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________________________________________________________________________

Dorthe Wildenschild

Systems that contain multi-phase flow in porous media are of interest in diverse fields including environmental engineering, hydrogeology, and petroleum engineering etc. One of the main descriptors of multi-phase flow in porous media is the relationship between capillary pressure and fluid saturation. Capillary pressure is inherently a pore-scale variable and is generally measured externally by a pressure transducer. As an alternative, it has recently become possible to estimate capillary pressure from images by using curvature values using the Young-Laplace equation. X-ray microtomography is the general platform to perform three dimensional imaging, and for producing measurements such as interfacial curvatures. To improve such curvature measurement, different image processing scenarios were tested and a novel curvature estimation method was developed. Results revealed that errors on the curvature estimates have been significantly reduced for both synthetic data and real data sets. Because curvature-based capillary pressure for disconnected non-wetting blobs differs from transducer-based capillary pressure due to
residual oil blobs experience less hysteresis as well as the influence of local pore morphology, connected fluid and disconnected fluid were considered separately.
Linking Pore-scale Fluid-Fluid Interfacial Curvature in Porous Media to Capillary Pressure and Local Morphology with A Novel Method for Curvature Measurement

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Tianyi Li, Author
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1. Introduction

Systems containing multi-phase flow in porous media are of interest in many different fields including environmental engineering, hydrogeology, and petroleum engineering. Nowadays, a great many environmental issues such as global warming, groundwater contamination, and oil recovery attract human interest. The current state of research has shown that a thorough understanding of flow in porous media has significant potential to help us solve some of these issues. Understanding the movement and physical properties of multi-phase fluids in porous media has significant applications in different environmental protection industries, in particular groundwater remediation of non-aqueous phase liquid (NAPL) contaminants.

Among the first quantitative studies of flow in porous media was that published by Henry Darcy in 1856, and porous media flow has been actively studied since. The models of multi-phase flow typically rely on extensions of Darcy’s law and empirical relationships. Capillary pressure (Pc) as a function of water saturation (S) is widely used to describe the capacity of porous media to retain or drain fluids. The relationship between capillary pressure and saturation not only depends on the physical properties of the system, but is also influenced by hysteresis, i.e., the difference in the Pc-S function between imbibition and drainage. Pc is generally measured externally by a pressure transducer. As an
alternative, it has recently become possible to estimate capillary pressure by using curvature values combined with the Young-Laplace equation (Armstrong et al. 2012b). Synchrotron based x-ray microtomography provides a robust way to perform three dimensional measurements, such as curvature analysis, without destroying the experimental samples (Andrew et al. 2014b, Wildenschild and Sheppard 2013). The general image processing procedures include noise removal, edge enhancement, segmentation etc. which transform greyscale data sets to labeled images. The labeled images are the foundation of various measurements such as porosity and saturation, Euler number analysis, interfacial area computation, and curvature calculation. Using the Young-Laplace equation to estimate curvatures, Armstrong et al. (2012b) demonstrated good agreement between transducer-based and curvature-based measurements when connected-phase interfaces were considered. The explicit agreement between transducer-based and curvature-based capillary pressure measurements displays promise for the use of image-based estimates of capillary pressure for interfaces that cannot be probed with external pressure transducers, and needs further analysis. The objective of this study is to (1) investigate whether the image-based capillary pressure estimation method extends to the interfaces between disconnected non-wetting phase and connected wetting phase, and (2) to fully understand the curvature distribution histograms for the disconnected oil phase interfaces during drainage and imbibition that were presented by Armstrong et al. (Figure 6 in Armstrong et al. 2012b).
Local morphology, changes in saturation, surface area and volume of disconnected non-wetting phase are considered during both drainage and imbibition. The results indicate that curvature evolution, and thus the capillary pressure between disconnected non-wetting phase and connected wetting phase is mainly controlled by local morphology instead of bulk capillary pressure.
In order to increase the confidence and accuracy of Pc estimates, it is necessary to control the quality of the original raw images, and improve current image processing procedures as well. Thus a specific objective of this research is to examine different image processing scenarios, analyzing results for each step using real image files as well as synthetic data sets.

A thorough image processing procedure has been established and implemented on existing data sets to further analyze interfacial behavior in that particular system: A novel method was introduced in this research to ameliorate curvature measurement with selective interface modification and distance weighting. Results indicate the errors were drastically reduced as compared to the approach used in Armstrong et al. (2012b).
2. Background

A multi-phase system of fluids in porous media usually consists of a gaseous phase, a liquid phase, and a solid phase. The gas phase is generally controlled by the atmospheric constituents such as N\textsubscript{2}, O\textsubscript{2}, CO\textsubscript{2}, etc., unless specified. The liquid phase is commonly dominated by water. Most scientific studies suggest that it is valid to assume water is incompressible because it only experiences pressure under -10 atm (Selker 1999). The solid phase often consists of complex organic and mineral compositions. For this research, glass beads are selected to imitate the solid phase for simplicity. Before introducing the details of this research, some definitions must be clarified in advance.

2.1 Porosity:

The porosity, or void fraction of a porous medium indicates the volume fraction which is not occupied by solids.

\[
n_v = \frac{\text{sample volume} - \text{volume of pure mineral}}{\text{sample volume}} = 1 - \frac{\rho_{vb}}{\rho_s}
\]

Eq #1

where \( n_v \) is porosity, \( \rho_{vb} \) is the dry bulk density and \( \rho_s \) is the solid density. The solid density \( \rho_s \) is a constant once the solid sample (material) has been chosen. Therefore, porosity is vaguely correlated to solid density. The bulk density of a medium mainly depends on two characteristics, the physical structure of the medium and its particle size distribution. Fine particles are easily organized into the space between larger particles which lead to relatively higher bulk density and lower porosity.
2.2 Degree of Saturation:

An important part of porous media research is to quantify and analyze liquid content. In most cases, liquid content is presented on a volume basis ($\theta$) as shown below.

$$\theta = \frac{V_w}{V_t}$$

$$w = \frac{M_w}{M_s}$$

where $V_w$ and $M_w$ are the volume and mass of the liquid phase respectively. $V_t$ is the total volume, and $M_s$ is the mass of the sample. Mass based liquid content ($w$) is also available but is rarely used. In reality, it is difficult to fully saturate all the pores with water because of the complexity of porous media. Some of the gas phase will remain in the pore space due to trapping in dead end pores. Conversely, during drainage from a fully saturated the porous medium, water held in thin films will remain in place unless heated continuously. Therefore, degree of saturation ($S$), which is defined based on the volume basis liquid content, is the best way to describe the quantity of water contained in a material.

$$S = \frac{\theta}{\theta_s}$$

where $\theta$ is the current liquid content and $\theta_s$ is the saturated liquid content.

The retention of water in soil is due to Van Der Waals attraction, electrostatic dipole, osmotic force, and surface tension. In porous media, the first three attractions are limited to thin layers or films and especially stagnant water. Surface tension on the other hand plays a major role for bulk water.
2.3 Surface Tension and Capillary Pressure:

Surface tension is the tendency of a liquid surface to allow it to resist an external force. It has dimensions of energy per unit area, or force per unit length as expressed in the following equation (Eq #4). At equilibrium, fluid pressures are the consequence of surface tension which curves interfaces in porous media. In order to be more specific, a capillary tube is the best example for clarification (Figure 2.1). The relationship between surface tension and pressure is described by the Young-Laplace equation, as shown below in equation 4.

\[ \Delta P = P_{nw} - P_w = 2\sigma_k \frac{2\sigma \cos \alpha}{R} \]

Figure 2.1 Model of Capillary Pressure
$\Delta P$ is the capillary pressure acting on the surface which is cause the pressure difference between the wetting phase $P_w$ and the non-wetting phase $P_{nw}$. $\sigma$ is surface tension of the interface and $k$ is mean curvature. The Contact angle ($\alpha$) is measured as the angle between the interface and the solid surface. $R$ is the radius of the capillary tube. When measuring contact angle for a given phase, the phase is determined as non-wetting for $\alpha > 90^\circ$ and wetting for $\alpha < 90^\circ$. As illustrated in the figure above, Laplace equation demonstrates that, for this particular case, the pressure of non-wetting phase is greater than the wetting phase, which bends the interface to a concave shape.

2.4 Mean Curvature:

For a two dimensional model, curvature is the inverse of the radius for the infinitely close inscribed circle at a particular point. A 3-D model is more complicated because the normal vector and tangent vector will be introduced into the curvature measurement as shown in Figure 2.2. All curves with the same tangent vector occupy the same normal vector. Taking all possible curvatures at a point on a surface into account, the maximum and minimum curvature values are called the principal curvatures with the symbol $K_1$ and $K_2$, respectively. The product of the principal curvatures ($K_1 K_2$) is called Gaussian curvature named by Carl Gauss with a dimension of $1/\text{length}^2$. Mean curvature, which is defined as half the sum of the principal curvatures $(K_1 + K_2)/2$, has been used widely in different research fields (Wernersson et al. 2011).
2.5 Wettability:

Wettability describes the preference of a solid surface to be in contact with one fluid rather than another. The term “preference” used in this definition may not be accurate, but it describes the balance of interfacial forces. A drop of a favorable fluid displace a less favorable fluid. In some extreme circumstances, the drop will either spread over the entire surface, or bead up depending on the wettability of the solid surface. As shown below in Figure 2.3, a water drop (blue) will spread over the whole surface if the solid is completely water-wet (right), resulting in a contact angle equals 0°. Conversely, it will bead up if the solid surface is oil-wet (left). For an intermediate wetting surface (middle),
the shape of the water drop depends on the balance among the three interfacial tensions as indicated below, which are $\sigma_{sw}$, $\sigma_{sa}$, and $\sigma_{aw}$ for solid-water, solid-air, and air-water, respectively.

\[
\sigma_{sw} = \sigma_{sa} \cdot \sigma_{aw} \cos \alpha
\]

Figure 2.3 Wettability and Contact Angle

2.6 The Characteristic Curves:

By using the Young-Laplace equation, we can equate pore size to the capillary filling pressure of each pore. Water saturation is the ratio of water volume to the total available pore volume as mentioned before. A typical relationship between capillary pressure and water saturation for a porous medium with a non-hosted particle size distribution is shown below in the characteristic curves (Figure 2.4). Generally, the characteristics curves have an S shape due to the pore size distribution. Figure 2.4 indicates that the imbibition process is delayed until the pressure is sufficient to fill the largest pores (pore body) in the media. Conversely, once the porous medium is filled, the drainage process does not begin until the pressure is high enough to drain the water out of the smallest pore necks. Therefore, Figure 2.4 gives information about the effect of pore size
distribution on fluid retention. Imbibition and drainage have different pathways, referred to as hysteresis, which will be clarified below.

Hysteresis is caused by the effect of characteristic pore radius and pore structures. The characteristic radii are denoted as $R_b$ for imbibition and $R_n$ for drainage as indicated in Figure 2.5. For a given saturation, there are two possible capillary pressures associated, depending on whether the porous medium is in the process of filling or emptying. Therefore, in order to estimate the capillary pressure, the history of the system must be known in advance.
Figure 2.5 The Characteristic Radii of Pore Space During Imbibition and Drainage (Modified after Selker 1999)
3. Literature Review

Flow of multiphase fluids in porous media has been studied via different methods for decades. Hydraulic properties of unsaturated porous media are commonly characterized by capillary pressure-saturation (Pc-S) curves. Because of the connection between capillary pressure and curvature described by the Young-Laplace equation, new avenues of exploration have opened up with recent developments in pore-scale imaging and image analysis. Interfacial curvature has been shown to be an important parameter in several studies (Cheng et al. 2004, Armstrong et al. 2012a).

Armstrong et al. (2012b) compared capillary pressure based on both curvature values and external pressure transducers. The results not only indicated agreement between these two methods, if connected phase interfaces were considered, but also demonstrated potential for the use of image-based estimation for those fluids that cannot be probed with external pressure transducers (Armstrong et al. 2012b). Two fundamentally different curvature estimates are available. The first approach is based on image intensity gradient (voxel-based). However, the application of voxel-based curvature estimation contains various limitations and is highly sensitive to image quality (Bullard et al. 1995, Thirion and Gourdon 1995). In the second approach, curvature is measured based on the triangulated surface generated from the segmented images (surface-based). The marching cubes algorithm is generally adopted for surface generation (Lorensen and Cline 1987). Although the second approach has been commonly used, the amount of smoothing needs
to be examined. Segmentation accuracy and image pixelization also affect the curvature estimates (Armstrong et al. 2012a).

Advances in digital image processing provide a direct method to non-destructively visualize porous space and the fluids within it. In particular, fluid features such as trapped blob morphology (Al-Raoush and Willson 2005, Schnaar and Brusseau 2005) and interfacial area (Culligan et al. 2006, Culligan et al. 2004, Narter and Brusseau 2010) can be measured via a three dimensional, high resolution image generated by x-ray computed tomography.

By using image analysis techniques, curvature values measured from the perspective of the water phase, can be used as a wettability indicator and calculated based on computed microtomography images (Brown 2012). Brown et al. (2014) also proved that curvature values are not only influenced by image resolution, but also affected by segmentation algorithms. A more accurate mean curvature estimate method would benefit further analysis (Brown et al. 2014).

Capillary pressure can be conveniently calculated based on bulk fluids mean curvature values, thereafter, employed for different needs. Curvature-based capillary pressure measurement was used for capillary number estimates, therefore, for the needs of non-wetting phase mobilization predictions (Andrew et al. 2014a, Armstrong et al. 2014). Jamaloei et al. (2011) further investigated the influences of wettability on the microstructure and the pore-scale mobilization of residual non-wetting phase in porous media. Mean curvature values were measured for capillary number and Weber number estimates (Yadali Jamaloei et al. 2011).
In addition, Haines jump which is a phenomenon describing non-wetting fluid displacing wetting fluid in porous media, is also related to curvature values. At the same capillary number, interfacial dynamics depend on the changes of viscosity and interfacial tension. Armstrong et al. (2015) illustrated that interfacial curvature value which is directly related to surface tension and pore morphology, is an important parameter to analyze for the study of Haines jumps (Armstrong et al. 2015). Instead of using x-ray computed tomography, Hsu et al. (2012) correlated the mobilization of disconnected non-wetting blobs with gravity force and hysteresis by implementing a non-invasive planar laser-induced fluorescence visualization technique for interfacial property measurement. Mean curvature values were estimated herein to test the gravity effects on residual oil blobs in porous media (Hsu et al. 2012).

As evident from the above referenced studies, surface-based curvature estimation approach has been used for different purposes. However, as far as what we know, most of the recent publications are mainly focused on image quality for curvature estimate improvement. How to increase the accuracy of curvature measurement regardless of image resolution needs to be studied and will be discussed in this research. Additionally, curvature-based capillary pressure applied to disconnected fluid interfaces needs to be further investigated and will be included as well.
4. Materials and Methods

4.1 Pore-Scale Imaging

4.1.1 Experiment Setup

The experiment discussed here was conducted at the Advanced Photon Source, Argonne National Lab in Chicago by Mark Porter in 2006 (Porter et al. 2010). The experimental system consisted of glass beads with a ratio of 35% 0.6mm diameter, 35% 0.8mm diameter, and 30% 1.0-1.4mm diameter packed in a 25.0mm long glass column with an inside radius 3.5mm. Two phases involved in this system are Soltrol 220 (non-wetting phase, ρ=0.79g/cm³, σ=0.0378N/m) and potassium iodide doped water (wetting phase, 1:6 mass ratio of KI:H₂O). A semi permeable hydrophilic membrane was placed at the bottom of the column in order to prevent the non-wetting phase from entering the water path. A rubber stopper containing the non-wetting phase outlet line was placed at the top of the porous medium inside the column. Wetting phase was controlled to ±1µl by a syringe pump (Gilson 402). The complete core holder assembly is shown below in Figure 4.1. Further details are provided in Porter et al. (2010).
The system was first fully saturated with wetting phase, followed by primary drainage (PD), main imbibition (MI), main drainage (MD), secondary imbibition (SI), and secondary drainage (SD). At each drainage or imbibition, a precise amount of wetting phase was pumped out of (drainage) or into (imbibition) the system at a flow rate of 0.6ml/hr. About 15 minutes were required by fluids to reach equilibration. Images were obtained from a 5.5mm section of the column at 13 µm/pixel resolution via x-ray computed tomography.
4.1.2 X-ray Computed Tomography:

X-Ray computed tomography is a non-destructive technology which is helpful for different research due to its powerful penetration ability. By using this technique, small objects inside a larger body can be visualized and analyzed without destroying it. The common components of x-ray microtomography setup are an x-ray source, an object to be imaged, and detectors as shown in Figure 4.2. The rotation stage provides images from different angles, resulting in a 3-D image instead of a projection (2-D) image. When x-rays penetrate the object, some are attenuated and others are transmitted. Therefore, it is necessary to control the energy of x-ray in order to control the image quality. The Beer-Lambert law relates attenuation with the sample thickness D via the following equation:

\[ I = I_0 e^{-\mu D} \]  

Eq #5

where \( I_0 \) is the original intensity, \( D \) is the thickness of the sample and \( \mu \) is the attenuation coefficient [L\(^{-1}\)], and \( I \) is the result intensity.

Figure 4.2 Synchrotron-Based x-ray Microtomography Setup (Wildenschild and Sheppard 2013)
In this research, synchrotron based computed microtomography was used for image generation due to its higher resolution. Synchrotron radiation is electromagnetic radiation produced by high speed electrons spiraling in a particle accelerator. X-rays are produced as a result of electrons decelerating.

X-ray attenuation depends on the density of a material and its elemental composition. Water and Soltrol attenuate x-rays to a similar degree. Thus, in order to identify the wetting phase in this system, potassium iodide was chosen as a dopant in this research because its attenuation will be significantly increased above a specific energy level as indicated in Figure 4.3. By changing the energy to above the photoelectric absorption edge for iodine [33.2 keV], the wetting phase will be easily distinguished due to its higher attenuation comparing with other phases.

![Figure 4.3 Photoelectric K-edge Plot for Iodine](image-url)
4.2 Data Collection

To generate a data point, images were generated from a 5.5mm section of the column at 720 angles over a 180 degree column rotation. The voxel resolution was 13 µm/pixel as mentioned before. The glass beads without fluids were scanned first as a “dry scan” image to allow for more straightforward segmentation. Generally, tomographic image processing procedures include acquisition and reconstruction, image processing with filtering, segmentation, and data analysis. All the two-dimensional radiographs were reconstructed and preprocessed with algorithms developed by GeoSoilEnviro Consortium for Advanced Radiation Sources (GSECARS) (Rivers et al. 1999) to produce 3D volumes of data. In the reconstructed grayscale image, the white area represents the water phase, glass beads appear grey, and the oil phase is black as shown below in Figure 4.4.

Figure 4.4 Reconstructed Raw Data File (MI 10.dat)
### 4.3 Noise Removal

The reconstructed volume data files were loaded into Avizo® for further processing.

Ideally, all pixel values in a uniform part of an image should have exactly the same intensity. But as seen in Figure 4.5, the original grayscale data file displays a relatively featureless histogram which does not benefit segmentation. In order to perform accurate data analysis, segmentation is a crucial step, and heavily relies on the fidelity of the images. Hence, filtering is necessary for noise removal in this case.

![Figure 4.5 Snapshots and Histograms for Grayscale (Top) and NLM Filtered (Bottom) Images](image)

In order to reduce the effects of image noise and blur, a non-local means (NLM) filter was first introduced due to its reported efficiency in noise removal (Buades et al. 2005). The search window, local neighborhood and similarity value, which define the window
size, the importance of each voxel in the window, and weight factor, respectively, were set to 4, 7, and 3. Based on the result shown in Figure 4.5 (top image), it was a challenge to identify peaks and valleys in the original histogram. However, after applying the non-local means filter, most of the noise was removed which leads to a more homogeneous image and a sharper histogram with distinct peaks representing oil, water, and beads, respectively (Figure 4.5, bottom image).

4.4 Segmentation

Image segmentation is a process of dividing an image into different regions. The main goal of segmentation is to identify and simplify an image into a new labeled image that is easier to analyze. In general, different kind of approaches are available such as converging active contours (Sheppard et al. 2004), watershed segmentation (Vincent and Soille 1991), Bayesian Markov Random Fields (Kulkarni et al. 2012). In this research, the watershed segmentation as described by Vincent et al. (1991) was adopted, which simulates rain falling and each drop eventually descends into a catchment basin as clarified in Figure 4.6. The watershed represents the ridge between each catchment basin. Each peak stands for a watershed while a valley is a catchment basin. As long as the water line is increasing, it approaches to the transition regions between each basin. The same theory is applied for image segmentation procedure. Areas of high intensity in the image represent a catchment basin, while all the valleys (low intensity areas) stand for watershed. Transition regions will be found based on intensity gradient and every single voxel eventually belongs to one phase at the end.
4.4.1 Initial Segmentation

Using the watershed segmentation method we first identify the known regions in the data by using a simple threshold. The solid phase can be easily identified from the dry image. Afterwards, the unknown transition regions, which were found at the point of inflection between the two known phases, are determined by considering the image intensity gradient. All the known regions will be put into the watershed module in Avizo®, in
which they are grown into the transitional region. This was also the approach used by Armstrong et al. (2012b).

Finally, the transitional regions will be minimized by the watershed module and labeled such that the 64,000 greyscale values are sorted into only 3 classes: oil, water, and beads. Figure 4.7 shows that all the glass beads in this system are now white in the labeled image, water phase is in gray, and oil phase is black.

Figure 4.7 Filtered Image and Labeled Image

The accuracy of image segmentation may influence further analysis. Therefore, it is necessary to compare the labeled image with the original grayscale raw data in order to check if the segmentation procedure is accurate. The following images (Figure 4.8) indicate some segmentation artifacts due to movement of a few of the unsintered glass beads. In the color labeled image, the core holder was cropped away and the outside was marked as black. The glass beads are labeled in light blue, while connected non-wetting (oil) phase is in dark blue. Disconnected non-wetting phase is green. Connected wetting (water) phase is red and disconnected wetting phase is yellow (not shown here). By comparing with the raw grayscale image, we see that two ganglia were misclassified as connected non-wetting phases as indicated by orange ellipses. Thus, a more accurate
image segmentation procedure is needed to correct for bead movement and is described in section 4.2.2.

![Grayscale Image and Segmented Labeled Image](image)

**Figure 4.8 Grayscale Image and Segmented Labeled Image**

### 4.4.2 Corrected Image Segmentation

As mentioned before, minor bead movement was found in every data set during both drainage and imbibition, which was the cause of the segmentation artifacts. In order to label the data correctly, segmentation must be implemented based on both the dry scan and every single partial saturated grayscale images. In other words, the static beads will be labeled based on the dry image and more beads generated from each grayscale image will be used for artifact removal. The result is shown below in Figure 4.9 for comparison. All the segmentation artifacts were classified to the bead phase now.
4.5 Surface Generation

Once the final segmented image is obtained, a three dimensional surface can be generated, for instance using the marching cubes algorithm (Lorensen and Cline 1987) implemented in Avizo®. This algorithm approximates a smoothed interface between different phases by generating thousands of triangles. The numbers of triangles associated with the resulting interface mainly depend on the resolution of the input images. Once the segmented image is labeled, the surface generation module can be applied with different smoothing type and extent. Smoothing has relatively little impact on curvature estimates if the principal radii of curvature are around 100 times the voxel size (Andrew et al. 2014a). For the dataset analyzed here, voxel size is of the same magnitude as the principal radii of curvature, which means the effect of smoothing cannot be neglected. The surface generation module in Avizo® provides two smoothing options: constrained...
and unconstrained smoothing. The first mode guarantees that no label is altered during surface generation, while unconstrained smoothing does the opposite. Smoothing extent is studied in detail due to its parabolic characteristics. In other words, higher smoothing extent does not necessarily result in an improved interface. The following table (Table 4.1) indicates that constrained smoothing with smoothing extent 3 not only preserves the original data, but also smooths the interface at the same time. Consequently, constrained smoothing with smoothing extent 3 is selected in this research.
Table 4.1 Comparison between Constrained (Top) and Unconstrained (Bottom) Smoothing with Different Extent

<table>
<thead>
<tr>
<th>Extent:</th>
<th>9</th>
<th>7</th>
<th>5</th>
<th>3</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><img src="image1.png" alt="Image" /></td>
<td><img src="image2.png" alt="Image" /></td>
<td><img src="image3.png" alt="Image" /></td>
<td><img src="image4.png" alt="Image" /></td>
<td><img src="image5.png" alt="Image" /></td>
</tr>
</tbody>
</table>
Figure 4.10 Surface View of MI 10 with (Left) and without (Right) Segmentation Artifacts

4.6 Regenerate Surface

With the new labeled images, interfaces between connected wetting and disconnected non-wetting phases were regenerated as shown above in Figure 4.10. By resegmenting the data, the segmentation artifacts, which lead to incorrect objects as indicated in the orange circles, were removed completely. Comparing with the previous study (Armstrong et al. 2012b), the new interface without segmentation artifacts produces an approximate unimodal curvature histogram instead of a bimodal histogram as shown below in Figure 4.11. More curvature analysis will be discussed in the result section.
Figure 4.11 Histograms of MI 10 with (Orange) and without (Blue) Segmentation Artifacts
5. New Curvature Estimation Approach

The smoothing type and extent used when generating the surfaces were thoroughly investigated in the previous chapter. Constrained smoothing with extent 3 was used for further analysis. A synthetic dataset of a meniscus in a capillary tube was generated to test the new curvature estimation approach. As shown below in Figure 5.1, no matter what the data size is, the edge of the interface, close to the solid surface, is always rough, which contributes to mean curvature outliers. As mentioned before, mean curvature values are not only a wettability indicator, but also related to capillary pressure prediction, and therefore requires meticulous calculation.

Figure 5.1 Synthetic Interface between Non-wetting Phase and Wetting Phase
In order to increase the accuracy of the mean curvature values, the triangles that are close to the solid surface should be eliminated. In other words, mean curvature should be estimated without taking the segments of the interface that is affected by proximity to the solid surface into consideration.

In Avizo®, interfaces are approximated by thousands of triangles generated by the marching cubes algorithm (Lorensen and Cline 1987). Each triangle is defined by three vertices as shown below in Figure 5.2.

To improve the curvature estimation, we introduced two new approaches that eliminate the effect of the solid surface on the curvatures.
5.1 Clipping

The basic idea is to measure the distance between each triangle and its shortest edge (nearest solid surface) as shown below in Figure 5.3.

![Figure 5.3 Shortest Edge Distance Estimation](image)

The contour line of this particular interface was first imported. Vertex distance calculation was performed based on Dijkstra’s shortest edge distance algorithm (Dijkstra 1959). The shortest edge distance of a specific triangle is estimated by taking the average of its three vertex distances. If its distance value is less or equal than a threshold, this particular triangle will be considered too close to the edge (solid surface) and eliminated at the end. Otherwise, the current triangle will be selected for mean curvature calculation. The general workflow of mean curvature modification procedures contains interface generation, vertex modification, triangles elimination, and curvature histogram reconstruction as indicated below in Figure 5.4.
Since each triangle has its own global coordinates, all the vertices can be exported into a three-column matrix in a Matlab file. The resulting Matlab file is going to be executed in order to load the triangles matrix into Matlab. More manipulations are needed due to the difference between Avizo Tcl script and Matlab syntax. The first Matlab script in the appendix can automatically directed to the desired folder, modify each vertex, and save the result into an Excel spreadsheet as output.

Afterwards, the modified vertex coordinates are loaded into the second Tcl script in the appendix. The main purpose of this script is to measure the shortest edge distance for each vertex and remove unwanted triangles (“clipping”). The distance value measured here will be implemented for curvature improvement as described later (“distance weighting”).

For clipping, a triangle will not be selected unless at least two corners meet the specific threshold. A single triangle with distance view is shown below in Figure 5.5 as an example. Presume the threshold is 10 pixels, the triangle on the left hand side meets the requirement and will be selected. Conversely, the one on the right hand side is eliminated.
from further consideration in this case. At last, mean curvature and distance values for the remaining and for the eliminated clipped triangles are exported in an Excel spreadsheet.

Subsequently, mean curvature histogram reconstruction is accomplished by using the third Matlab script in the appendix. The mean curvature values for the clipped triangles are subtracted from the original mean curvature histogram. Recalculating the mean curvature value for the clipped surface is not an option because curvature calculation is based on twenty surrounding triangles.

A synthetic capillary tube with an internal radius of 50 voxels was utilized here for verification. Theoretically, the mean curvature value of this meniscus is \(-0.02\) pixel\(^{-1}\). The reconstructed histogram is shown below in Figure 5.6 which indicates that the curvature value after “clipping” is narrowed and closer to the theoretical value.
5.2 Distance Weighting

The distance value for each triangle is estimated simply by taking the average of its three apical distance values, thereafter, used as a weight factor for curvature measurement. The “distance weighting” mean curvature value can be estimated based on the following equation:

$$k = \frac{\sum (Curvature \times Distance)}{\sum Distance} \quad \text{Eq #6}$$

where “Curvature” is the curvature value (1/pixel) for each triangle. “Distance” (unit: pixels) in the numerator is the averaged distance value for each triangle which gives more weight factor to those triangles far from the edge, and less weight factor for those close to the edge. The denominator of this fraction is the sum of all the distance values (unit in
pixels). Consequently, the overall distance weighted mean curvature value (k) can be estimated with a unit 1/pixel.

Additional synthetic capillary data sets with different radii were generated for further analysis and will be used only in Chapter 6.1. A semi-sphere was cropped by a column in order to create a perfect concave meniscus as shown below in Figure 5.7. With these synthetic data sets, the theoretical mean curvature values for each data set can be calculated by using the reciprocal of each radius. The surface area of each data is different due to the different radii. In order to perform identical influence study, same portion of each meniscus was eliminated (clipped). The results for comparison will be discussed in Chapter 6.

![Figure 5.7. The Synthetic Capillary Tube Interfaces](image-url)
6. Results

6.1 Improvement of Mean Curvature Estimates

In this study, the following five different mean curvature estimation approaches were tested for comparison. Percentage error was calculated based on the following equation:

\[
\% \text{error} = \frac{|\text{Calculated Mean Curvature} - \text{Theoretical Mean Curvature}|}{\text{Theoretical Mean Curvature}}
\]  

<table>
<thead>
<tr>
<th>Approaches:</th>
<th>Details:</th>
<th>Formula:</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Original</td>
<td>Mean curvature calculated based on the original histogram.</td>
<td>[k = \frac{\sum \text{Curvature} \times \text{Intensity}}{\sum \text{Intensity}}]</td>
</tr>
<tr>
<td>2. Negative Only</td>
<td>Mean curvature calculated based on all the negative numbers in the original histogram. (Yellow Part) as in Armstrong et al. (2012b)</td>
<td>[k = \frac{\sum \text{Curvature} \times \text{Intensity}}{\sum \text{Intensity}}]</td>
</tr>
<tr>
<td>3. Distance Weighting Only</td>
<td>Calculated based on the curvature and distance values for each triangle.</td>
<td>[k = \frac{\sum \text{Curvature} \times \text{Distance}}{\sum \text{Distance}}]</td>
</tr>
<tr>
<td>4. Clipping Only</td>
<td>Mean curvature calculated based on the remaining interface.</td>
<td>[k = \frac{\sum \text{Curvature} \times \text{Intensity}}{\sum \text{Intensity}}]</td>
</tr>
<tr>
<td>5. Distance Weighting with Clipping</td>
<td>Calculated based on the curvature and distance values for the remaining interface.</td>
<td>[k = \frac{\sum \text{Curvature} \times \text{Distance}}{\sum \text{Distance}}]</td>
</tr>
</tbody>
</table>

Table 6.1. Approaches for Mean Curvature Estimation
The results in Figure 6.1 indicate that approaches one and two introduced approximately 6 to 9 percent error. Without clipping, the distance weighted mean curvature calculation reduced the error to less than 3 percent if the radius of the semi-sphere is greater than 30 voxels. Comparing the original approach with the clipping only approach, surface clipping contributed to the error reduction mainly because of interface segments are associated with mean curvature outliers and therefore is most effective for larger objects (<3% error for sphere > 20 voxels).

The combination of clipping and distance weighting demonstrates the percentage error can be reduced to less than 1 percent for the larger objects (radius > 25 voxels), which means approach 5 is the optimal mean curvature calculation method. Therefore, for mean
curvature estimation, using a combination of both clipping and distance weighting generates the most accurate results.

6.2 Effect of Segmentation Artifacts on Curvature Estimates

The segmentation artifacts were successfully removed by resegmentation as mentioned before in Chapter 4. Interfaces between connected wetting and connected non-wetting phases, connected wetting and disconnected non-wetting phases were generated by using surface-generating algorithm in Avizo® with constrained smoothing and smoothing extent 3 as described in Chapter 4. The target interface was first isolated and the mean curvature was measured. The two principal curvature values were calculated based on each triangle of the interface, and mean curvature was estimated as the average of these two principal curvatures.

As shown in Figure 6.2, the interfacial curvature frequency histograms for connected wetting and connected non-wetting phase appear as expected. For both main imbibition and secondary imbibition, as water saturation increases, capillary pressure should decrease. In other words, mean curvature values should move closer to zero. Conversely, mean curvature values for main drainage and secondary drainage should be far from zero as water saturation decreases.

The effect of resegmentation, i.e., removing artifacts due to bead movement, is illustrated in Figure 6.3.
Figure 6.3 shows that the curvature-based estimate corresponds quite well with the transducer-based measurement for both of the two imbibition. On the other hand, results for the two drainage curves show relatively poor agreement with the transducer-based measurement. The results also show that curvature-based capillary pressure without segmentation artifacts is slightly closer to the transducer-based measurement. Compared with an entire interface, segmentation artifacts only contributed a small portion of the mean curvature values, so the effect on capillary pressure estimates is relatively minor.
Figure 6.2 Mean Curvature Histograms for Connected Wetting and Connected Non-wetting Phases
Figure 6.3 Effect of Segmentation Artifacts on Pc-S Curves for Main Imbibition, Main Drainage, Secondary Imbibition, and Secondary Drainage (curvature estimation was based on method 2 as the same as in Armstrong et al. (2012b))
6.3 Improved Curvature-based Capillary Pressure Estimates

A previous study by Armstrong et al. (2012b) had demonstrated that by considering connected fluid interfaces, transducer-based capillary pressure and curvature-based capillary pressure agreed well for imbibition at equilibrium, but not so well for drainage. Two possible hypotheses provided by Armstrong et al. (2012b) why curvature-based capillary pressure differed from transducer-based capillary pressure in their data include: short equilibration time and inaccurate mean curvature measurement (Armstrong et al. 2012b). The latter of these two hypotheses has been studied herein. We introduced a novel mean curvature estimate mentioned in Chapter 5. By applying this method to the connected fluid interfaces, the new curvature distribution histograms were calculated and shown in Figure 6.4. Result shows most of the positive mean curvature values were removed from the histograms. Interfacial tension was read from table and capillary pressures were again estimated using the Young-Laplace equation. Results show very favorable agreement between transducer-based capillary pressure and curvature-based capillary pressure during both imbibition and drainage. Figure 6.5 compares Pc estimates using this new method (gray triangle), to transducer-based estimates (blue diamond), and to the estimates of Armstrong et al. (orange square). Armstrong’s Pc estimates show a good match for the two imbibition and relatively poor agreement for the two drainage, as mentioned above. We see that mean curvature estimates were significantly improved by eliminating interface near the solid surface and taking distance weighting into consideration. The final results shown in Figure 6.6 display a similar characteristic Pc-S curves as mentioned before in Figure 2.3.
Figure 6.4 Connected Fluid Interfacial Mean Curvature Distribution for MI MD SI SD with Clipping
Figure 6.5 Pc-S Curves based on Connected Fluid Interfaces for MI MD SI SD with Clipping and Distance Weighting
Figure 6.6 Comparison between Final Curvature-based Pc and Transducer-based Pc for Connected Fluids Main Imbibition & Drainage (Top) and Secondary Imbibition & Drainage (Bottom)
6.4 Mean Curvature Estimates for the Interfaces between Disconnected Fluids

Curvature-based capillary pressure estimates were significantly improved for connected fluids. The disadvantages of transducer-based measurement is that the transducers cannot probe into any disconnected fluid. Yet, based on the images, we can evaluate the pressure state of disconnected non-wetting phase. The interfacial curvatures were measured for disconnected fluids interfaces here for analysis. As shown below in Figure 6.7, each histogram of imbibition and drainage appears to follow the same trend as discussed above, i.e. mean curvature are increasing or decreasing monotonically. However, some of the histograms display bimodal distributions, which suggest the need for further investigation.

The left-hand small peaks in the following bimodal histograms indicate that some of the disconnected non-wetting phase exists in a different capillary pressure state. In order to discover why some of the disconnected ganglia were trapped at an earlier capillary pressure, three ganglia as highlighted in Figure 6.8 were isolated and analyzed individually.
Figure 6.7 Mean Curvature Histograms for Connected Wetting and Disconnected Non-wetting Phases
Two kinds of single ganglion interfaces were generated in Avizo®. The opened surface, defined as the interface between two different fluids, was used for interfacial area measurement between the disconnected non-wetting phase and the connected wetting phase. Total surface area and volume of disconnected non-wetting phase were estimated via the closed surface thereafter. The example of opened surface and closed surface is shown below in Figure 6.9 for illustration.
For the first isolated disconnected non-wetting ganglion, results are shown below in Figure 6.10 and Figure 6.11. The volume of this ganglion was consistently increasing during both main imbibition and main drainage. In addition, this trend applied to total surface area and interface area. Likewise, similar phenomenon was found for secondary imbibition and drainage as well. As shown below (Figure 6.12), the isolated non-wetting ganglion was squeezed and confined into a pore body during main drainage. Non-wetting phase redistribution and restabilization are the two reasons why the volume of disconnected non-wetting ganglion is changing. A larger volume has greater potential to generate more surface area if deformation is neglected. Unlike the gas phase, the liquid phase in porous media is generally considered as incompressible. In other words, the volume of a single disconnected oil blob should not be affected by bulk capillary pressure that much. The total volume, surface area and interfacial area increased drastically after main drainage 03, which indicates that another oil blob became disconnected and merged with the first ganglion during main drainage 05.
Figure 6.10 The First Single Ganglion Analyses for Main Imbibition and Main Drainage
Figure 6.11 The First Single Ganglion Analyses for Secondary Imbibition and Secondary Drainage
Figure 6.12 Single Disconnected Non-wetting Phase (MD 01 MD 05 MD 11) with Isosurface View
Unexpectedly, the second and the third disconnected non-wetting ganglia highlighted in Figure 6.8 behaved different than the first one. As shown below from Figure 6.13 to 6.15, the total volume of these particular disconnected non-wetting phases remained the same while the interfacial area and the total surface area of these two ganglia behaved unpredictably during both drainage and imbibition. This indicates that the topology of the disconnected non-wetting phase was not only affected by the capillary pressure, but also was influenced by the surrounding physical conditions, i.e. the shape of the surrounding pores. More analysis were needed for further investigation.
Figure 6.13 The Second Single Ganglion Analyses for Main Imbibition and Main Drainage
Figure 6.14 The Second Single Ganglion Analyses for Secondary Imbibition and Secondary Drainage
Figure 6.15 The Third Single Ganglion Analyses for Secondary Imbibition and Secondary Drainage
Brown et al. (2012) has shown in a previous study that the change of wettability during three-phase flow is mainly due to x-ray exposure and the contact with non-wetting fluid. Assuming wettability alteration is not the case in these *two-phase* experiments, the wettability of solid surface should remain unchanged. Therefore, the contact angles between the wetting phase, the non-wetting phase, and the solid surface should be the same in each drainage or imbibition process. Also, the redistribution of the disconnected non-wetting phase can be neglected due to the small volume changes. Hence, local morphology, which is one of the most important porous media properties, was introduced here for thorough study. Since non-wetting phase restabilization happened during each drainage and imbibition, each disconnected non-wetting ganglion was pushing toward or away from the pore neck. As shown below in the following 2-D cartoon (Figure 6.16), the interfacial surface area decreases as the interface gets closer to the pore neck, conversely, increases as the interface approaches the pore body. This is the most reliable explanation for the unpredictable topology characteristics. We therefore conclude that the pressure of some of the disconnected non-wetting phase is controlled by pore morphology rather than bulk fluid pressure, and that is the reason for the secondary peak in the histograms.

![Figure 6.16 Interface Simulation in Porous Media](image)
The mean curvature values for disconnected oil blobs shown in Figure 6.7 were converted to capillary pressure via the Young-Laplace equation for investigation. The curvature-based and transducer-based Pc is shown in Figure 6.17. No matter during the process of imbibition or drainage, the capillary pressure of disconnected non-wetting phase remained the same. Noticeably, the capillary pressure for the first two data sets in main imbibition as highlighted were relatively higher comparing with others which should be eliminated. The main reason is due to the limited quantity of disconnected non-wetting phase. Only one single oil blob was presented in these two data sets which doesn’t represent capillary pressure for all the disconnected non-wetting phase at all. Consequently, once the non-wetting phase became disconnected, the capillary pressures of disconnected oil phase behaved constant.
Figure 6.17 Comparison between Curvature-based Pc and Transducer-based Pc for Disconnected Fluids
Main Imbibition & Drainage (Top) and Secondary Imbibition & Drainage (Bottom)
7. Discussion and Conclusion

Previous study hypothesized that longer equilibration time and more accurate curvature estimates would allow for capillary pressure stabilization and more similar curvature-based Pc and transducer-based Pc values (Armstrong et al. 2012b). Equilibration time was limited by beam line availability which is not considered in this study. More equilibration time would be helpful if longer beam time granted in the future. New curvature estimation method was introduced and tested on both synthetic data and real data sets. Both of the distance weighting and the clipping has a very positive effect on mean curvature estimates. By taking clipping and distance weighting into consideration simultaneously, results revealed that the curvature estimation errors were significantly reduced for both of the synthetic data and the real data sets.

Image segmentation artifacts were mainly due to the movement of the unsintered glass beads. Curvature values for the interfaces between connected wetting and connected non-wetting fluids were not influenced by segmentation artifacts that much due to its higher surface area. Conversely, segmentation artifacts had significant negative effects on curvature estimates for interfaces between the disconnected non-wetting phase and the connected wetting phase. Therefore, as expected, taking connected wetting and connected non-wetting phases into consideration, the curvature-based capillary pressure estimates were slightly improved by eliminating segmentation artifacts. However, based on the results, the removal of segmentation artifacts mainly contributed the curvature estimation improvement for disconnected non-wetting phase. The new curvature histogram in Figure
6.6 shows better result comparing to the former curvature analysis (Armstrong et al. 2012b).

Previous study demonstrated that curvature-based capillary pressure and transducer-based capillary pressure agreed well for imbibition at equilibrium if connected fluid interfaces were analyzed (Armstrong et al. 2012b). As mentioned earlier, the new curvature estimation method has great potential for curvature estimates improvement, which produced better Pc-S curves for main imbibition, main drainage, secondary imbibition, and secondary drainage as shown previously in Figure 6.6. Compared with the transducer-based capillary pressure, the new curvature-based Pc showed good match for all the imbibition and drainage.

Taking disconnected non-wetting phase and connected wetting phase interfaces into consideration, the questionable mean curvature estimates observed in Fig 6 in Armstrong et al. (2012b) were thoroughly studied in the previous chapter. This study showed that through use of our improved curvature estimation algorithms, we were able to produce more realistic looking curvature distributions (Figure 6.7 here vs Figure 6 in Armstrong et al). Our results also suggests that redistribution and restabilization are the main factors for the changing volume of disconnected non-wetting phase. Apparently, the curvature values and topology for disconnected non-wetting ganglia were not only influenced by the bulk pressure difference, but were even more strongly affected by the local morphology. A 2-D model provided in the previous chapter can roughly simplify the unpredictable phenomenon. But a more complicated 3-D model would be really helpful which need further investigation in the future.
As mentioned in the previous chapter, linking curvature-based capillary pressure estimates for disconnected non-wetting ganglia with bulk pressure difference is not an option. In other words, disconnected non-wetting phase and connected non-wetting phase differ in curvature at a given capillary pressure. The main reason is disconnected fluids are strongly affected by pore morphology. As shown above in Figure 6.16, taking disconnected fluid interfaces into consideration, curvature-based Pc remain constant regardless of the system saturation status, which indicates that residual non-wetting ganglia are less hysteretic. Therefore, curvature-based capillary pressure estimates will not extend to disconnected fluids for the needs of Pc-S curve study. These two phases should be considered individually.

In this study, a novel curvature estimation method was developed which significantly contributed to scientific parameter measurement, hence, is helpful for understanding and prediction of the fate and transport of multi-phase fluids in porous media. In the environment, once NAPLs snap-off and becomes disconnected, it becomes difficult to mobilize and remediate it because it is held in place by capillary forces. Study of disconnected non-wetting fluid will therefore support improved understanding of processes related to clean-up of NAPLs in groundwater and to other multi-phase systems in the environment. A more accurate mean curvature value can also be determined as a better wettability indicator which benefits groundwater remediation and oil recovery for petroleum engineering field.

Moreover, curvature-based capillary pressure measurement is an alternative way to estimate Pc for both connected fluid interfaces and disconnected fluid interfaces.
However, curvature-based \( P_c \) for disconnected oil blobs show no relationship with the bulk capillary pressure or the history of either imbibition or drainage. For further research, a 3-D model for oil blobs would be really helpful and benefits the understanding of how the disconnected non-wetting phase behaves in porous media.
8. Bibliography


9. Appendices

Appendix One

clc
clear
test=zeros(1000000,6);
for j=1:4
    i=['MD';'MI';'SI';'SD'];
    path =
    ['X:/res_vols/res_vol3/Nov2006LooseBeads/Tianyi/5Phase_Tianyi/Connected_Wetting_Connected_Nonwetting/Matlab/',i(j,:)]
    cd (path)
    list=dir(fullfile(path,'*.m'));
    nFile=length(list);
    for k=1:nFile
        file=list(k).name;
        run(fullfile(path,file));
        s=surface.faces;
        b=s-1;
        n=length(b);
        t=ceil(n/1000000);
        filename = [file 'faces.xlsx'];
        if n>1000000
            test=zeros(1000000,t*3);
            test(1:1000000,1:3)=b(1:1000000,1:3);
            test(1:n-1000000,4:6)=b(1000001:n,1:3);
            xlswrite(filename, test);
        elseif n>2000000
            test=zeros(1000000,t*3);
            test(1:1000000,1:3)=b(1:1000000,1:3);
            test(1:n-1000000,4:6)=b(1000001:2000000,1:3);
            test(1:n-2000000,7:9)=b(2000001:n,1:3)
            xlswrite(filename, test);
        else
            xlswrite(filename, b);
        end
    end
clc
clear
Appendix Two

# Triangle number
set i 0
# increase the value of "number" in order to see if all the vertices >= threshold
set too_close 0

# this is an example for how the first lines of s=surface.faces;
set v "
2 0 1
1 3 2
4 0 2
4 2 5
6 7 3
6 3 1
11 9 8
10 11 8
12 9 11
11 13 12
15 14 16
16 17 15
14 12 13
13 16 14
17 19 18
15 17 18
22 20 21
21 23 22
25 24 20
20 22 25
26 24 25
...
...
66 67 69
70 68 69
69 71 70
73 72 70
70 71 73
75 74 72"
set tri 0
set x 0
set va ""
"Surface View" buffer setValue 2 1
"Surface View" fire
"Surface View" buffer setValue 1 1
"Surface View" fire

create HxSpreadSheet MasterTable_WS
MasterTable_WS setNoRemoveAll 1
MasterTable_WS addColumn " " string
MasterTable_WS addColumn "MeanCurvatureRemaining" string
MasterTable_WS addColumn "Distance" string
set rowIndex 0

foreach n $v {
    if {["ShortestEdgeDistance" getValue $n] >= 2} {
        incr too_close
        set x [expr $x+["ShortestEdgeDistance" getValue $n]]
    }
    # ####################################################################
    # check for end of line (three vertices for each triangle)
    if {$tri<2} {
        incr tri
    } elseif {$tri==2} {
        set tri 0
        # ####################################################################
        # >=2 for remaining surface; <2 for clipped surface
        if {$too_close >= 2} {
            "Surface View" selectTriangles $i
            set va "$va ["MeanCurvature" getValue $i]"
            set x [expr $x/3]
            incr rowIndex
            MasterTable_WS setValue 1 $rowIndex [MeanCurvature getValue $i]
            MasterTable_WS setValue 2 $rowIndex $x
        }
        incr i
        set too_close 0
        set x 0
    }
}
Appendix Three

clc
clear
n=xlsread('MeanCurvature.xlsx');
m=xlsread('MeanCurvatureClipped.xlsx');
i=length(m);
j=length(n);
for x=1:i
    temp=0;
    for y=1:j
        if m(x)>=n(y,1)&& m(x)<n(y+1,1)
            if temp==0;
                if n(y+1,2)>0
                    n(y+1,2)=n(y+1,2)-1;
                    temp=1;
                elseif n(y,2)>0
                    n(y,2)=n(y,2)-1;
                    temp=1;
                end
            end
        end
    end
end
filename=('MeanCurvatureRemaining.xlsx');
xlswrite(filename,n,1)