Ultra-fast in-situ X-ray studies of evolving columnar dendrites in solidifying steel weld pools.

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Abstract. High-brilliance polychromatic synchrotron radiation has been used to conduct in-situ studies of the solidification microstructure evolution during simulated welding. The welding simulations were realized by rapidly fusing ~ 5 mm spot in Fe-Cr-Ni steel. During the solid-liquid-solid phase transformations, a section of the weld pool was placed in an incident 50-150 keV polychromatic synchrotron X-ray beam, in a near-horizontal position at a very low inclination angle. Multiple high-resolution 2D detectors with very high frame rates were utilized to capture time resolved X-ray diffraction data from suitably oriented solid dendrites evolving in the weld pool. Comprehensive analysis of the diffraction data revealed individual and overall dendritic growth characteristics and relevant melt and solid flow dynamics during weld pool solidification, which was completed within 1.5 s. Columnar dendrite tip velocities were estimated from the experimental data and during early stages of solidification were exceeded 4 mm/s. The most remarkable observation revealed through the time-resolved reciprocal space observations are correlated to significant tilting of columnar type dendrites at their root during solidification, presumably caused by convective currents in the weld pool. When the columnar dendrite tilting are transformed to respective metric linear tilting velocities at the dendrite tip; tilting velocities are found to be in the same order of magnitude as the columnar tip growth velocities, suggesting a highly transient nature of growth conditions.

1. Introduction
In a fusion welding process, the ability to weld a metal to itself or to another strongly depends on dendritic growth dynamics, which determine the morphological evolution of the advancing solidification interfaces and the spatiotemporal pattern building at the advancing front. Thus microstructure at the weld center, where the final growth fronts meet and solidification terminates, is dictated by the full history of the crystal growth process. Weld center boundaries form critical morphological features and are the regions where weld failures often occur. In most instances weld fusion zone (FZ) is deemed as a mini casting and solidification behavior of the FZ is derived from the extrapolation of the scientific knowledge gained from the investigations of ordinary solidification processes, which usually take place at orders of magnitude lower cooling rates [1,2] compared to the real welds.

With the advancement of the computational material science, many solidification mechanisms were hypothesized via computer simulations, e.g. ref. [3,4] establishing self-consistent theories. Synchrotron X-ray or neutrons studies [5-8] emerged as a powerful tool during last two decades to investigate
microstructure evolution in solidifying metallic alloys in-situ. X-ray and neutron experimental parameters are generally limited by the mutual compromise between the material properties, beam brilliance, detector capability and the required time resolution. Although a few experimental attempts were focused on welding related phase transformations eg. Ref. [8-10] employing X-ray diffraction (XRD) techniques, however a critical allusion of the solidification features in the weld pool, especially related to the crystal growth were not reported until recently [11]. In this contribution, we provide further analysis of columnar growth and related dynamic effects found in ultra-fast synchrotron XRD experiments [11].

2. Experimental configuration
The experiments were carried out at the European Synchrotron Radiation Facility’s ID15A beamline. Fe-Cr-17.3%-Ni-11.1%-Mo-2.1%-C<0.1% austenitic steel was used as the sample material and 2 mm thick and 30 mm x 20 mm rectangular samples were used for the experiments. The primary heat source employed to simulate a real spot-welding process comprised seven 1 kW Halogen lamps equipped with elliptical reflectors. The lamps were mounted in a custom-built rig that allowed for individual adjustments to superpose infrared and near infrared energy emitted from all the lamps into a common focal spot of approx. 5 mm diameter at the center of the sample Argon flow was brought in at the sample surface just adjacent to the weld spot to reduce oxidation rates at the pool surface and weld pool depths in the experiments were varied between 0.6 – 1.5 mm. Generally melting initiated after a few seconds from switching on the Halogen-lamps to full power, followed by stabilization of a molten pool within 1-3 seconds. As soon as complete melting in the part of the pool was verified through the illuminated incident X-ray beam, pool freezing was initiated by powering off the halogen heaters and the power-off time is referred hereafter as t = 0 s. Typically, the observed region of the weld pool re-solidified completely within 1.0 - 1.5 s. For each welding experiment, a sample was placed on top of a resistive heater with the surface normal tilted 0.035 rad towards the incident beam, as outlined in Fig.1. The incident X-rays were 50 - 150 keV polychromatic. The incident beam was positioned concentric to the pool surface center, 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. Diffraction data were collected with a Pco.dimax® CMOS equipped with an image intensifier, The CMOS spans a 2016 x 2016 pixel array, and was read at 1.0 kHz frame. Ref.[11] provides further details on this experimental configuration.

![Fig 1. Schematic configuration of time resolved X-ray diffraction experiments.](image_url)

3. Experimental observations and data analysis
During the experimental observations, a remarkable motion of individual discontinued XRD peaks in the early stage of solidification was appeared. At the beginning, these discontinued XRD peaks moved in very fast radially and each peak only appeared for few milliseconds only. After a while, lasting XRD peaks were appeared with reducing angular movement rates, the average angular motion rate for
one experiment is shown in Figure 2.a. It was ascertained that the combination of the polychromatic beam and the ultra-high frame rate is essential to realize a traceable evolution of XRD peaks, especially at < 400 ms. Subsequent to the discontinued XRD peaks, continued XRD peaks began to appear with, still substantially high moving rates, but their moving rates drastically reduced to a minimum during next 500 ms. A considerable grain re-orientation is attributed via such XRD peak movements and if a monochromatic X-ray beam or a fewer time resolutions was employed, only a mist like diffraction pattern could be detected e.g. ref.[10]. Initial discontinued XRD peaks were attributed to equiaxed type free moving grains in the melt pool and, subsequently appeared continued XRD peaks were attributed for columnar grains [11].

Figure 2. a) Average angular motion rate for XRD peaks binned for every 100ms, error bars indicate the reported minimum and maximum during the period. b) Schematic illustration of XRD observations for corresponding columnar crystal tilting, and growth. The tilting angle is exaggerated here for better illustration.

In order to analyze the data related to the columnar type grains quantitatively, time evolving integrated diffracted intensity, $I_h$, reciprocal lattice vector, $r^*$ length along the major axis (angular direction), $d_a$ and azimuth angle, $\psi$ for each XRD peak was extracted from the background corrected image sequence(s). While $\psi$ was observed to remain nearly at a constant value, prominent variations in $I_h$, $r^*$ and $d_a$ were recognized, in relation to the growth and orientation of a crystal. The observed XRD peak and potentially attributed crystal characteristics are schematically illustrated in Figure 2.b. As mentioned in the previous section above, XRD data were collected frame by frame at a fixed rate of 1.0 kHz and in order to analyze each frame, raw pixel intensities are required to undergo few processing steps for necessary quantification. Background intensity caused by diffuse scattering from air or the liquid metal was assessed from frames captured with the weld pool completely molten, and found to be weak and fairly constant across the solid angle covered by the detector. Accordingly, basic corrections were performed simply by subtracting the mean background intensity value from all pixels to arrive at a set of net pixel intensities for each frame. After background intensity subtraction, individual XRD peaks emerged as islands of interconnected pixels with net intensities above the pre-defined threshold in each frame. For complete mathematical details of the image processing procedure readers are referred to ref.[11].

According to the theory of kinematical diffraction $I_h$ from a crystal fully bathed in the incident beam, and for a XRD peak associated with the reciprocal lattice vector $h= h_a^* + k_b^* + l_c^*$, where $h$, $k$, $l$ are the Miller indices, and $a^*$, $b^*$, $c^*$ reciprocal unit cell vectors, can be expressed as,

$$I_h = V_c^{-2} K_h |F_h|^2 \cdot V_g$$

(1)

where $V_c$ 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,

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

(2)

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. Discussion

As the lasting (continued) XRD peaks observed in the experiments were attributed to columnar crystals [11]. Angular motion of such a XRD peak can be characterized as re-orientation via tilting of the crystal, as illustrated in Figure 2.b. Angular motion of XRD peaks could also occurred due to the thermal contraction, but observed \(dr*/dt\) were ~3-4 orders of magnitude above the resulting \(dr*/dt\) from the lattice parameter changes [12] at ~ 300 K/s cooling rate. In fact, thermal contraction effects are distinguishable in the latter part of the XRD image sequence(s). By successful indexing of a few reflection pairs belonging to the same columnar crystals with respect to \(<100>\) growth direction [2,10] allowed a number of the columnar crystal re-orientations and growth to be analyzed in more detail with \(I_h(t)\) of the respective XRD peaks. The time evolution of a set of individual columnar type grain volume fraction, \(f_g(t)\) evolutions were extracted and one example grain volume fraction curve is presented in Figure 3.a. Most of the analyzed columnar crystals that grow with primary tip directions at angles < 15º with respect to the incident beam. The average columnar grain length measured from the corresponding micrographs was taken as an input to the calculations, in the relevant experiments. The derived \(V_g(t)\) evolution for columnar crystals were computationally fitted to an evolving parabolic shape envelope. The initial growth rates found to be up to 4 mm/s initially, but were dropped by two orders of magnitude or more within 200 ms. In addition to the increase in intensity, the expansion of the XRD peak spot size, mainly along the radial direction (measured as \(d_s\)) was also observed later. However, the observation was identified as an effect of the crystal volume expansion, as suggested via a strong correlation between \(d_s\) and \(V_g\) rather than an evidence of a deformation (bending along the crystal trunk), through the similarity between the XRD peaks \(I_h\) increase (which is represented by the crystal volume fraction) and the relevant XRD peak width change presented in Figures 3.a and 3.b.
Experimental evidence on columnar crystal tilting from the root as explained here is novel in the case of welding or similar rapid solidification processing, and is to our knowledge, not incorporated in any simulation models, although its presence should be well-recognized. In the processes such as what we have studied, where high cooling rates employed, advection and convection is extremely strong and we suggest the flow driven solid-liquid momentum as the driving force for the tilting of solidifying columnar crystals. Very low yield strength of the alloy near melting point together with the relatively slender crystal roots could also be critically contributed for such as crystal tilting. As established via computational studies [1,13], even the absence of the arc pressure or Lorentz forces; surface tension driven forces can impose a reasonably strong flow in the weld pool, as similar to the conditions exists in the experiment(s). The maximum fluid flow value calculated for the experimental conditions via an established analytical relationship [1] is over 400 mm/s. The presence of a strong convection was evident in the post-welded samples which typically showed pitting in the center of the weld together with a continuous protruded ring of solidified material outside its edges.

Figure 4. (a and b) Evolution of growth and tilting for two representative columnar crystals. Average growth velocities are binned for every 5 ms and both the crystals grow with their major component parallel to the incident X-ray beam direction.

The reason for not incorporating such crystal dynamics presented here to the current solidification science may only be justified by a complete lack of relevant experimental data. Owing to the high temperatures tilting take place, no evidence from the post characterization could be anticipated. At the preliminary spell of the solidification, the columnar dendrite growth rates and the bending velocities are found to be nearly at the same order of magnitude as illustrated in Figure 4. As a implication of simultaneous growth and rotating/tilting and prevailing flow, the solute boundary layer around the growing crystals would be nearly wiped out, and may leave the anisotropy and diffusion control effects [1,3,4,14] nontrivial for microstructure formation in the solidification conditions presented here and could expect generally in fusion welding or any other rapid solidification process.

Our experimental evidence emphasize sever inadequacy to affiliate conventional crystal growth theories on weld or similar rapid solidification and also serves to underline the role of liquid flow as a controlling mechanism for the crystal growth, at least in the early to intermediate stages of such rapid solidification. Thus, the experimental results serves to stress that melt flow and liquid to solid momentum transfer is an obligatory aspect to address and account in future theoretical work and simulation models for weld solidification or any other similar solidification conditions which involve strong flow, rather than relying on direct extrapolation of conditions and exchange terms used for ordinary transient solidification processes.

5. Summary
Synchrotron radiation have been used to carry out in-situ time resolved studies of evolving microstructures in steel alloy weld solidification, with ultra-fast 2D X-ray diffraction pattern capture
rate. Experimental results with subsequent data analysis brings a quantitative insight to the highly
dynamic physical phenomena existing in weld solidification processes or any other similar dendritic
crystal growth taking place under very high melt flow environments. The study has shown that
substantial crystal rotation, tilting and motion is present, especially in the earlier stages of
solidification, which is likely to have a strong influence on the transient behavior of growing crystals
in welds. The experimental findings presented here will influence and emphasize on the critical factors
to be considered in the progression of the solidification and subsequent defect formation mechanisms
theories related to the weld process. Furthermore, the experiment and the results set an example for
future extremely fast in-situ XRD.

References
[1] DebRoy T and David SA 1995 Rev. Modern Physics 67 85
[2] David S A, Babu S S and Vitek J M 2003 JOM 55 14
[3] Jeong J H, Goldenfeld N and Dantzig J A 2001 Phys. Rev. E 64 041602
[4] Granasy L et al. 2004 Nature Mater. 2 645
[5] Mathiesen R H et al. 1999 Phys Rev Lett. 83 5062
[6] Buffet A et al. 2007 Physica Status Solidi: Applications and Materials Science 204 2721
[7] Yasuda H et al. 2011 ISIJ International 51 402
[8] Elmer J W, Wong J and Ressler T 2000 Scripta Mater. 43, 751
[9] Babu S S et al. 2002 Acta Mater. 50 4763
[10] Yonamura M et al. 2010 J. App. Physics 107 013523
[11] Mirihanage W U et al. 2014 Acta Mater. 68 159
[12] Seki I and Nagata K 2005 ISIJ International 45, 1789
[13] Tanaka M and Lowke J J 2006 J. Phys. D: Appl. Phys. 40, R1
[14] Tomorr H et al. 2006 Nature Mater. 5 660