are re-orienting in consequence of the processes involved. In fact, it would not have been possible to track individual grain growth or grain rotation, as evident here, in solidifying weld pools by monochromatic X-rays. Even though Yonemura et al. [9] in their monochromatic X-ray study mainly report on transformations that take place after solidification, they also briefly report observations of “mist-like” data in the very first few frames that are ascribed to solidification. Given that the frame rates reported are in the range 10–100 Hz, it is reasonable to assume, the rate is inadequate to capture grain growth in a truly time resolved manner during the actual weld pool freezing stage. At least if the pool solidified completely within 1–2 s, like in the current study, which also compares well to typical pool solidification times reported in literature [4–6]. It therefore seems tempting to suggest that the observations of Yonemura et al. of diffuse scattering during solidification, at least in part, can be ascribed to time blurring during single frame exposure by crystals rotating in and out of the diffraction condition. Accordingly, the current study may be the first to report on real individual time-resolved crystal growth and solidification related dynamics in freezing weld pools. Columnar grain orientations seen in the weld cross-section micrographs in Fig. 4 support the assumption that the heat flow is closely radially symmetric from the weld centre spot. A simple linear extrapolation to approximate the cooling rate at the initiation of solidification in the pool was carried out from cooling rates extracted from thermal contraction of the lattice at \( t > 750 \text{ ms} \), i.e. after columnar grain motion has come to a complete stop. The cooling rates extrapolated linearly in time to \( t = 0 \text{ s} \) are in excess of \( 1000 \text{ K s}^{-1} \), and reasonably close to the cooling rates associated with similar size welds [18]. Further comparison to Elmer et al. [18], suggests that the average thermal gradient across the FZ centre to the periphery could be \( \sim 200 \text{ K mm}^{-1} \). In future extensions of the method presented, further efforts should be made to extract spatiotemporal data on the temperature field in the pool.

Even though our XRD data analysis suggests considerable convective effects in the weld pool, the primary heating employed in the experiments did not involve any electromagnetic arc or plasma as common to many real fusion welding processes. Accordingly, flow is presumed to be driven mainly by a combination of high cooling rates and the surface tension/chemistry of the molten pool, in line with computational studies found in the literature [5,6], where considerable flow velocities are predicted, even in the absence of any arc pressure or Lorentz forces. The maximum possible flow velocity in the melt pool calculated for the experimental conditions via an analytical relationship [4] is over \( 400 \text{ mm s}^{-1} \). Weld-centre depletions and edge-region protrusions found in the welded samples also confirm the presence of a relatively strong convection which drives melt from the centre and out of the weld pool. The detected crystal motion, which is most prominent at early stages, is believed to be caused mainly by liquid–solid momentum transfer and related back to convection as the driving force. Correlating the grain motion and cessation of it with the grain growth; as shown in Fig. 7, it is readily seen that the overall solid volume fraction, \( f_{\text{avg}} \), and individual crystal volume fractions, \( f_g \), at the same time are \( \sim 0.4 \), which could support the conclusion that at this time the solid fraction in the illuminated pool region reaches a value where there is simply not enough bulk liquid left to uphold appreciable convection.

For quantitative comparison of columnar growth with convection-driven tilting, approximate time-dependent crystal length evolution, \( l(t) \), can be extracted via a parabolic envelope fitting to \( f_g(t) \) and the average columnar dendrite length, as measured from the solidified sample micrographs. The resulting tip growth velocities, \( d l(t)/d t \) are found to exceed \( 4 \text{ mm s}^{-1} \), and on average, the columnar grains peak at growth velocities of \( 2.4 \text{ mm s}^{-1} \). At the emergence of the \( f_g(t) \) curves in Fig. 7, the very initial tran- sients [19] in tip growth rates are missing. However, we can anticipate that \( c \)-dendrites may set out to grow at the regions outside the X-ray illuminated volume, mostly at the boundaries of the weld pool and initial transients in these regions are not illuminated by our incident X-rays. From \( l(t) \) it is also possible to calculate metric tilting velocities, \( \dot{h}(t) \) \( \omega_0(t) \), for the columnar tips. Fig. 9 presents the maximum values for tip-tilting velocities, which have been extracted for 36 columnar grains from three spot-welding.

![Fig. 9. Statistics for the maximum dendrite tip-tilting velocities estimated for 36 different columnar crystals analysed in three separate welding sequences. Crystals that appear after \( t \sim 700 \text{ ms} \), when motion terminates, have been excluded from the data.](image-url)

sequences. The data reveal a distribution where $\sim 80\%$ of the crystals have maximum tip-tilting velocities of $\geq 0.5 \text{ mm s}^{-1}$.

Experimental evidence of strong crystal rotations and tilting from the root is novel to the case of welding or similar rapid solidification processing, and apparently not incorporated in any simulation models, although its presence in principle could be rationalized hypothetically, as it is well-recognised that many real welding processes are concerned with very strong convection currents [4–6]. When growth and tilting are compared, especially at the initial stage of columnar growth, i.e. $\sim 250 \text{ ms} < t < \sim 450 \text{ ms}$, the columnar crystal growth velocities and the tilting velocities are found to be of the same order of magnitude, as illustrated in Fig. 10.

Thus, it seems reasonable to assume that solute pile-up ahead of the columnar front is effectively removed by convection, and in addition the columnar tips are moving, such that growth at this stage should be rather heavily influenced by the present hydrodynamics. Therefore, it is questionable to what extent growth models developed for more conventional solidification problems could be extrapolated to account properly for the situations that apply to rapid solidification during early to intermediate stages in weld processing. The experimental findings presented here serve to underline the role of melt flow both on crystal growth and on crystal motion, and may serve to stress that these effects should be taken more thoroughly into account in future simulation models for weld solidification. The reasons for not incorporating crystal motion into the current weld solidification simulation schemes may be a complete lack of relevant experimental benchmark data. Owing to the early stages at which tilting take place, little evidence from post-experimental characterization could be anticipated.

5. Summary

In situ time-resolved studies of evolving microstructures in steel alloy weld solidification were carried out using ultrafast polychromatic X-ray diffraction. Data analysis has brought quantitative insight to individual and overall crystal growth rates in solidifying welds. The study has shown that substantial crystal rotation, tilting and motion are present, especially in the earlier stages of solidification, and are likely to have a strong influence on the transient behaviour of growing crystals in welds. Furthermore, the experimental method and analysis developed within this study could be quite readily adapted to other in situ studies of welding or other rapid phase transformations in materials science.

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