Title:

Measurement of Anode Anisotropy by Micro X-ray Computed Tomography

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Abstract:

Micro X-ray Computed Tomography (CT) is an instrumental method for recording inner structure 3D images without damaging the observed volume. A methodology has been developed by SINTEF for CT of carbonaceous materials, yielding detailed structural views and quantification of the pore, void and grain distribution within volumes of 10 mm to 130 mm diameter.

Example results are shown from a study of anode anisotropy. A series of vertical cores from a prebaked anode was analysed, selected due to clear difference in top, middle and bottom level structure. The CT data was calibrated and connected to the physical position in the anode. The data was analysed with various methods that quantify the degree of anisotropy. The analysis show that the anodes have a 10-15 cm wide zone of high anisotropy, while the top and bottom of the anode are more isotropic. Some possible reasons for mid-height anisotropy are discussed.

Keywords:

µCT, Anode Cracking, Anisotropy, Image Analysis

Paper:

1. Background

In the recent years, Micro X-Ray Computed Tomography (µCT) has emerged as a good alternative to microscopy and visible light image analysis, especially for studies of carbon anodes. Carbon is comparatively transparent to x-rays, so there are less artefacts during reconstruction of the data, and the signal-noise ratio is excellent. As a collaboration between SINTEF and Hydro Aluminium, successful imaging and interpretation have been made of porosity, shape of grains, anode structure, distribution of grains, distribution of the anode binder matrix, associated mixing quality, and evaluation of packing faults. Investigations have also been made of risk patterns connected to cracking and, from spent anode samples, the degree of electrolyte penetration into the anode wear surface. The main advantage of µCT is that the technique can be used to image large samples non-destructively at a good resolution within a reasonable time and cost. Since the features of interest are inside the material, a method with depth penetration like µCT is an excellent way of investigating internal structure. Qualitative features of anodes made both in pilot scale and anodes produced at different industrial plants can be compared and experiences in production can be seen directly in the resulting product. Several advantages and possibilities of high resolution µCT were discussed in a previous TMS paper [1].

Most anodes are formed by unidirectional vibro-forming, and it is the experience of the authors that the aggregate packing pattern in the green anode shows considerable vertical versus horizontal anisotropy. The main cause for this is the gravity-driven packing combined with the elongated shape of the coke grains. The degree of anisotropic packing versus isotropic packing has been studied using µCT images from the recorded image stacks and even illustrating 3D movies.

This paper summarizes the findings from a µCT investigation of vertical anisotropy difference through an anode based on cores sampled at several heights. The anode combines both good structure and a clear cracking pattern at several heights. This suggests good green paste and problematic vibro-forming. However, the paper focusses on the technique and the imaging results, which is a good example of the versatility of the method, and do not discuss the causes apart from a brief mention in the Results and Discussions section.

2. Data Acquisition, Processing and Export

The µCT data was acquired by a Nikon XT H225 ST instrument (cone beam volume CT). It has a 225kV maximum acceleration voltage and employs a reflection source selectable between four different targets: Cu, Mo, Ag and W. The usable sample size range is from 5 to 150 mm diameter. The detector has 2000x2000 pixels, so the resulting resolution (voxel size) for the usable sample size range is 2.5 to 75 µm, depending on the sample size.

For the current work, a tungsten reflection target was used, with an acceleration voltage of 135 kV and a current of 200 µA. The radiation was not filtered. The imaging was done with an integration time of 1 second, amplification of 18 dB, with 6283 projections per 360º. The distance from source to sample was 221.72 mm, distance from source to detector was 1127.5 mm, resulting in a voxel size of 39.33 µm. The images were exported as 16-bit TIFF and processed in the public domain software ImageJ [2] using custom macros for this project.

For maximum resolution, the sample must fill the scan volume width. Since the cores are longer (~25 cm) than wide (~7 cm), the best results are obtained by doing several scans along the sample length, and then merging the data as a post processing step; the instrument software cannot do this automatically. Because the X-rays are reflected from the target, the angle and thus the energy is slightly different between the top and the bottom of the scan. Since higher energy X-rays penetrate the sample more easily, the calculated attenuation values are slightly (~2%) lower at the top compared to the bottom of each scan. This error was calculated and corrected from a scan of a calibration piece of pure isostatic graphite of the same size. 5 overlapping scans were done for each sample.

3. Analysed Samples

The drill positions of the cores are shown in **Table 1** and illustrated in **Figure 1** with three core positions "TOP", "MIDDLE" and "BOTTOM". The mid-height core will be influenced by the extra pressures from the stub-hole nearby, and the bottom core will be influenced by the extra pressure from the slots nearby.

The drill position of each core was saved in a metadata-file with the same root name as the µCT data files. The image processing macros use this information for calculating the physical position (coordinate within the anode) of each voxel, for easier documentation and tracking during reporting.

***Table 1**: Position of drilled cores*

Label	Position	X [mm]	Y [mm]	Z [mm]
V8T	TOP	685	530	600
V9T	MIDDLE	920	530	500
V8B	BOTTOM	685	530	0

When describing the images from the µCT recording, a slice is a cross-section and a stack is a sequence of slices. A stack can be presented sequentially as a 3D movie and such movies are exported in both the X, Y, and Z directions.

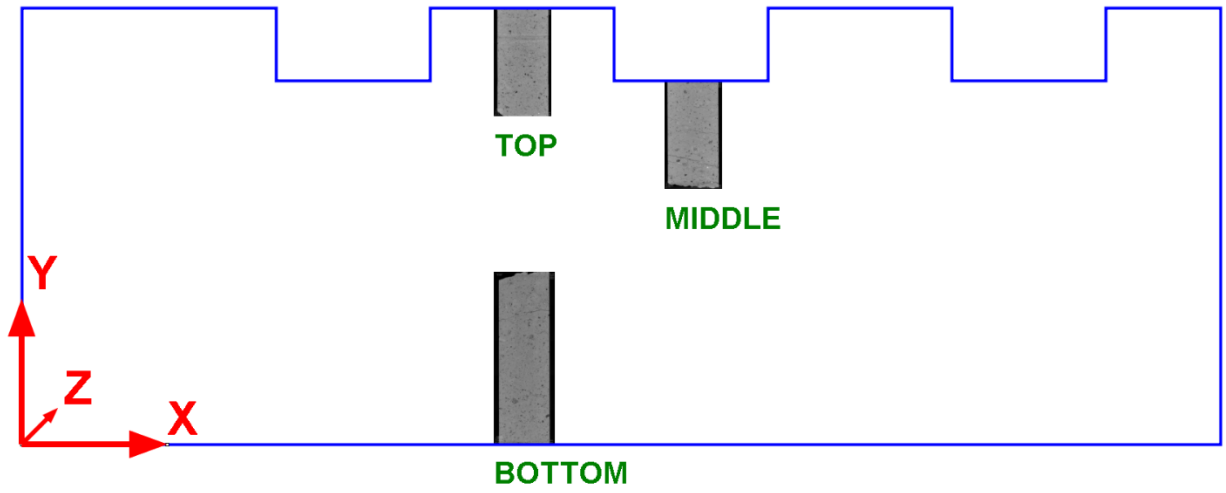

***Figure 1:** Position of drilled core samples.*

The definition of the orientation of the coordinate system can hopefully be established as an agreed upon convention which is very valuable in all types of 3D mapping of anode properties. By this convention, the Z axis is the direction of the current (and vibro-forming) while the XY plane is thus in the transverse direction; parallel to the anode wear surface (and potroom floor). The X-axis is along the longest edge of the anode. A core, or any position, is thus defined by (X, Y, Z), where the X-position is the distance from the anode short edge, the Y-position is the distance from the anode long edge and the Z-position is the distance from the anode operational surface; the wear surface. As a convention, origo is at the corner, away from the centre channel and down to the right when looking towards the centre channel. The coordinate system is thus right-handed.

The given core X-, Y-, Z-positions are saved in a metadata file connected to the µCT datasets. The methodology now reads this metadata and connects it to the coordinate system of the µCT data automatically, enabling easier labelling of the images.

Figure 2 shows a vertical cross-section of the merged dataset of the core labelled BOTTOM, which was drilled from the anode wear surface. **Figure 3** shows a magnified view (size 50x40mm) of Figure 2 at Z-position (height) 175mm, while **Figure 4** and **Figure 5** show magnified views at 105mm and 35mm respectively. **Figure 5** shows good, even quality anode structure indicating good mixing with good wetting of the grains and binder matrix. There are no mixing flaws, and both coke and butts were apparently well wetted.

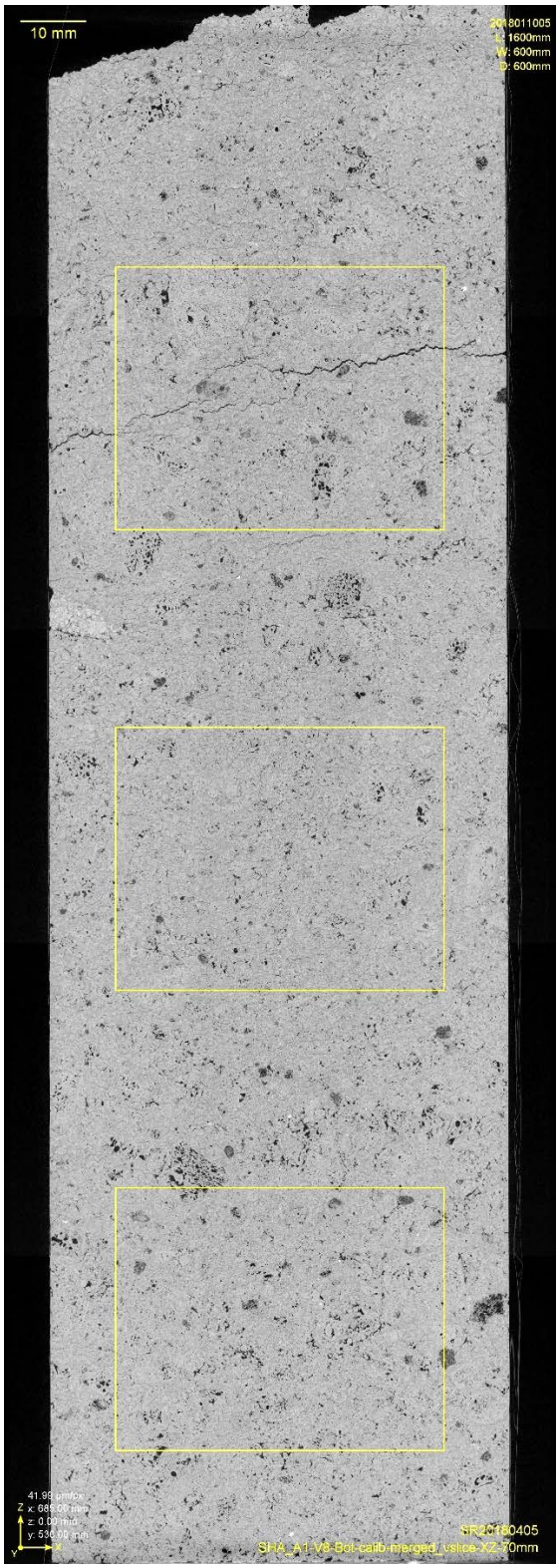

Figure 2: *Vertical slice of core BOTTOM drilled from bottom*

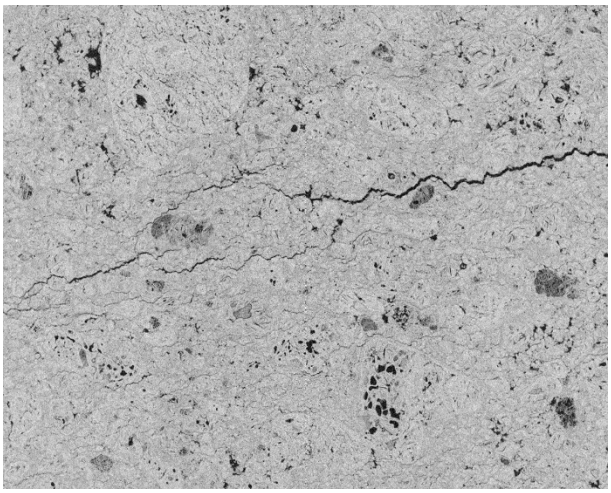

Figure 3: *Magnified (50x40mm) Figure 2 at Z-pos. 175mm*

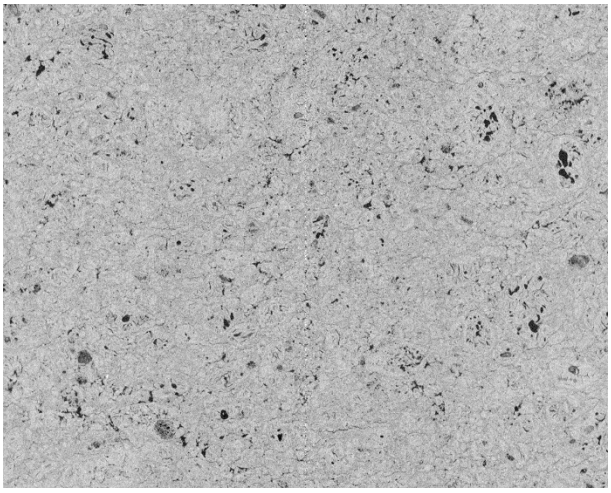

Figure 4: *Magnified (50x40mm) Figure 2 at Z-pos. 105mm*

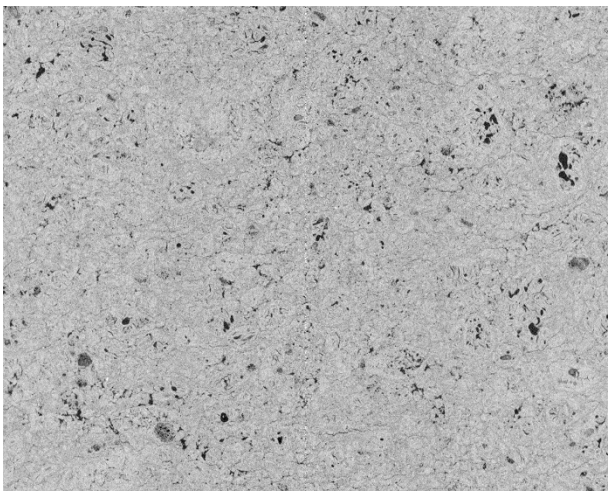

Figure 5: *Magnified (50x40mm) Figure 2 at Z-pos. 35mm*

Moving upward in the core, to **Figure 4** and especially **Figure 3** there are hairline cracks at increasing height (distance) from the anode wear surface (Z-position). The crack in Figure 3 is about 250 µm at the

widest. The crack is not continuous, it shows branching and spread around grains and there are parallel thin cracks creating a zone of cracking.

An important part of the post-processing of the data is calibrating the attenuation values. The grey tone in the images is linear proportional to the X-ray attenuation value. The calibration is done against a graphite reference, which means the grey tone can be compared between all images. **Figure 5** shows moderate variation in grey tone, as does **Figure 4**, however, **Figure 3** shows greater span with more dark grey areas, suggesting higher porosity, and possibly an effect similar to the cracks.

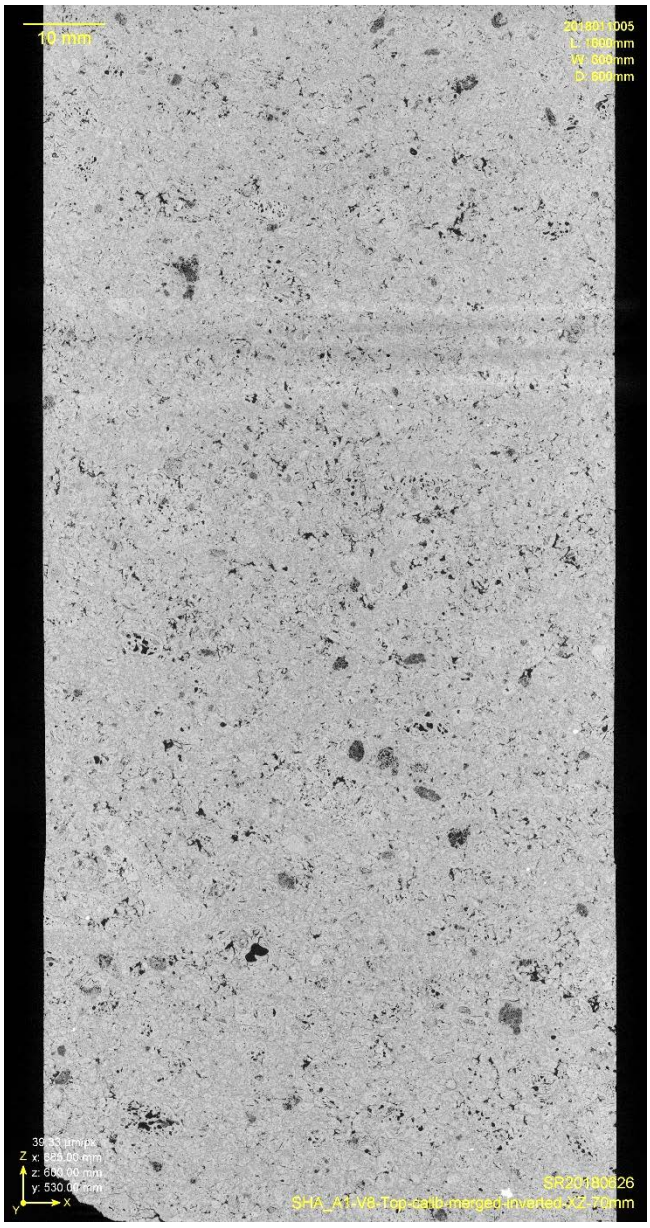

Figure 6: *Vertical slice of core TOP*

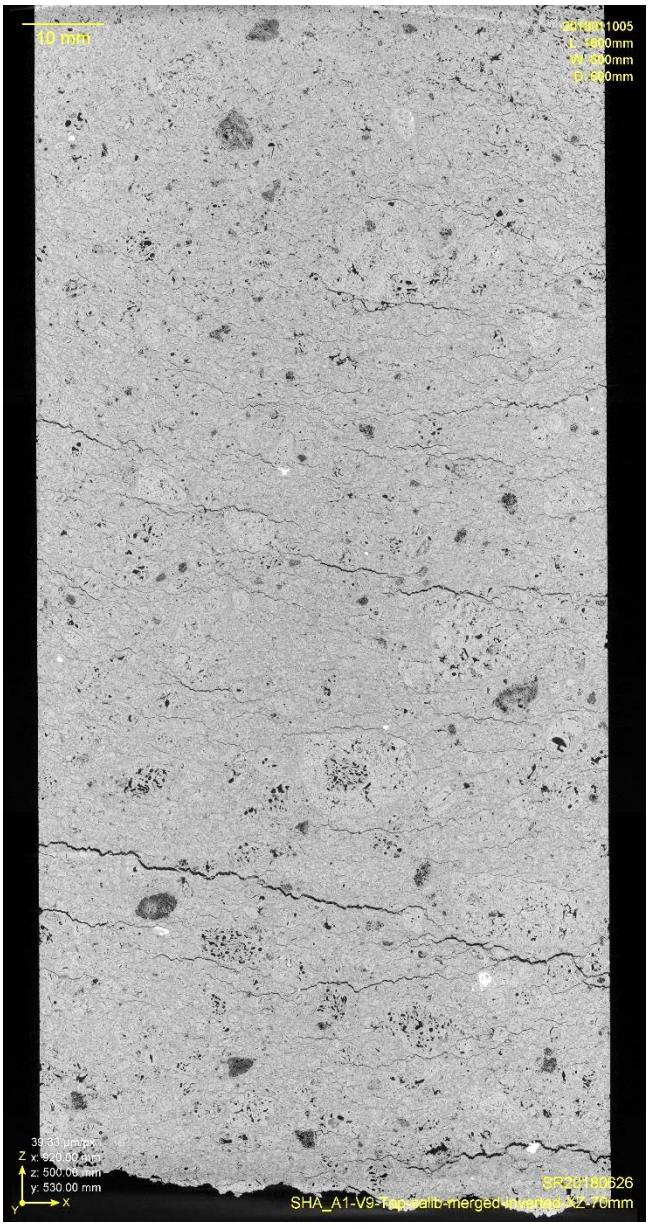

Figure 7: *Vertical slice of core MIDDLE*

Figure 6 shows a vertical slice of core TOP drilled from top of anode, and **Figure 7** shows a vertical slice of core MIDDLE drilled from bottom of the stub-hole. MIDDLE has considerably more visible cracks.

4. Image Analysis Method

For analysis of the anisotropy of the µCT data, a plugin named OrientationJ was used. The mathematics is described in [3]. The software characterizes the orientation and isotropy properties of a region of interest (ROI) in an image, based on the evaluation of the gradient structure tensor in a local neighbourhood. Three parameters are calculated: Orientation, Energy and Coherency. Orientation is simply the angle of the structure tensor. The Energy value indicates the amplitude of the tensor. The Coherency indicates if the local image features are oriented or not: Coherency is 1.0 when the local structure has one dominant orientation and is 0.0 if the image is essentially isotropic in the local neighbourhood. So Coherency is therefore a direct measure of the anisotropy of a structure.

The local neighbourhood window size was chosen as 4x4 pixels (168 x 168 µm) for the method development. This size was found to best be able to detect the relevant cracks. A problem with this method is that since it only considers a small local neighbourhood, it cannot separate anisotropy due to "real" cracks and local anisotropy due to coke pores, which are also cracks, but on a smaller scale. The latter will however be randomly distributed angle-wise, so if the calculated values are averaged over an area in the image of say 10x10mm, only "real" cracks should contribute to the anisotropy.

Figure 8 through **Figure 13** shows example results from the appliance of the method for a small image.
Figure 8 shows a 60x40mm crop from vertical slice of core BOTTOM as the original density image.
Figure 9 shows a vector plot of the Energy and Coherency properties multiplied.
Figure 10 shows the Energy values as an image, while **Figure 11** shows the Coherency values as an image. **Figure 12** shows the Orientation angles as an image, while **Figure 13** shows a histogram plot of these angles from -90 to 90 degrees. The colour scale of **Figure 12** is thus illustrated.

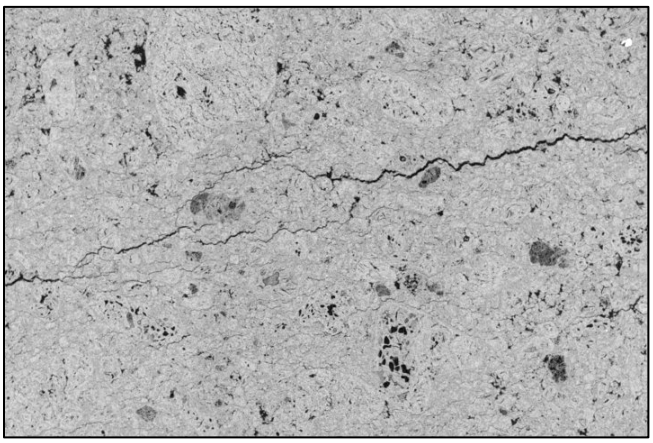

Figure 8: *60x40mm crop from vertical slice of core BOTTOM*

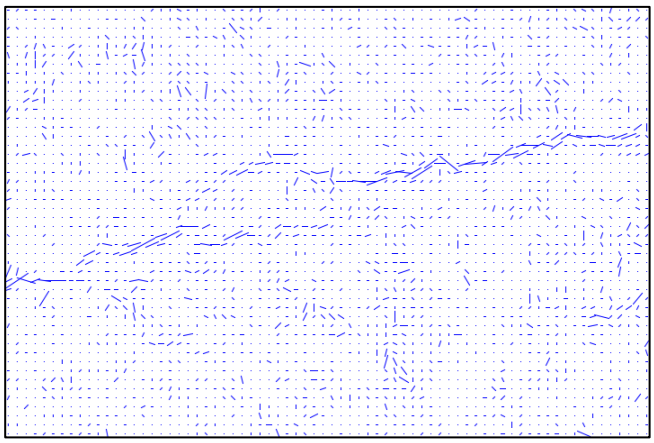

Figure 9: *Energy multiplied by Coherency as a vector plot*

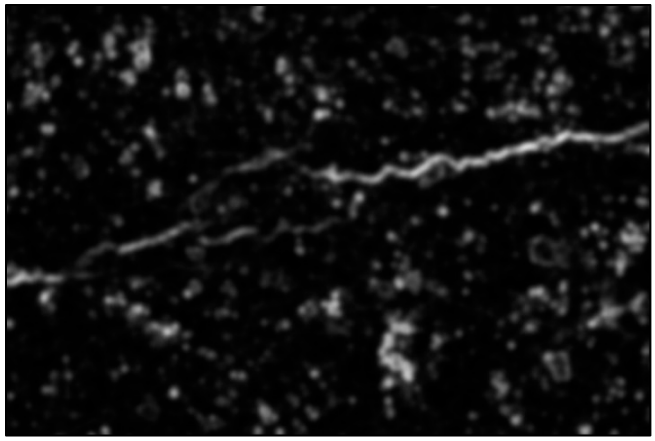

Figure 10: *Energy image*

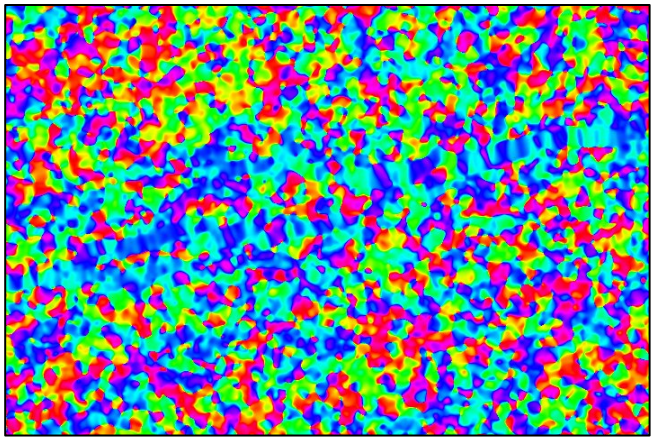

Figure 12: *Orientation image*

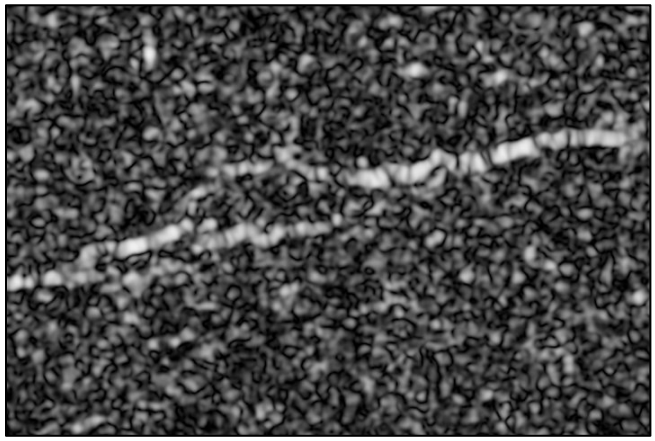

Figure 11: *Coherency image*

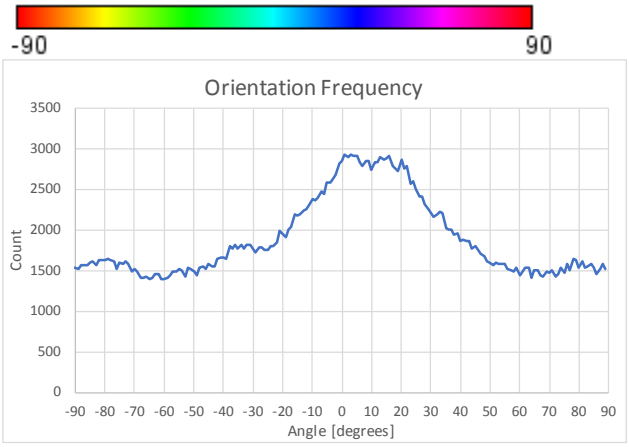

Figure 13: *Orientation Frequency from angle -90 to 90 degrees*

The energy value was not used in the analysis, since it was found to give high values along the edges of the coke pores and therefore does not say anything about the anode isotropy. Compare the coke grain a bit low right of the middle in Figure 8, Figure 10 and Figure 11. The coke grain has high Energy but low Coherency.

5. Results and Discussion

As a means of quantifying the degree of local anisotropy as a function of Z-position (distance from the anode bottom), average measurements were calculated for a slice height of 10mm. A window size of 8x8 pixels (336 x 336 µm) was used for the local gradient calculation, or better separation of the cracks from the coke pores. **Figure 14** shows a plot of the average Coherence value for these slices for all three cores. Z position 0 is at the anode bottom, while Z position 600 is at the anode top. An increasing amount of anisotropy is observed at increasing distances from both the bottom and the top, and the difference is nearly symmetrical. Since no sample was available for the middle of the anode, the plot data is incomplete.

The µCT is an excellent way of illustrating inner structure at a new level of detailing. However, good interpretation is still a challenge. Several hypotheses can be set forth on causes for the different patterns.

It is not the task of this paper to discuss this. Some possible explanations of the uneven distribution of anisotropy can be suggested:

1) The distribution of the anode paste in the form before vibro-forming starts can be uneven, causing pressure gradients and strain through the green anode body during the forming.
2) The vibro-forming is unable to compress the middle of the anode with the same intensity as the top and bottom parts, which are exposed to more vibration pressure due to the nearness to the steel top and bottom.
3) As an alternative effect of 2), if the vibro-forming is very intensive, the movement of the paste in the middle of the anode can be greater, creating a greater challenge to the cohesion of the binder matrix.
4) The packing of the paste can yield a zone where the anode is too dense, so pitch volatiles do not escape easily during baking. This can cause build-up of pressure in the middle where the volatile pressure is higher.

As seen, multiple hypotheses can be formulated. What is clear, however, is that the risk of anode cracking can be associated with the anisotropy of the anode as measured with the described method. The anisotropy is a property of the anode structure, and cannot be explained by random variations.

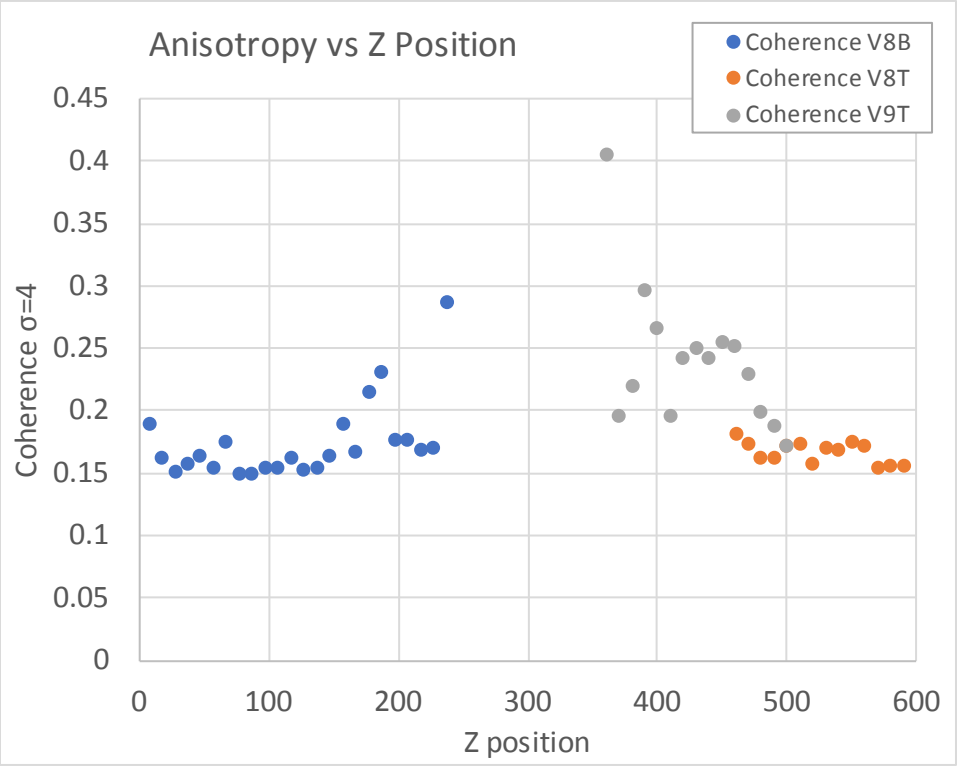

Figure 14: *Coherency versus Z-position for 60x10mm crop of all samples*

6. Acknowledgement

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7. References

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2. Rasband, W.S.: ImageJ, U. S. National Institutes of Health, Bethesda, Maryland, USA, www: http://imagej.nih.gov/ij/, 1997-2018
3. OrientationJ, a plug-in for ImageJ. Written by Daniel Sage at the Biomedical Image Group (BIG), EPFL, Switzerland, available on internet at http://bigwww.epfl.ch/demo/orientationj/