

Supporting Information File

Quantitative imaging of gas adsorption equilibrium and dynamics by X-ray Computed Tomography

Ronny Pini^{a,*}, Lisa Joss^b, S. A. Hosseinzadeh Hejazi^c

^a*Department of Chemical Engineering, Imperial College London, London, United Kingdom*

^b*School of Chemical Engineering and Analytical Science, The University of Manchester, Manchester, United Kingdom*

^c*Amirkabir University of Technology, Tehran, Iran (Islamic Republic of)*

Abstract

This document contains the following sections:

S1. Uncertainty analysis of X-ray CT images

S2. Image segmentation

References

1. Joss, L. and Pini, R. (2017). *The Journal of Physical Chemistry C* **121**(48), 26903–15.

*Corresponding author

Email address: r.pini@imperial.ac.uk (Ronny Pini)

S1. Uncertainty analysis of X-ray CT images

As described in Sections 2 and 3.4 of the main manuscript, the amount adsorbed in moles per unit volume, n^a , is obtained by combining a scan acquired with CO₂ (\overline{CT}_a) with the corresponding registered scan of the bed saturated with Helium ($t = 0$, \overline{CT}_{He}):

$$n^a(t) = \frac{\overline{CT}_a(t) - \overline{CT}_{He}(t = 0)}{p_1 M_m} \quad (S1)$$

where $M_m = 0.044 \text{ kg}\cdot\text{mol}^{-1}$ is the molar mass of CO₂ and $p_1 = 1 \text{ HU}\cdot\text{m}^3/\text{kg}$ is the calibration factor. Eq. S1 was applied to compute pellet-, slice- or column-averaged amount adsorbed by using pellet-, slice- or column-averaged CT numbers. The uncertainty associated with the computed amount adsorbed at each measurement scale was estimated by following a protocol described in a previous publication [1]. Figure S1a-c shows the histograms obtained upon subtracting two single scans that have been acquired in sequence with the adsorption column saturated with Helium. Prior to subtraction, the scans have been reconstructed using different resampling schemes to yield voxels of different sizes up to the volume of one 1 mm-thick slice of the column ($V \approx 210 \text{ mm}^3$), as indicated in Table S1. The values of the obtained difference image follow a normal distribution centred in zero and with a standard deviation that decreases with increasing voxel volume. Because of the random nature of the noise associated with the X-ray CT images, the uncertainty associated with the CT value of any given voxel can be readily obtained as, $\sigma_{CT} = \sigma_{\Delta}/\sqrt{2}$, where σ_{Δ} is the standard deviation of the difference image (Figure S1a-c). The values obtained for σ_{CT} for each resampling scheme are plotted in Figure S1d and they are listed in Table S1, together with the uncertainty in the computed amount of CO₂ adsorbed, σ_n . The latter is readily obtained upon applying classic rules of error propagation to Eq. S1:

$$\sigma_n = \frac{\sqrt{2}\sigma_{CT}}{p_1 M_m} \quad (S2)$$

Res. scheme	Voxel dimension	Error CT	Error n^a
$x \times y \times z$	$(x \times y \times z)\text{mm}^3$	σ_{CT} [HU]	σ_n [mmol·cm ⁻³]
$16 \times 16 \times 1$	$1.14 \times 1.14 \times 1$	± 4.4	± 0.141
$16 \times 16 \times 2$	$1.14 \times 1.14 \times 2$	± 4.0	± 0.128
$32 \times 32 \times 2$	$2.27 \times 2.27 \times 2$	± 3.0	± 0.095
$32 \times 32 \times 4$	$2.27 \times 2.27 \times 4$	± 2.3	± 0.074
1 mm thick slice	210	± 0.4	± 0.012

Table S1: Image resampling schemes applied in this study, voxel dimensions and the associated uncertainties for the measured voxel CT value and the computed CO₂ adsorbed amount. The original image resolution is $(x \times y \times z)\text{mm}^3 = (0.071 \times 0.071 \times 1)\text{mm}^3$.

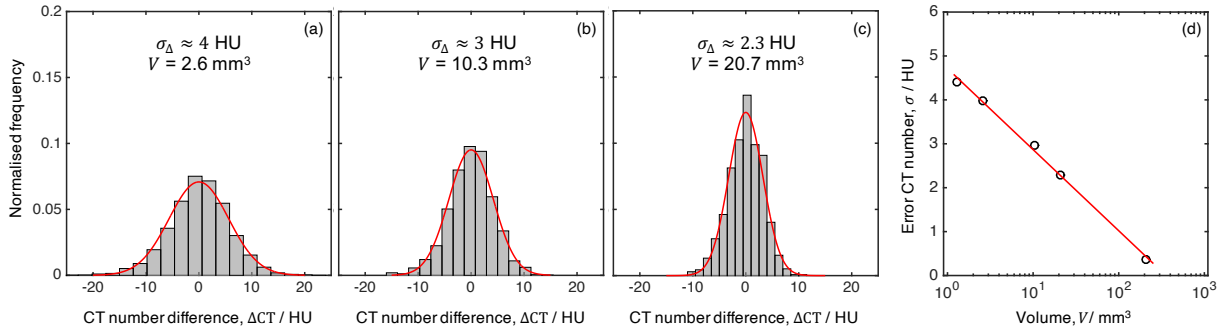


Figure S1: Histograms of the difference images obtained upon subtracting two scans of the rock sample acquired in sequence at the same location. The images have been reconstructed by applying different resampling schemes (see Table S1). The histograms have been fitted with a normal distribution (red solid curve) centred in zero and with a standard deviation, σ_{Δ} .

S2. Image segmentation

To extract adsorption isotherms of individual pellets within the adsorbent column, the XCT images were segmented using the procedure described below. The procedure uses a marker-controlled watershed segmentation and was implemented in MATLAB 2015a through the combination of various standard MATLAB functions. The workflow of the segmentation process is shown in Figure S2 and follows these steps:

1. **Image normalization** to modify the range of intensity values to $[0,1]$, i.e. $\overline{CT}_N = \overline{CT}/1000 + 1$ for $-1000 \leq \overline{CT} \leq 0$; values outside this range are set to either 0 or 1.
2. **Write the grayscale image** to a graphics file (.tiff) using `imwrite` (Figure S2A).
3. **Contrast enhancement** using contrast-limited adaptive histogram equalisation (`adapthisteq` using default parameters, Figure S2B).
4. **Marking of foreground objects** in the given image `I` by the morphological operation “opening-by-reconstruction” that uses disk-shaped structuring elements.
 - (a) Creation of the structuring element:
`se = strel('disk',4).`
 - (b) Application of the structuring elements to obtain markers by erosion:
`Ie = imerode(I, se)`
 - (c) By using the obtained markers, perform a morphological reconstruction of `I`:
`Iobr = imreconstruct(Ie, I);`
 - (d) Dilate the resultant image using the same structuring elements to obtain new markers:
`Iobrd = imdilate(Iobr, se)`
 - (e) Second morphological reconstruction (Figure S2C):
`Iobrcbr = imreconstruct(imcomplement(Iobrd), imcomplement(Iobr));`
`Iobrcbr = imcomplement(Iobrcbr);`
5. **Compute the background markers** (`bgm`).
 - (a) Binarise image to yield Figure S2D (`level` is identified manually for each slice by visual inspection):
`Ibw = im2bw(Iobrcbr,level);`
 - (b) Compute distance transform of the binarised image and its watershed transform:
`D = bwdist(Ibw); DL = watershed(D);`
 - (c) Watershed ridge lines (`DL == 0`) are set as the background markers (Figure S2E):
`bgm = DL == 0;`
6. **Compute the foreground markers** (`fgm`) by calculating the regional minima of the distance transform (Figure S2F): `fgm = imextendedmin(D,1)`

7. **Compute the watershed transform of the segmentation function** by modifying the gradient magnitude image.

(a) Compute the gradient magnitude of original image:

```
gmag = imgradient(I);
```

(b) Modify the gradient magnitude image, so that its only regional minima occur at the foreground and background marker pixels (Figure S2G):

```
gmag2 = imimposemin(gmag, fgm | bgm);
```

(c) Compute the watershed-based segmentation (Figure S2H):

```
L = watershed(gmag2)
```

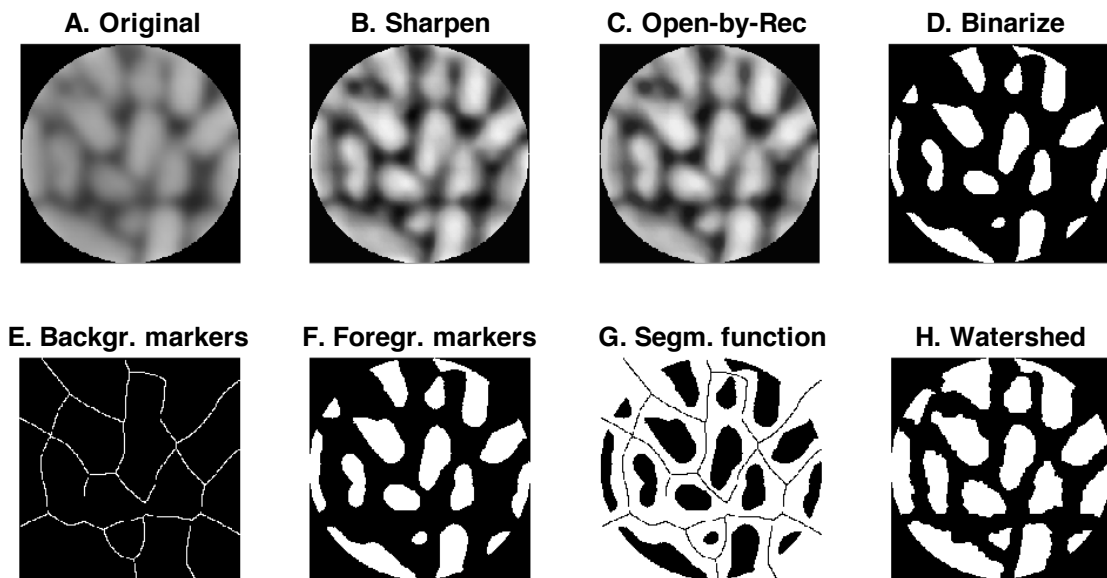


Figure S2: Image processing workflow for marker-controlled watershed segmentation (one slice shown as example).

The steps above were applied to each slice independently by using the same parameter values, with the exception of the threshold level in item 5. Upon stacking together the segmented slices, cross-sections belonging to the same pellet were merged together, resulting in the three-dimensional reconstruction shown in Figure S3.

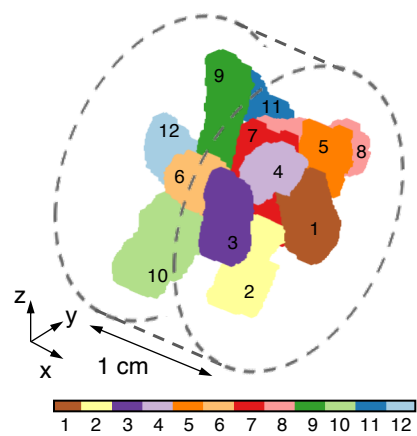


Figure S3: Three-dimensional reconstruction of a 1 cm-long section of the adsorption column, where 12 individual pellets have been identified by segmentation and color-coded accordingly.