An image processing pipeline for in situ dynamic x-ray imaging of directional solidification of metal alloys in thin cells

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Received: 20 December 2022 / Revised: 12 June 2023 / Accepted: 17 June 2023 / Published online: 16 July 2023
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Abstract
We present an image processing algorithm developed for quantitative analysis of directional solidification of metal alloys in thin cells using x-ray imaging. Our methodology allows to identify the fluid volume, fluid channels and cavities, and to separate them from the solidified structures. It also allows morphological analysis within the solid fraction, including automatic decomposition into dominant grains by orientation and connectivity. In addition, the interplay between solidification and convection can be studied by characterizing convection plumes in the fluid, and solute concentrations above the developing solidification front. The image filters used enable the developed code (open-source) to work reliably even for single images with low signal-to-noise ratio, low contrast-to-noise ratio, and low image resolution. This is demonstrated by applying the code to several dynamic in situ x-ray imaging experiments with a solidifying gallium–indium alloy in a thin cell. Grain (and global) dendrite orientation statistics, convective plume parameterization, etc. can be obtained from the code output. The limitations of the presented approach are also explained.

1 Introduction

Solidification is a central aspect of many industrial applications, particularly in metallurgy, e.g., production of nickel-based superalloys, lightweight aluminum and magnesium alloys, etc. (Amoorezaei et al. 2012; Kao et al. 2020; Stefanescu and Ruxanda 2004). A well-known and common problem is the risk of defect formation during these processes. Segregation of solute species originates at the microscale, but propagates to and emerges at the macroscale (macrosegregation), leading to nonuniformity of the distribution of the inter-metallic phases in industrial alloys (Beckermann 2002). In addition, during solidification of alloys, partitioning of elements leads to the formation of a solute boundary layer in the vicinity of the liquid–solid interface. In cases where the density of the solute may be lighter than that of the bulk liquid, buoyancy forces in the boundary layer directed back toward the bulk liquid cause the formation of solute plumes that emanate from the solid–liquid interface. Under certain conditions, the escaping solute can form stable channels called chimneys. After complete solidification, these disturbances remain as defects in the castings known as freckles, which are essentially anisotropic alloy composition inhomogeneities in the form of channels with diameters of few primary dendrite arm spacings and lengths varying from millimeters to centimeters (Reed et al. 2009; Auburtin et al. 2000; Beckermann et al. 2000; Kao et al. 2020; Saad et al. 2015; Shevchenko et al. 2013) (https://hdl.handle.net/2027.42/76001).

It is therefore desirable to control solidification such that defects do not occur. However, control requires understanding the underlying physics, particularly for the mushy zone (which is essentially the solidification front), and solidification processes in liquid metal alloys are very complex, with many possible regimes of pattern formation depending on system parameters (e.g., temperature gradient, cooling rate, alloy component mass fractions) (Amoorezaei et al. 2012; Asta et al. 2009; Haxhimali et al. 2006; Strickland et al. 2020; Stefanescu and Ruxanda 2004). Within the mushy zone, there exists an interplay of many physical mechanisms on different length scales: primary...
and secondary dendrite arm growth, liquid–solid interface instabilities, liquid mass flow near the interface and in the bulk (in general, both natural and forced convection), concentration transport, liquid flow in through solidified dendrite structures and remelting, global and local temperature dynamics, etc. (Asta et al. 2009; Ramirez and Beckermann 2003; Shevchenko et al. 2015, 2017; Stefanescu and Ruxanda 2004; Anderson and Guba 2020; Soar et al. 2020). One way to control such complex dynamics is by applying magnetic field to the domain where solidification occurs—however, one then has to consider additional physical mechanisms within the system, such as liquid flow damping or forcing by the Lorentz force, which also includes a thermoelectric contribution. This and other factors introduced by magnetic field application significantly alters solidified microstructures (Kao et al. 2020; Cai et al. 2020; He et al. 2020; Eckert et al. 2013).

A very prospective and commonly used method for studying solidification dynamics without or with applied magnetic field is by using downscaled model systems—Hele-Shaw cells where binary alloy solidification can be observed at the meso-scale (i.e., dendrite grains with spatially resolved individual dendrites) using in situ dynamic x-ray transmission contrast radiography. Even though one obtains only the projections of the solidified microstructures, it has proven to be a very effective means of probing systems with solidification processes for physical insights (Reinhart et al. 2020; Saad et al. 2015; Kao et al. 2020; Becker et al. 2020; Delalieu et al. 2010; Werner et al. 2021; Karagadde et al. 2014; Kao et al. 2019; Mirihanage et al. 2014; Shevchenko et al. 2013, 2015, 2017; Soltani et al. 2022; Tang et al. 2021; He et al. 2020; Boden et al. 2008). In addition to the challenges associated with imaging, there is also the matter of extracting valuable information from the acquired images. Ideally, to get the full picture of system dynamics, one has to separate the liquid from the solid, identify the solidification front and any liquid enclosures within the solidified microstructure, and obtain the microstructure skeletons. One could then perform orientation analysis for the skeletons, derive the primary dendrite spacing statistics, determine the local velocity with which the solidification front travels, as well as measure the solute concentration near the solidification front as it moves, since the concentration largely determines the front evolution. In this regard, detecting convective plumes and analyzing their shapes are also of interest, as is velocimetry in the liquid flow regions. In addition, it could also be of interest to detect and separate different grains (if any) within the microstructure seen in the images. Of course, the problem lies in doing all of the above automatically and reliably, which is relevant given the amount of images usually acquired in X-ray radiography experiments and the amount of information captured within each image.

However, while there exist solutions for some of the above problems, most appear to be limited to segmentation/detection of dendritic structures (Nenchev et al. 2020; Tassenberg et al. 2020; Wan et al. 2021; Hughes et al. 2021; Viardin et al. 2022; Wang et al. 2021). In Nenchev et al. (2020); Tassenberg et al. (2020); Wan et al. (2021), the focus is the detection of the dendrite cores from images of planes normal to the solidification direction—this, and the fact the algorithm presented in Nenchev et al. (2020), Tassenberg et al. (2020) uses template matching as one of its stages, makes it hardly applicable to studying x-ray images with directionally solidifying dendrite “forests” (e.g., as in Kao et al. (2020), Saad et al. (2015); Shevchenko et al. (2013, 2015); Boden et al. (2008)) where planes parallel to the growth direction are imaged. In addition, at least observing the demonstrated application examples, it seems that these methods should be reliably applicable in the cases when the images are fully filled with dendrites, i.e., in instances where there are both liquid and solid regions, one must first be separated from the other using different methods. Dendrite tip tracking is performed in Hughes et al. (2021) by segmenting the upper part of the solidified structures growing upwards. The key aspect of the segmentation procedure is to use the difference between two consecutive frames to highlight the newly formed solid, segment the relevant region, and then derive the tip coordinates. The utilized approach also enables tracking the solidification front. However, in cases where the differences between the frames are smaller and significant noise is present, the algorithm could be expected to run into performance issues and a more general approach is desired. A versatile approach using neural networks for automated detection of equiaxed dendrite detection was reported in Viardin et al. (2022). Another important example of automated dendrite segmentation using neural networks was shown in Wang et al. (2021) where, unlike in Viardin et al. (2022), a binary mask for the solid structure was predicted instead of detecting separate dendrites. It is, however, worth pointing out that the example images/cases presented in Viardin et al. (2022), Wang et al. (2021) do not exhibit significant noise, which is often present even after some temporal averaging in dynamic x-ray radiography experiments where exposure times are relatively low—it is therefore not clear how well these methods will perform under such conditions.

The lack of a systematic approach to image processing beyond methods for segmentation presents a problem, since it has been clearly demonstrated that the microstructure evolution must be analyzed in conjunction with the other processes in solidifying systems. Currently, the most common tool used for image analysis in the field is ImageJ with its many custom plugins developed by the community (Schindelin et al. 2012; Schneider et al. 2012). While ImageJ is open-source, with an impressive arsenal of methods, many of them are not automated, robust, or publically available. In contrast, it would be very convenient to have an open-source all-in-one solution for x-ray image analysis. It should also be noted that such code could be applied to
the output of numerical simulations as well, the difference being that the latter do not have the image noise associated with experimental measurements. Thus, more direct comparisons between simulations and experiments, which seem to be largely lacking, could be possible.

This was the motivation for us to present our solution—the first version of the open-source code developed for automatic analysis of dynamic x-ray radiography images of directional solidification processes studied using Hele-Shaw cells. The current version does not yet have an integrated optical flow component (e.g., like the code used in Boden et al. (2008)), but otherwise it meets the above-mentioned analysis functionality requirements. Moreover, it was designed for robustness and is quite resilient to image noise and low image contrast. Utilized approaches to image and data processing combine both well-known state-of-the-art and our original methods, particularly for solid structure segmentation and dendrite grain analysis. The performance of the methodology implemented in the code is demonstrated on data from in situ x-ray radiography experiments (Shevchenko et al. 2013, 2015).

2 Image characterization

2.1 Image acquisition

All images used in this paper were acquired at the X-ray lab at Helmholtz-Zentrum Dresden-Rossendorf (HZDR). The Hele-Shaw solidification cell with dimensions $35 \text{ mm} \times 25 \text{ mm} \times 0.15 \text{ mm}$ was imaged at 1 frame per second with a 1 s exposure time. The imaging system utilized the Phoenix X-ray XS225D-OEM X-ray tube and is described in more detail in He et al. (2020), Saad et al. (2015), Kao et al. (2020), Shevchenko et al. (2013, 2015). For each image sequence recording, dark current signals and x-ray beam profile signals were recorded for subsequent image correction and normalization during pre-processing. For every set of system parameters, repeated recordings were made to ensure the results are reproducible, as well as for redundancy.

2.2 Image properties

Images are 16-bit gray-scale TIFFs and the field of view (FOV) typically has a $\sim 760 \times 576 \text{ px (pixel)}$ image size with a pixel size $[13.7;37.6] \mu\text{m}$ (actual image size varies between different image sequences due to boundary cropping). Figure 1 is an example of an acquired image of a dendritic network in the solidifying Ga-In alloy (Shevchenko et al. 2013, 2015).

To simplify further description and analysis, it is important to at least informally define key image features. In Fig. 1, one can see the solidification front (SF) outline with a black dashed curve. Here we define the SF as the envelope of the FOV region with the solidified structures. A more precise operational definition in the image processing context will be given in Sect. 3.3. The region (or multiple) containing solidified structures delimited by the solidification front is the solid zone (SZ). This zone may also include liquid cavities isolated from the bulk liquid.
and channels that are connected to the bulk liquid (above the SF), e.g., one such channel will later form from the largest of the closed liquid pools highlighted in Fig. 1. Thus, the liquid zone (LZ) is the difference between the FOV and the SZ, minus the cavities and channels. These definitions will be used throughout the rest of this article.

The images exhibit Poisson (multiplicative) noise, as well as salt-and-pepper noise due to momentarily overexposed or unresponsive ("dead") camera pixels. The x-ray beam flux over the FOV is nonuniform, with a fall-off near the edges of the acquired images. The contrast-to-noise ratio (CNR) is different for the LZ and the SZ. The convective plume CNR in the LZ is initially rather good, but typically degrades over time as the solid fill factor (SFF, the ratio of the SZ area to the FOV area) of the cell increases—this is because the solute is ejected above the SF, the LZ is saturated, and the contrast between the liquid alloy components diminishes. In addition, the CNR in the SZ may also vary over the image since there is solute flow across the solidified structures, potentially occluding them. The signal-to-noise ratio (SNR) is usually adequate for structures in the SZ, but it is rather low for the convective plumes in the LZ. In addition, some of the images may exhibit larger-scale artifacts – for instance, as shown in Fig. 1 with red dashed frames. In this case, the artifacts are the spots where the two parallel walls of the Hele-Shaw cells were fused together.

The following assumptions are made regarding the images and the physical system:

- When treating the SZ, the noise is considered white Gaussian. This is because after pre-processing (Supplementary Information) the image luminance does not vary too much at length scales much greater than the dendrite thickness and inter-dendrite spacing (Fig. 1) – this is in contrast with the LZ (compare the luminance distribution within the dark-blue dashed frame against that above the SF).
- Persistent larger-scale artifacts (i.e., not pixels with outlying luminance values) in the images, if present, are considered stationary over time.
- Dendrites in the SZ have linear or only slightly curved shapes.
- Dendrites may overlap in the imaging plane, and thus, their X-ray radiography projections may cross.

Given the above, the following considerations determine the methods of choice:

- Low image resolution means that care must be taken when attempting to remove noise from dendrites. This is because the pixel size is a significant fraction of the dendrite width. At the same time, the textures in the solid domain are rather fine (inter-dendrite spacings are roughly of the same order of magnitude as the dendrite width). The SNR is such that the dendrites are corrupted by noise enough that methods which are not texture-/morphology-aware cannot achieve satisfactory non-destructive denoising.
- Liquid flow across the dendrites acts as correlated noise when attempting to derive dendrite morphology. This further complicates the solid structure analysis – treating this issue jointly with the Gaussian noise stemming from under-exposure does not produce good enough results and a separate approach is needed.
- While the CNR at the SF is rather high in the example seen in Fig. 1, in other cases the SF is not as smooth and is less contrast. Note that segmenting liquid cavities and channels can be even more difficult. Therefore, dedicated filters are required to significantly increase the CNR of the liquid/solid phase boundary before SZ/LZ segmentation.
- The segmentation method for SZ/LZ separation must reliably work under potentially varying image quality: the ray produced by the X-ray tube may flicker and has nonuniform intensity; solid fraction increase and solute ejection into the bulk liquid strongly change the image luminance distribution both locally and globally; these effects should be modeled by the utilized segmentation method.

3 Image processing

3.1 Assumptions and considerations

The developed image processing code must enable in-depth analysis of both the LZ and SZ over time, as well as the dynamics associated with the SF evolution. Therefore, the objectives are as follows:

1. Segment the LZ and SZ.
2. Derive the SF.
3. Identify channels connected to the LZ, and also liquid cavities within the SZ.
4. Segment the convective plumes within the LZ for shape analysis.
5. Extract skeletons of the structures (in this case dendrites) within the SZ.
6. Perform orientation analysis for the structures identified within the LZ.
7. Decompose the SZ structures into subdomains (grains) by orientation and connectivity.
8. Measure the solute concentration near the SF.

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• Methods used for LZ denoising must be such that the shapes of convective plumes are not overly deformed or smeared out, but denoising here is much less constrained than in the SZ.

• Image quality varies greatly across different experiments and image sequences, both our own and those performed by other researchers – it is therefore worthwhile to develop a code that is resilient and can operate under adverse conditions potentially much worse than what is seen in Fig. 1.

• Such a code with many components and methods will inevitably have a rather large number of parameters – these should either be mostly fixed/general or should be quickly optimizable.

Image processing is organized in stages outlined in Algorithm 1. Throughout the paper, we will provide the default parameters for the various procedures involved.

**Algorithm 1: Overall structure of the image processing pipeline**

**Input:** Raw image sequence

1. Pre-process images (Supplementary Information)
2. Remove image artifacts (Supplementary Information)
3. Segment the SZ (Section 3.3.1 & Algorithm 2)
4. Identify channels connected to the LZ, and also liquid cavities within the SZ (Algorithm 3)
5. Derive the SF and segment the LZ (Algorithm 3)
6. Extract skeletons of the structures (in this case dendrites) within the SZ (Algorithms 4, 5 & 6)
7. Perform orientation analysis for the structures identified within the LZ (Algorithms 7 & 8)
8. Decompose the SZ structures into subdomains (grains) by orientation and connectivity (Algorithms 7 & 8)
9. Measure the solute concentration near the SF (Algorithm 9)
10. Segment the convective plumes within the LZ for shape analysis (Algorithm 10)

**Output:**
- SF shape, height map and growth rate over time
- Solute concentration dynamics near the SF
- Shape dynamics for convective plumes in the LZ
- Dendrite structure maps with highlighted features
- Dendrite orientation spectra for the SZ
- Dendrite orientation spectra and relative areas for grains identified within the SZ

3.2 Image pre-processing and artifact removal

Image pre-processing is performed in *ImageJ*—the details are outlined Supplementary Information. Then the images are normalized and saved, and passed to *Wolfram Mathematica* for further processing. Sometimes the images will still contain artifacts even after cropping and pre-processing (for example, as in Fig. 1). While the flat-field correction (FFC) during pre-processing ensures that such artifacts are no longer strong outliers, these image areas still significantly affect image luminance histograms and may interfere with image filtering (especially BM3D, significantly disrupting patch matching) and segmentation. We therefore use a procedure that identifies and inpaints these defects, i.e., makes them seamless with respect to the surrounding image textures – the procedure is explained in Supplementary Information.

3.3 Liquid/solid zone separation

3.3.1 Image filtering

Prior to segmentation, image filtering is performed to increase the CNR for the LZ/SZ boundaries, including liquid cavities (CNR tends to be especially low) and channels.

Here, the filters were applied such that they also eliminate dendrite structures while preserving larger-scale liquid zones and larger spaces between dendrites that are filled with liquid. It was decided to use block-matching 3D (BM3D) filtering (Dabov et al. 2006, 2007, Lebrun 2012, Makinen et al. 2020), since, unlike other tested solutions, it consistently
preserved the SZ shape well and also strongly increased the CNR of channels and cavities within the SZ.

We perform image filtering in two stages. First, BM3D is applied. An overview of the utilized BM3D version, its implementation and settings are provided in Supplementary Information. The second filtering stage is the non-local means (NM) filter (Coll and Morel 2005), which we use to mitigate any artifacts left over and/or produced by BM3D and further increase the CNR for the SF and cavities and channels within the SZ. The details regarding the NM filter are given in Supplementary Information. Once the filtering is done, one can proceed with the SZ segmentation.

### 3.3.2 Solid zone segmentation

Despite significant improvements to SNR and CNR due to filtering (Sect. 3.3.1), segmentation is still a challenge due to generally imperfect reference-based FFC, spatially varying SNR and CNR, X-ray beam fluctuations and strongly varying SFF and solute concentration in the LZ over time, as well as formation, growth/shrinking and disappearance of cavities and channels. The tests show that (at least in our cases) global segmentation methods, even advanced ones, are not able to detect the SZ stably and accurately over an entire image sequence that usually starts with no solid zone and possibly ends with $\text{SFF} \approx 1$. We have therefore opted for an empirical "physics-aware" model that computes an adaptive binarization threshold for the filtered images. The segmentation steps are summarized in Algorithm 2.

#### Algorithm 2: SZ segmentation

**Input:**
- Pre-processed image sequence (Supplementary Information)
- Filtered image sequence (Section 3.3.1)
- (Optional) Apply Gaussian/median filtering to the raw images
- Compute the mean inverse luminance (MIL) for the raw images
- (Optional) Filter the MIL time series
- Compute the MIL-based adaptive threshold time series (1)
- Segment the SZ from the filtered (i.e. post BM3D and NM) images using the adaptive threshold

**Output:** SZ masks for the entire image sequence

The initial sequence of raw images is used, because it is desirable to capture the beam fluctuations as well. Optionally, small-radius median/Gaussian filtering may be applied first to mitigate outliers in the images. Afterward, the mean inverse luminance (MIL) $\langle 1 - I(t) \rangle$ is computed for all (normalized) images in a sequence, where $t$ is the time/frame index. To avoid over-fitting the adaptive threshold to the MIL time series, it is (optionally) filtered using the Gaussian total variation (TV) filter (Rudin et al. 1992). The adaptive threshold $\tau(t)$ for images is computed from MIL as follows:

$$\tau(t) = 1 - C_1 \cdot f_1(t) \cdot f_2(t) \cdot g_1(X) \cdot g_2(X);\quad f_1(t) = L_{TV}(1 - I(t)),$$

$$f_2(t) = (g_1 \circ g_2)(C_2 + 1 - f_1(t)).$$

where $g_1(X) = X / \text{min}(X)$, $g_2(X) = X / \text{max}(X)$, $X(t)$ is time series, $C_1 > 0$, $C_2 \geq 0$, $p \in \mathbb{R}$ and $L_{TV}$ is the TV filtering applied (if necessary) to the MIL time series.

The adaptive part of the threshold consists of two contributions: $f_1(t)$ and $f_2(t)$, where $f_1(t)$ is tied to the SFF of the FOV – the greater the SFF, the greater the MIL is because the SZ attenuates the X-ray significantly more intensely than the LZ; $f_2(t)$ is the correction based on $f_1(t)$ that accounts for the fact that, as the SFF increases, the solute is pushed out of the solid zone and the LZ becomes significantly more saturated. Both $f_1(t)$ and $f_2(t)$ also capture the instantaneous global illumination changes that could result from X-ray beam flickering. Moreover, $f_2(t)$ plays a very important role in cases where flow in the LZ rapidly transitions between natural and forced convection regimes (e.g., under the influence of applied magnetic field), since these transitions are accompanied by significant changes in the LZ luminance. The SZ is segmented by binarizing the inverted filtered images with respective thresholds $\tau(t)$. More details about this segmentation approach and the utilized parameters are found in Supplementary Information.

#### 3.3.3 Liquid zone, solidification front, cavity and channel segmentation

Next, liquid cavities and channels are identified, the SF is derived and the LZ is segmented—the steps involved are outlined in Algorithm 3. Steps 1 generates the SZ mask does not contain the areas occupied by artifacts – this is the mask used for the dendrite structure analysis in Sect. 3.4. Then the liquid phase mask is obtained in Step 2. Image padding in Step 3 is done because the cavities can also have a boundary
at the bottom and the sides of the FOV. The top boundary is not padded because once the SF has passed the top of the FOV it is impossible to distinguish cavities at the FOV top from channels. This way, the boundary component removal leaves only the cavities in the processed masks.

Steps 4 to 9 fill and exclude the detected cavities from the liquid phase mask (Step 4) and, if necessary, one can also remove leftover artifacts (if any) from SZ segmentation (Step 5); the closing transform (Haralick et al. 1987) (Step 6) is performed using disk structuring elements to fill the asperities in the 0- and 1-valued level sets in the mask – the asperities with length scales below a user-defined disk element size are filled conformally to the nearby level set boundary shape; the filling transform (Gonzalez and Woods 2006) (Step 7) completes the channel filling wherever it was imperfect after closing; Steps 8 and 9 detect channels as the differences between the output of Step 7 and the masks after cavity filling while removing small-scale segments that physically do not correspond to channels, i.e., are either too small to be classified as such or simply are artifacts.

Algorithm 3: LZ, cavity and channel separation & SF derivation

**Input:** SZ masks for the entire image sequence (Algorithm 2)

*Get the SZ & liquid phase masks with artifact areas excluded*

1. SZ: multiply the SZ masks by the inverted artifact mask
2. Liquid phase: invert the result and multiply by the inverted artifact mask

*Segment cavities & channels*

3. Cavities: apply 1-px image padding (pixel value 0, all image boundaries except for the top) to the output of Step 2, remove border components, then revert the image padding
4. Subtract the cavity masks from the corresponding liquid phase masks (Step 2) and invert the resulting images
5. (Optional) Perform segment size thresholding for the resulting masks
6. Invert the masks, then apply the closing transform
7. Apply the filling transform and invert the result
8. From the output of Step 7, subtract the difference between it and its segment size-thresholded version
9. Channels: find the image difference between the output of Steps 4(5) & 8 and perform segment size thresholding for the result

*Derive the SF & segment the LZ*

10. Apply 1-px image padding (pixel value 0, all image boundaries except for the bottom) to the artifact mask, remove border components, then reverse the image padding
11. Apply the closing transform
12. Add the outputs of Steps 7 & 11
13. LZ: invert the output of Step 12
14. SF: perform edge detection for the output of Step 12

*SF correction & smoothing*

15. Apply small-radius Gaussian filtering to the SF masks
16. Normalize the images and apply Otsu thresholding
17. Perform morphological thinning

**Output:**
- SZ with artifacts excluded
- Segmented LZ with artifacts excluded
- Cavity masks
- Channel masks
- SF states
To segment the LZ, one adds the artifact segments outside the SZ to the masks with filled channels and cavities (Steps 10 to 13). The SF is obtained by applying the Canny edge detection (Canny 1986) to the LZ mask. The SF is then smoothed with a small-radius Gaussian filter to eliminate noise introduced by the preceding operations, after which Otsu binarization (Otsu 1979) is applied and the thinning transform (Gonzalez and Woods 2006) is performed so that the SF is exactly 1 px thick. The physics that can be derived from the sequence of the SF states over time are shown further in Sect. 4. Parameters for Algorithm 3 are shown in Supplementary Information.

### 3.4 Solid domain analysis

#### 3.4.1 Image partitioning & scan region identification

Once the SZ has been segmented, one can proceed with the analysis of the solidified structures within the SZ. Since the local textures generally strongly vary throughout the SZ, as does their CNR, it was decided to split the FOV into a number of partitions, determine which ones contain enough of the SZ for significant analysis (scan regions), and perform local filtering and feature extraction. This procedure is detailed in Algorithm 4.

**Algorithm 4: Image partitioning & scan region identification**

**Input:**
- Pre-processed images (Supplementary Information)
- SZ masks with artifact areas excluded (Algorithm 3)

1. Partition the pre-processed images into grids square patches with side lengths based on image dimensions (2)
2. Partition the SZ masks into corresponding square patches; memoize output
3. Assign all patches their position indices; memoize
4. Compute the SFF for the SZ mask patches and assign the values to the respective image patches
5. Designate the image patches with SFF > \( \varepsilon_{SFF} \) (\( \varepsilon_{SFF} > 0 \), user-defined) as scan regions

**Output:** Scan regions for further analysis (Algorithm 5)

The pre-processed images (Supplementary Information) are partitioned into a regular grid of square patches with side lengths given by

\[
L = s \cdot \min \text{dim}(z) \tag{2}
\]

where \( z \) is the input image and \( s \) is the scaling factor.

Some patches, especially for the images near the beginning of the sequence, are going to be mostly filled with liquid, and thus are not eligible for solid structure analysis. Identifying which regions to scan for solid structures saves a significant amount of computation time. To determine the patches where significant amounts of the solid phase are present, the SFF is computed for every patch using the SZ mask patches. The patches with the SFF greater than a user-defined threshold \( \varepsilon_{SFF} \) are designated as scan regions and are passed to Algorithm 5 for further analysis. The memoized image position indices for the patches will be used later for the FOV reconstruction. More details on Algorithm 4 are given in Supplementary Information.

#### 3.4.2 Scan region filtering & dendrite skeleton extraction

The identified scan regions are processed as indicated in Algorithm 5.
Algorithm 5: Scan region filtering & dendrite skeleton extraction

**Input:** scan region images (Algorithm 4)

**Prepare filter input**
1. Re-scale the images and perform color tone mapping (CTM)
2. Re-scale the images again and perform FFC

**Filter the images**
3. Apply BM3D filtering
4. Perform two iterations of non-local means masking (NMM)
5. Apply soft color tone map masking (SCTMM)
6. Apply FFC

**Extract dendrite skeletons**
7. Segment dendrites using 2-threshold hysteresis binarization
8. Invert the mask
9. Apply the thinning transform
10. Perform size thresholding
11. Multiply the resulting masks with their corresponding SZ mask patches (Algorithm 4, Step 2)
12. Perform morphological pruning (optionally in multiple passes)
13. Remove border pixels
14. Perform size thresholding

**Output:** Dendrite skeleton masks

Here, the strategy is to use BM3D to denoise the dendrite textures as non-destructively as possible. However, we have found that in general it can be difficult to obtain good results without preparing the images first. This is why the first stage is image normalization, color tone mapping (CTM) and FFC. The CTM(\(x, c\)) operation maps the colors (in this case the gray-scale values) of the input image \(x\) image using gamma compression with a global compression factor \(c\) (Tone Reproduction 2004) and thus compresses the dynamic range—this has the effect of dramatically increasing the CNR of the dendrites. Meanwhile, FFC uses a coarse polynomial fit of the image luminance map to perform background correction (flattening) without reference (Wolfram Research 2017). In some cases, this helps to reduce the large-wavelength correlated noise due to liquid metal flow across the dendrites.

Next, the scan regions are filtered. One first applies BM3D to restore the dendrite textures in images, then two iterations of non-local means masking (NMM) are applied to mitigate any leftover correlated noise and increase dendrite CNR. NMM could be viewed as a generalized, locally adaptive version of unsharp masking—it takes the input image \(x\) and transforms it into an output image \(y\) as follows:

\[
y = 2 \times x - w_{nm} \ast \text{NM}(x, r_l, r_p)
\]

(3)

where \(\text{NM}(x, r_l, r_p)\) is the NM filter, \(w_{nm}\) is the NM mask weight, and \(r_l\) and \(r_p\) are explained in Supplementary Information. We have previously applied NMM to particle detection in neutron radiography images of particle-laden liquid metal flow for a similar purpose, with good results (Birjukovs and Lappan 2022; Birjukovs et al. 2022). Afterward, the soft color tone map masking (SCTMM) is applied for further background reduction and CNR enhancement. SCTMM transforms an original normalized image \(x\) to output \(y\) in the following way:

\[
y = x \ast (x - (1 - \text{CTM}(x, c)))
\]

(4)

The motivation and principles behind SCTMM are explained in detail in Birjukovs et al. (2021) and the applications in neutron imaging of bubble and particle flow in liquid metal are demonstrated in Birjukovs et al. (2021), Birjukovs and Lappan (2022). Finally, another FFC iteration is performed.

Dendrite skeleton extraction is done in eight steps. One starts with double-Otsu hysteresis binarization (Gonzalez and Woods 2006), followed by mask inversion, morphological thinning and size thresholding. Then the resulting skeleton masks are multiplied with their respective SZ mask patches, which crops the skeleton parts that are within the liquid phase areas and thus cannot actually be dendrites. Afterward, the remaining skeleton asperities are removed with multi-pass morphological pruning (Gonzalez and Woods 2006), then border pixels are removed and size thresholding is done again.

The parameters for Algorithm 5 are specified in Supplementary Information.
3.4.3 Resolving unoriented & overlapping structures

After dendrite detection, the skeleton segment orientations ($\varphi$, with respect to the image $X$ axis) must be measured. However, it may be the case that the image filters do not properly resolve primary/secondary dendrites or cases with dendrite overlaps. In addition, some segments may not have clearly defined orientations, or could simply be leftover artifacts. To identify and correct such unresolved (in the sense of orientation) structures within all IWs, a procedure outlined in Algorithm 9 is performed for all IWs.

Algorithm 6: Resolve unoriented skeletons

**Input:** IWs with dendrite skeletons (Algorithm 5)

1. Identify unresolved skeletons
   - Compute orientation angles ($\varphi$, with respect to the image $X$ axis) and aspect ratio ($\chi$) for dendrite segments
   - Colorize resolved (oriented) segments ($\chi > \chi_c$, $\chi_c \geq 1$ is user-defined) by their $\varphi$
   - Separate resolved and unresolved segments into different masks

2. Resolve the skeletons (for masks with unresolved segments)
   - Detect skeleton corner points
   - Detect skeleton branch points
   - Threshold and filter corner and branch points from skeletons

3. Reassemble IW skeletons
   - Re-evaluate segment orientations for masks with (formerly) unresolved segments (Step 7)
   - Colorize resolved (oriented) segments ($\chi > \chi_c$) by their $\varphi$
   - Add the output of Step 9 to the previously set aside masks with initially resolved segments (Step 3)

**Output:** IWs with resolved dendrite skeletons colorized by their orientations

Algorithm 6 uses the aspect ratio $\chi$ as a criterion to determine if the skeleton segments have a resolved (i.e., well-defined) orientation. Since morphological thinning is one of the steps in Algorithm 5 and Algorithm 6 does not add new pixels to masks, most of the skeleton lines should have a 1 px thickness and, provided they are long enough, therefore also high $\chi$. Both very small segments and large skeletons, with unresolved overlapping branches, will have low $\chi$ and will be passed for further processing. Colorizing the segments by $\varphi$ will play a key role later during dendrite grain decomposition (Sect. 3.5), but in Step 3 of algorithm 6 it is used to separate the masks with initially resolved and unresolved segments. The choice of the color scale does not matter as long as it is normalized (see Sect. 3.5) and the palette is visually convenient for the user.

Once the masks with unresolved skeletons are separated, corner point detection is performed using the Harris-Stevens method (Harris and Stephens 1988; Sánchez et al. 2018). Then, morphological branch points are detected (Wolfram Research 2010). Afterward, the detected corner points are filtered by selecting corner pixel clusters above a defined size threshold, combined with the detected branch points, and then pixel clusters with exceeding a defined size threshold are removed from the resulting mask. Finally, the size-thresholded combined mask is subtracted from the input mask with unresolved skeletons. With this done, the resolved skeletons are now assigned colors based on $\varphi$ subject to the $\chi > \chi_c$ criterion, and the resulting masks are recombined with those containing initially resolved skeletons.

3.4.4 Assembling the global dendrite skeleton

At this point, all the remaining unresolved (white-colored) skeletons within IWs are considered unoriented dendrites and/or artifacts. These are excluded from any subsequent analysis. One can now reassemble the global (FOV) dendrite skeleton image by tiling the IWs according to their position indices from Step 3 of Algorithm 4. It is also now easy to generate maps with color-coded dendrite orientations with highlighted cavities and liquid/solid boundaries.

3.5 Dendrite grain decomposition

Before proceeding with decomposition of the resulting global dendrite skeleton into grains, a global orientation ($\varphi$) spectrum must be computed for the assembled skeleton. It is not only of physical interest, but will be used in Algorithm 7...
as well. While a global $\varphi$ spectrum is certainly relevant, it is often desirable to distinguish areas of "coherent" dendrite growth, i.e., dendrite grains with their areas and mean dendrite $\varphi$. The presented dendrite grain decomposition (DGD) method does this by considering $\varphi$ similarity and proximity of the dendrites detected within the FOV, and it does so by exploiting the color-space representation of $\varphi$ generated by Algorithm 6.

### 3.5.1 Detecting dominant dendrite grains

DGD is performed in three stages: a primary scan which detects dominant dendrite grains; a refined scan which checks if the larger grains should be subdivided further and if the smaller grains are eligible; a filtering step which resolves ambiguities and overlaps between the detected dendrite grains. The first step of the DGD procedure is outlined in Algorithm 7.

**Algorithm 7: Detecting dominant dendrite grains**

| Input: Assembled global dendrite skeleton (Section 3.4.4) |
|-----------------------------------------------------------|
| 1. Compute the global $\varphi$ spectrum from the global skeleton |
| 2. Filter the $\varphi$ spectrum & detect dominant peaks |
| 3. Find dendrite segments near the $\varphi$ peaks in the color-space |
| 4. Build grain masks that cover the dendrite segments |

**Output:** Separate masks for dominant dendrite grains

The $\varphi$ spectrum is computed by measuring dendrite segment $\varphi$ (this time without the $\chi > \chi_c$ constraint) and lengths, and then constructing a histogram with uniformly-sized bins with values $\rho \in [0:1]$ weighed by the dendrite lengths. Length weights are used to account for boundary pixel removal in IWs which may break up longer dendrites into fragments. More details are found in Supplementary Information.

Since there is a mapping between $\varphi \in (-\pi/2; \pi/2]$ and the dendrite skeleton color values (normalized) due to Algorithm 6, one can now find the segments in the global skeleton that correspond to the selected $\varphi$ peaks. The procedure is explained in Supplementary Information.

When the dendrite clusters corresponding to each $\varphi$ peak are found, a mask must be created for them that will delimit and separate them as one grain. This is done by applying the closing transform with disk structuring elements to the skeleton clusters, which fills the spaces between the dendrite skeletons while not affecting the outlying parts of the skeletons, i.e., preserving the shapes of the dendrites that protrude from the bulk of the cluster. Note that this may generate more than one grain mask per $\varphi$ peak, since dendrite clusters with very similar orientations may be sufficiently far apart. Thus, grains are identified, accounting for both dendrite orientations and spatial distribution. Afterward, the resulting grain-covering binary masks are thresholded by their area, and the remaining masks are separated for further analysis with a refined scan. The parameters for the above steps are also found in Supplementary Information.

#### 3.5.2 Refined dendrite grain scan

Once dominant grains are identified for each $\varphi$ peak, they are subjected to a secondary scan that is designed to check whether the originally recognized grains need to be further subdivided. This is done to both (implicitly) ensure grain uniqueness, minimize overlaps, and resolve smaller areas within the larger grains that have distinct enough orientations. The scan follows steps similar to those of Algorithm 7, but with the following modifications:

1. Steps 1 and 2 are now applied to the dendrites within the grain masks, not the global skeleton.

2. Area-adaptive $\varphi$ spectrum resolution is used.

Prior to $\varphi$ spectra calculation, the dendrite skeletons belonging to the grains are isolated by multiplying the grain masks by the global dendrite skeleton. The area-adaptive resolution is set up such that, on the one hand, the algorithm can resolve finer differences in orientations within the initial grains and detect the underlying $\varphi$ peaks, while on the other hand not using exceedingly large resolution for smaller grains with relatively few dendrites. In the latter case, the algorithm would otherwise treat the noise in the spectrum as significant peaks despite the filtering. Details are provided in Supplementary Information.

Note that in some cases the refined scan may eliminate a grain instead of simply keeping or subdividing it. The former can happen if the areas of the resulting resolved grains are below the threshold, although such cases should be quite rare.

#### 3.5.3 Resolving ambiguities & performing cleanup

When the initial grains have been scanned again and kept or decomposed further and/or eliminated, the final DGD step is performed—ambiguities and overlaps between the grains are resolved. Two cases must be treated here: the previous
stages of DGD have, in separate instances, generated two grains with almost identical (i.e., overlapping) masks and this is indeed one and the same grain; there is partial overlap between the grains, but it is physical since the grains are adjacent and the dendrite orientation changes very slowly from one grain to the other, i.e., there exists a transition zone instead of a sharp boundary. In the former case one of the masks is redundant, and in the latter the overlap zone must be identified and designated as such, since no clear distinction between the two grains can be made in the transition zone. Finally, there is also the matter of potentially leftover dendrites with \( \varphi \) values that are significant outliers with respect to the mean \( \varphi \) for the grains. These issues are addressed by Algorithm 8.

**Algorithm 8:** Resolving ambiguities & performing grain cleanup

**Input:** Dendrite grains output by the refined scan (Section 3.5.2)

1. Compute the overlap masks for dendrite grain pairs
2. Compute the uniqueness factors \( u \in [0; 1] \) for the grain pairs
3. Discard redundant grains (\( u < u_c, u_c \) is user-defined)
4. Designate the overlap masks for the pairs of remaining unique grains as overlap zones and subtract them from the grain masks
5. Perform total area thresholding for the resulting masks, then secondary size thresholding for the underlying segments – the output gives the final grain masks
6. Multiply the global skeleton by the final grain masks to isolate the respective dendrites
7. Remove dendrites with outlying \( \varphi \) values and perform dendrite size thresholding

**Output:** final dendrite grain masks & skeletons

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Fig. 2 FOV image processing: a raw image, b pre-processed image (Supplementary Information), c image with artifacts (note the image corners in a) inpainted as explained in Supplementary Information and d final image after BM3D and NM filtering (Sect. 3.3.1). The color scheme here and further, unless stated otherwise, is identical to Fig. 1. Note the enhanced contrast of the SF, liquid channels and cavities.

(c)
The first step is performed by multiplying all possible pairs of dendrite grain masks. Then the uniqueness factors defined as $u = 1 - \frac{S_\cap}{\langle S \rangle}$ are computed for all dendrite grain pairs, where $S_\cap$ is the overlap mask area and $\langle S \rangle$ is the mean grain area for the pair. Pairs with $u < u_c$ are considered redundant, and only one grain mask from such pairs is kept. The overlap masks for all the other grains are kept as overlap zones and subtracted from the unique masks. The remaining segments are then thresholded both by total and individual areas. Afterward, the resulting masks are multiplied by the local skeleton to isolate the grain dendrites. Finally, the dendrite skeletons are filtered by orientation and length: $\langle \theta \rangle$ is measured for the grain dendrites and segments outside the $\langle \theta \rangle \pm 5\sigma$ interval (by default) are eliminated, followed by length thresholding. This concludes the DGD process. Relevant parameters are found in Supplementary Information.

### 3.6 Liquid domain analysis

#### 3.6.1 Measuring solute concentration above the solidification front

Measuring the solute concentration above the SF involves the following considerations:

- Filtering the noise in the liquid region above the SF as non-destructively as possible, i.e., not to alter the luminance field too much, as it can later be used with the

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**Fig. 3** Boundaries between segmented SZ/LZ (black contours), accounting for areas with removed persistent artifacts, for different time stamps a–d in ascending order obtained by segmentation using Algorithm 2 and 1

**Fig. 4** Stages of segment classification for separated SZ/LZ: a segmented SZ, b mask a with non-border artifacts removed (case none present here), c liquid cavity mask, d LZ mask without cavities and border or persistent artifacts, e mask b but without border artifacts (none were present), f combined masks c and d, g masks separated by the SF, and h liquid channel mask. This case corresponds to the image sequence considered in Fig. 2
Beer-Lambert law to assess the concentration of the solute.

- LZ/SZ segmentation will never be perfect, and dendrite tips could be slightly above the SF, yielding errors in the luminance/concentration measurements—this must be mitigated.

Both are addressed by Algorithm 9.

**Algorithm 9: Measuring solute concentration above the solidification front**

**Input:**
- Pre-processed FOV images without artifacts (Supplementary Information)
- Solidification front masks (Algorithm 3)

1. Apply median filtering to the FOV images
2. Apply the bilateral filter
3. Perform NM filtering
4. Define the SF-conformal buffer zone by shifting the SF contour mask upwards
5. Define the concentration sampling zone by extruding the SF-shaped boundary upwards from the buffer zone upper boundary
6. Compute the mean luminance for vertical pixel bands within the sampling zone over the FOV width
7. (Optional) Use the Beer-Lambert law to convert the luminance to the solute concentration

**Output:** Mean luminance/concentration above the SF over the FOV width for all time stamps

We find that the combination of median, bilateral (Tomasi and Manduchi 1998) and NM (in this order) filters in Steps 1-3, after some parameter tuning, yields a sufficiently well-filtered luminance/concentration field without offsetting the values too much or significantly affecting the larger-wavelength features. The bilateral filter is chosen in particular because of its luminance value range filter component, since with the right settings it should preserve luminance level sets within the images that are fairly close. Once the images are processed, sampling zones are defined for every frame. To address the above-mentioned issue with dendrite tips possibly being above the SF, a buffer zone is created where no sampling occurs—this is done by shifting the SF mask (curve) upwards by a distance $d_{buf}$. Starting from this level, the SF curve is then extruded over a distance $d_{samp}$ to create the SF-conformal sampling zone for the concentration measurements from the filtered images. Here, the mean values are computed for 1 px-wide vertical strips of $d_{samp}$ pixels above the buffer zone over the width of the FOV. Parameters are provided in Supplementary Information.

### 3.6.2 Convective plume segmentation

With convective plume segmentation, the luminance value preservation is not as serious a concern as long as the shapes are preserved. Here the idea is to filter the images and then decompose the resulting filtered LZ luminance map into $N_{level}$ level sets (clusters) ranked by luminance. One can then re-assemble $N_{plume}$ level sets with the highest luminance back together to obtain the masks for the convective plumes above the SF. This is done via Algorithm 10.
Algorithm 10: Convective plume segmentation

Input:

- Pre-processed FOV images without artifacts (Supplementary Information)
- LZ masks with filled channels and cavities (Algorithm 3)

1. Perform Steps 1-3 from Algorithm 9 (with different parameters)
2. Gaussian filtering
3. NM filtering
4. Create a buffer zone above the LZ boundary (SF) in the LZ mask with filled cavities and channels using morphological dilation
5. Multiply the filtered image by the resulting mask
6. Decompose the resulting image into level sets
7. Re-assemble selected level sets to get convective plume masks

Output: Convective plume masks & level sets

To isolate the liquid region within the FOV, the image is multiplied by the LZ mask. However, before multiplication, a buffer zone is created above the LZ mask by morphological dilation using disk structuring elements with a radius of 15 px. The rationale here is the same as in the case with the concentration measurements above the SF – to eliminate the dendrite tips that may have been imperfectly segmented. If not removed, these would form "parasitic" level sets and reduce the actual level set count within the LZ.

The luminance (concentration) level sets for the convective plumes are obtained using the K-medoids method (Kaufman and Rousseeuw 1990) with $N_{\text{level}}$ medoids – the level sets are then ranked by their mean luminance (with the excluded solid and buffer region having the lowest, zero luminance after mask multiplication) and the top $N_{\text{plume}}$ are re-assembled into the output mask for the convective plumes.

Parameters for Algorithm 10 are given in Supplementary Information.

Fig. 5 Examples showing how the SF (red curves) is traced based on the final SZ mask

Fig. 6 IW processing via Algorithm 5: a a colorized relief plot of an IW (projection of a pre-processed image), b IW after re-scaling, CTM and FFC, c BM3D output, d results after 2 iterations of NMM, one SCTMM iteration and FFC, e output after 2-Otsu (hysteresis) binarization and image inversion, and f final result after size thresholding and thinning, overlaid on top of the grayscale version of d. Image luminance increases from color light blue to white

Fig. 7 An example of detected IW skeletons for an image after cropping to the SZ mask, e.g., Fig. 4c. This case corresponds to Figs. 2 and 4
4 Application examples & performance

To demonstrate the resilience of the proposed solution, it was decided to test the developed methodology and code for conditions that are purposefully made worse than one would expect. Specifically, we have opted to use only a single frame for FFC for pre-processing for an image sequence. This way the background features, i.e., setup elements caught within the FOV, artifacts, and the X-ray beam profile are still compensated for, but the resulting SNR is significantly lower.

4.1 Image filtering & segmentation

First, consider the stages of FOV processing prior to SZ and LZ segmentation. Figure 2 shows characteristic examples of input images versus the pre-processed and then filtered output. Figure 2 (more examples in Supplementary Information) is a good example where many of the image features that can realistically be expected are present.

Artifacts appear as overexposed corners and areas at the bottom image boundary in (a). A thermocouple is located in the upper-left corner of (a), visibly protruding from the left image boundary. Note also the tape for affixing the thermocouples, visible as rectangular areas with greater opacity at the left and upper boundaries of (a). Liquid channels and cavities are present in the lower part of the FOV, better visible in (b). One can observe that even with single frame FFC, the luminance distribution in (a) about the X-ray beam axis is almost entirely negated in (b), and so are the elements attached to the imaged liquid metal cell. However, as opposed to (a), noise is amplified in (b) and becomes visibly coarser-grained. Afterward, the artifacts are removed, and the result is shown in (c) – the formed artifact areas should no longer affect the NM and BM3D filter patch matching procedures. Finally, BM3D and NM are applied, in that order, resulting in an output seen in (d) with much more contrast LZ/SZ boundaries, as well as cleaner liquid metal cavities and channels. While the liquid metal plume in the upper part of (b) has been dramatically diffused, it is of no concern here, as the result of this image filtering routine is used only for LZ/SZ separation (filters in Algorithm 9 are used for concentration measurements in the LZ instead).
The results of segmentation (Algorithm 2, using (1)) after FOV filtering are presented in Fig. 3 (with more examples in Supplementary Information). Figure 3 represents an easier case where the front has a rather simple shape, with cavities forming and disappearing, and a channel forming over time to the left of the cell center line. An example of a time series for the dynamic segmentation threshold and the rationale behind the used parameters are given in Supplementary Information.

After SZ/LZ segmentation is complete, these segments are further differentiated to separate liquid cavities and channels from the SZ, and to determine the shape of the SF. There are also safeguards against artifacts that can potentially be left over after LZ/SZ segmentation, as it is hardly possible to always find optimal $C_1$, $C_2$ and $p$ immediately. Therefore, it is good if the code has backup options. Figure 4 illustrates the steps involved in separating segment classes (more in Supplementary Information).

Once cavities are identified (Algorithm 3), one readily obtains the bulk LZ segment, and can then derive the SF and find the channels extending below the SF. Examples of how the SF is traced along the SZ segment boundaries is shown in Fig. 5. The resulting SF edge masks will be used later for...
Fig. 13 Results of DGD for the case corresponding to Fig. 12: a dendrite segments colorized by dendrite grain IDs with grain mask overlays, with orange areas representing liquid metal cavities and bulk liquid with channels highlighted as gray areas, and b grain dendrite skeletons overlays. The background in b is a post-processed raw image (pre-processing followed by image inversion, reference-less FFC, CTM, re-scaling and sharpening) with the LZ masked.

Fig. 14 Results of DGD for the case considered in Fig. 10. Note the transition zone (black-colored dendrite segments) in a between the two grains to the left.

Fig. 15 Dendrite orientation $\phi$ spectra for the grains identified via the DGD process, as shown in Fig. 14. Colors for plots a–b correspond to the grain colors in Fig. 14. The legend in d shows the grain area fraction $S$ (with respect to the SZ mask area), mean dendrite orientation $\phi$ and its standard deviation $\delta\phi$. 

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concentration measurements above the SF and for convective plume segmentation in the LZ.

### 4.2 Analysis of solidified structures

With the SZ identified along with cavities and channels, one can now turn to the analysis of the solidified structures within the SZ. As outlined in Algorithm 4, first the FOV image is partitioned into IWs (an example is shown in Supplementary Information), and then each partition is processed using Algorithm 5. An example of this is provided in Fig. 6 (and another in Supplementary Material). Notice that the noise makes the identification of dendrites in the initial images (a) quite difficult, and the CNR for dendrites is quite low, mainly due to larger-wavelength correlated noise stemming from liquid flow. Note also that the dendrites have lower X-ray transparency than the surrounding liquid, and therefore it is the liquid that is colored white in (a).

Subfigure (b) shows that CTM and reference-less FFC make the dendrites and the spacings in between much clearer, but the SNR and CNR are unchanged. However, we found that this stage dramatically boosts the performance of BM3D, which is the next stage, the output of which is seen in (c). While the structures in Fig. 6c are already clearly discernible to the human eye, the CNR is still lower than desired for reliable segmentation. Since BM3D does not mitigate the correlated noise (uncorrelated Gaussian noise model is used for BM3D), the next stage with two iterations of NMM correction and SCTMM is applied (another example is shown in Supplementary Material), after which reference-less FFC is applied again. NMM takes care of much of the correlated noise, SCTMM boosts CNR, and FFC acts as post-NMM large-wavelength background cleanup.

Afterward, double-Otsu hysteresis segmentation and image inversion yields the dendrite mask (e), and morphological thinning with size thresholding generates the IW dendrite skeleton (f). Now all that remains is to crop the dendrite skeletons, multiplying their masks by the SZ mask projected onto IWs – this process is demonstrated in Supplementary Material. After cropping is done for every IW, one has solid structure skeletons for the entire FOV – an example of IW skeletons output by Algorithm 5 for the case considered in Figs. 2 and 4 is shown in Fig. 7.

Observe, however, that many of the IWs in Fig. 7 exhibit multiple dendrite crossings and branching, making the orientation analysis problematic. To mitigate this issue, Algorithm 6 is applied to each of the IWs. Extra Figures in Supplementary Information show the steps involved in this procedure. Although the results have certain imperfections and a small amount of information may be lost in IW skeletons in general, we find that the set of parameters that we have selected for Algorithm 6 allows recovering much more information that would otherwise be lost. This is very clearly illustrated in Figs. 8 and 9 where one can see the significant difference between before and after Algorithm 6 is applied.

As noticeable in Fig. 9, some of the new segments still do not exhibit clear orientations or are otherwise ambiguous, but most of the information otherwise inaccessible from Fig. 8 has been recovered with minimal losses. With this, one can now re-assemble the processed IWs into a global skeleton, which yields results showcased in Fig. 10, and more in Supplementary Material).
While Fig. 10 is already quite informative, there is more to be extracted from the assembled skeletons. One can measure the orientation ($\theta$) spectrum, computing the relative orientation frequency by weighing over dendrite segment lengths to account for gaps due to IW boundaries and dendrite interruptions due to other reasons. An example of this is shown in Fig. 11b where the $\theta$ spectrum is computed for a reconstructed dendrite skeleton. Note that the unoriented dendrite skeletons (e.g., colored black in Fig. 10b) do not count toward the spectrum. One can also compute the median $\theta$ for every IW to have a coarser but simplified overview of how dendrite $\theta$ are distributed over the FOV—this is seen in Fig. 11a. In addition, by computing $\theta$ spectra for an entire image sequence, one can observe the dynamics over time. Statistics for liquid cavity (orange areas in Fig. 10b) areas, aspect ratio, orientations, etc. can also be computed per frame and their dynamics visualized. The same is true for channels extending into the SZ (e.g., Fig. 10b).

Importantly, the $\theta$ spectrum shown in 11b is later used for the DGD procedure (Algorithm 7), which is used to decompose the global dendrite skeletons into grains. Examples of this process are shown in Fig. 12 and in Supplementary Material. Figure 12a is similar to Fig. 10a, but is stripped of all background with only dendrites remaining—here the most intense $\theta$ spectrum peak shown in

**Fig. 19**  Mean SF dynamics over cell width: a height $\langle Y \rangle$ and b vertical velocity $\langle dY/dt \rangle$ over time $t$ for the case shown in Fig. 3

**Fig. 20** Solute concentration dynamics above the SF and along the cell width $X$ over time $t$ for the case shown in Fig. 3. Concentration is expressed via image luminance (encoding GaIn concentration via the Beer-Lambert law) in relative units (with respect to the initial frame after Algorithm 9 is applied) and is color-coded as shown in the color bar to the right.

While Fig. 10 is already quite informative, there is more to be extracted from the assembled skeletons. One can measure the orientation ($\phi$) spectrum, computing the relative orientation frequency by weighing over dendrite segment lengths to account for gaps due to IW boundaries and dendrite interruptions due to other reasons. An example of this is shown in Fig. 11b where the $\phi$ spectrum is computed for a reconstructed dendrite skeleton. Note that the unoriented dendrite skeletons (e.g., colored black in Fig. 10b) do not count toward the spectrum. One can also compute the median $\phi$ for every IW to have a coarser but simplified overview of how dendrite $\phi$ are distributed over the FOV—this is seen in Fig. 11a. In addition, by computing $\phi$ spectra for an entire image sequence, one can observe the dynamics over time. Statistics for liquid cavity (orange areas in Fig. 10b) areas, aspect ratio, orientations, etc. can also be computed per frame and their dynamics visualized. The same is true for channels extending into the SZ (e.g., Fig. 10b).

Importantly, the $\phi$ spectrum shown in 11b is later used for the DGD procedure (Algorithm 7), which is used to decompose the global dendrite skeletons into grains. Examples of this process are shown in Fig. 12 and in Supplementary Material. Figure 12a is similar to Fig. 10a, but is stripped of all background with only dendrites remaining—here the most intense $\phi$ spectrum peak shown in

**Fig. 21** Analyzing Ga-rich convective plumes in the LZ above the SF (Algorithm 10): a pre-processed image with the outlines of the buffer zone (black contours) extended from the SF and the artifacts (e.g., lower-left and upper-right corners), and b convective plumes segmented and highlighted outside the buffer zone (black areas with white boundaries). The level sets representing the plumes are colorized by image luminance (representative of the solute concentration). The color scheme is as in Fig. 20. This case corresponds to the one shown in Fig. 3 (close to the beginning of the image sequence, where the plumes are the most intense).
Fig. 11b is considered. In (b), one can see the segments with \( \varphi \) within an interval of the selected \( \varphi \) peak (Algorithm 7) and (c) shows the result of thresholding by color-space distance. Afterward, the grain masks are constructed using the closing transform, then size-thresholded, which results in grain masks as seen in (d). These operations are performed for every peak in the global \( \varphi \) spectrum that survives filtering and thresholding (Algorithm 7).

When the refined scans and grain cleanup (Sect. 3.5.2 and Algorithm 8) are done, one can assemble the resulting grains (both their masks and dendrite segments) within the FOV, examples of which are shown in Figs. 13 and 14. It becomes visible in Fig. 13, and especially (b) that the detected grains indeed constitute the major dendrite clusters with coherent (sufficiently similar) \( \varphi \) sets. Note also that in (a) the grains separated by the liquid cavities (e.g., upper-left corner and the lower-right part of the FOV) are correctly separated, even the ones with sufficiently similar \( \varphi \). This is because liquid cavities are accounted for in the grain cleanup process. Observe that the detected dendrite skeletons match the landscape seen in the background of (b).

The same holds for Fig. 14, where in (a) one can see a black-colored cluster of skeletons designated as undetermined, i.e., they do not belong to either of the two adjacent grains with certainty and are a transition zone. This is because both grains are very close to this area, and the \( \varphi \) of dendrites within this zone is somewhere between the mean \( \varphi \) for the two grains in question. It might seem that the lower part of one of the grains in (a), highlighted with light blue, should be treated as a separate smaller grain because the long diagonal cavities seem to split it in two. However, one can clearly see in (b) that they are actually connected via one of the dendrites that bridges a narrow gap between the two liquid metal cavities. A similar situation holds for the grain highlighted with light red in Fig. 13a. If one does observe some small clusters of grains with similar \( \varphi \) in Figs. 13b and 14b that were discarded by the DGD process, then these grains are most likely below the size threshold. One can, of course, always adjust settings accordingly if capturing smaller grains is necessary.

Once the dominant grains are detected, their relative areas (with respect to the SZ area) and \( \varphi \) statistics can be determined, which is demonstrated in Fig. 15 for the case seen in Fig. 14. Again, note that this can be done for all or selected frames in an image sequence to observe the dynamics of grain formation, fragmentation and how their statistics change.

4.3 Solidification front dynamics

In addition to the above, having derived the SF, one can now look at its dynamics. Figure 16 shows how the SF height over the cell width changes over time for the case shown in Fig. 3, as the SF advances vertically upwards and the SFF increases.

This also contains the shape information to some degree. If a more continuous dynamics visualization is needed, one can generate an image as in Fig. 17 where the color encodes the front height over time and cell width can be clearly seen for an entire image sequence. Figure 17 is obtained by median-filtering the front height matrix with a 2-pixel (element) kernel radius. The resulting matrix can then be used to calculate the matrix of instantaneous SF propagation velocity, which is shown in Fig. 18. In the case of velocity, outlier removal and bilateral filtering (\( \mu_b = 2 \) pixel value range factor and Gaussian kernel scale \( \sigma_b = 21 \)) are performed. If mean dynamics are of interest, they can be readily derived from the above matrices. The corresponding results are shown in Fig. 19.

Solute concentration can be measured above the SF as explained in Algorithm 9 and plotted for different locations along the cell width over time, which is showcased in Fig. 20.

Note the maximum spot within \( X \in (8.2;12.3) \) mm and \( t \in (1505;1755) \) s, which corresponds to a rapid channel opening at the SF (Fig. 3). Figure 9 reveals the sudden
appearance of a highly concentrated "trace" in the same region as the velocity minimum/maximum in Fig. 18. Once a liquid metal cavity breaches the SF and a channel forms, much greater X-ray transmission is consistently measured, as it should be, implying increased Ga concentration. The trail shift to the left is simply due to the change in the location of the channel outlet at the SF, which is caused by remelting. Observe also the banded structure for \( t \lesssim 1500 \) – these are not artifacts due to data processing, but rather physical concentration oscillations above the SF, as well as the result of SF fluctuations.

### 4.4 Convective plume segmentation

Finally, one can analyze what occurs above the SF in the bulk of the LZ further by examining the convection plumes using Algorithm 10. The results of its applications to example frames representing different cases are shown in Figs. 21–22. As with the concentration measurements just above the SF, the buffer zone mask is intended to prevent the interference from potentially sticking out dendrite tips that in general may occur above the derived SF for low SNR/CNR images. In addition, the artifact areas must be excluded, since any information contained there is otherwise meaningless. This makes obvious that the code successfully segments the plumes in the showcased examples and largely preserves their shapes despite the clearly visible large-grained noise.

This multiple level set representation, in conjunction with morphological analysis shown in Fig. 4, as well as the SF dynamics and SZ analysis, should provide a wealth of details, enabling in-depth analysis and physical interpretation of solidification processes studies using experimental setups that are similar in principle to those considered in this paper. It is worth noting that what can be seen in the above sub-figures (a) a purposely lowered SNR. Given the results seen in (b), higher SNR images should enable even better results. This, of course, also holds for the SZ analysis.

### 5 Further improvements & extensions

While many of the necessary features are already present in the code, there is most certainly room for functionality expansion and performance boosting. Specifically, we propose the following:

- Transition to GPU implementations of BM3D and NM filters, such as the ones presented in Honzátko and Kruliš (2019); Davy and Ehret (2021) – this should greatly reduce computational time per image sequence.
- Test a possibly better suited BM3D version, BM3D-SAPCA (shape-adaptive principal component analysis) (Dabov et al. 2009).
- Integrate an optical flow velocimetry code by Liu et al. Liu (2017); Liu et al. (2021) directly into the presented image processing code via MATLink for a more in-depth liquid flow analysis and seamless start-to-finish data processing. This will be invaluable, in particular when combined with the presented plume segmentation and the SF analysis routines.
- Improve upon the IW partitioning approach to eliminate the gaps between the skeletons from separate IWs – this would require a robust method that combines the overlapping parts of the IWs. Successfully implementing this will further boost the quality of dendrite structure analysis.
- Derive solid phase thickness over the cell and Ga/In concentrations.
- Compute inter-dendrite primary spacing.
- Demonstrate that the showcased methodology readily translates to higher-resolution images, e.g., from synchrotron measurements and numerical simulation output.

### 6 Conclusions

To summarize, we have demonstrated a robust and noise-resilient image processing pipeline for analyzing directional metal alloy solidification processes in laboratory-scale experiments using Hele-Shaw liquid metal cells and dynamic X-ray imaging. The developed methodology at present allows one to segment liquid and solidified zones within the field of view, detect the skeletons of solidified structures in the solid zone, perform orientation analysis, detect dominant dendrite grains (if any), quantify the dynamics of the solidification front and solute concentration above it, detect and separate liquid metal channels and cavities, as well as segment and characterize convective plumes in the liquid zone. Even with artificially lowered SNR, the code performed reliably and the demonstrated performance is such that in-depth physical analysis of images is feasible. With the addition of extra features in the future, such as optical flow velocimetry for the liquid zone and convective plumes, as well as other improvements outlined above, the presented methods and code should be of great value to the relevant scientific community for further physics-focused research.

**Supplementary Information** The online version contains supplementary material available at https://doi.org/10.1007/s00348-023-03671-2.

**Author contributions** MB developed and implemented the image processing code, and processed the data used in the paper. NS provided...
the data (x-ray images) and helped test the code together with MB, SE provided funding. The first version of the manuscript was written by MB. All co-authors (MB, NS and SE) contributed to manuscript editing and review prior to submission.

Funding  This research is supported by Helmholtz-Zentrum Dresden-Rossendorf (HZDR) and a DAAD Short-Term Grant (2021, 57552336). The authors acknowledge the project “Development of numerical modeling approaches to study complex multiphysical interactions in electromagnetic liquid metal technologies” (No. 1.1.1.1/18/A/108) wherein some of the utilized image processing methods were developed.

Data availability  Both input and output for the image processing code, as well as associated visuals, are available on demand—please contact the corresponding authors.

Code availability  The code is open-source, and is available on GitHub: Mihails-Birjukovs/Meso-scale_Solidification_Analysis.

Declarations

Conflict of interest  None to declare.

Ethical approval  Not applicable.

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