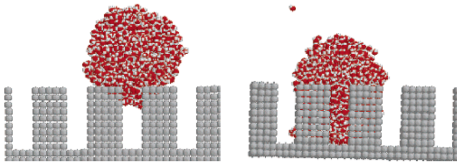
Sophie Finnigan

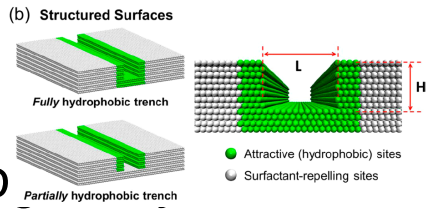
Constructing Complex Mesoscopic Surfaces

IBM

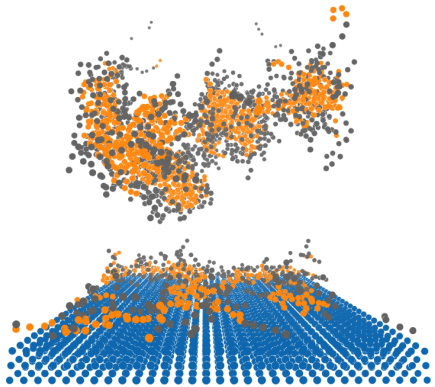
Introduction



- Surface interactions important to a wide range of applications
- Surfaces are naturally complex
- Current models are simple with limited hetero



- Fluid-surface interactions not accurately simulated or understood
- Lack accurate calculation of adsorption isotherms



Aims

- Create a methodology to generate complex surfaces
- Fits within flow
- Generates input ready for simulations
- Surfaces to be used to study:
  - Surface adsorption - calculation of adsorption isotherms
  - Competitive adsorption
- Development of complementary post-processing tools

Which simulation methodology?

Molecular dynamics (MD)

System evolves by Newtons laws of motion

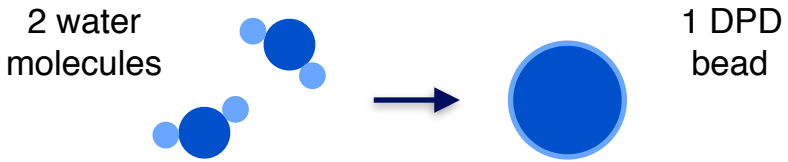
$$\nabla^2 r_i = \nabla v_i = F_i m_i$$

$$F_i = -\frac{\partial}{\partial r_i} U_i$$

U_i is a potential energy function

Dissipative particle dynamics (DPD)

Coarse-grained MD:



$$F_{ij} = F_{ij}^C + F_{ij}^D + F_{ij}^R$$

$$F_{ij}^C = \begin{cases} \frac{1}{2} A_{ij} r_{c,ij} \left(1 - \frac{r_{ij}}{r_c}\right)^2 & r_{ij} < rc \\ 0 & r_{ij} \geq r_c \end{cases}$$

Which simulation methodology?

Molecular dynamics (MD)

- Greater chemical insight into the surface
- Larger range of existing surfaces and parameters

- Time and length scale not large enough for surface adsorption
- Metastable structures due to slow dynamics
- Initial configuration needs consideration

Dissipative particle dynamics (DPD)

- Suitable for studying surface adsorption
  - Large system sizes can simulate bulk concentrations
  - Longer time scales can achieve equilibrium structures

- Limited previous surface modelling
  - Lack of parameters
- Loss of chemical information

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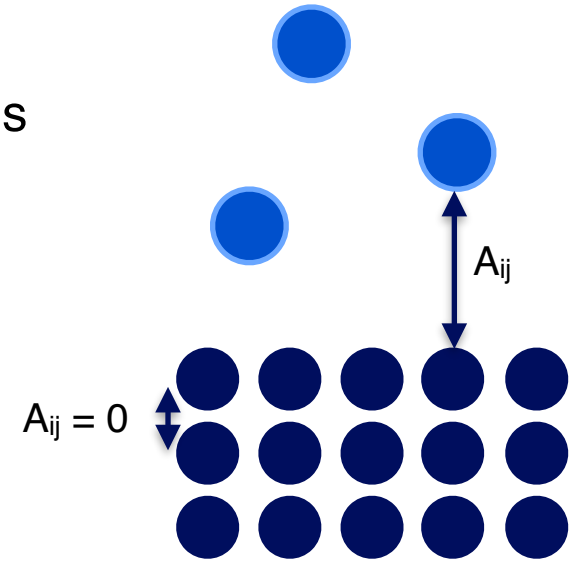
DPD Surface Modelling

Surface modelled as frozen beads:

- Intermolecular interaction with system beads
- No interaction with surface beads
- Fixed positions

DL Meso input files:

- CONFIG, CONTROL, FIELD



Surface Generation

Initial surface specification:

- Surface species
- Surface fractions
- Number of surface layers (n_z)
- Surface bead density (ρ_s)

Parameter Library

A_{ij} interaction parameters

FIELD

System dimensions

Surface thickness (l)

Number of beads (n_x, n_y)

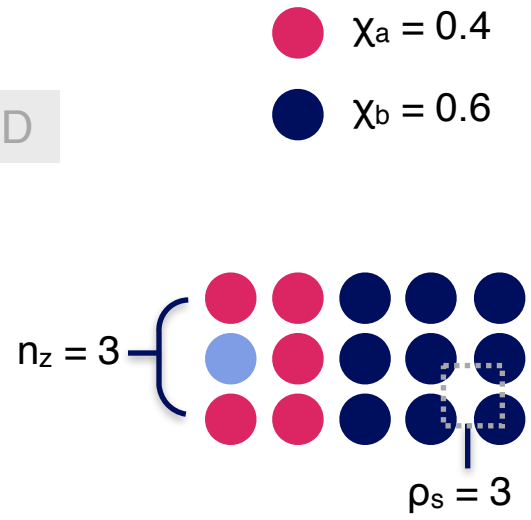
Lattice spacing

Updates cell volume

CONTROL

Calculates positions (x, y, z coordinates)

Surface CONFIG



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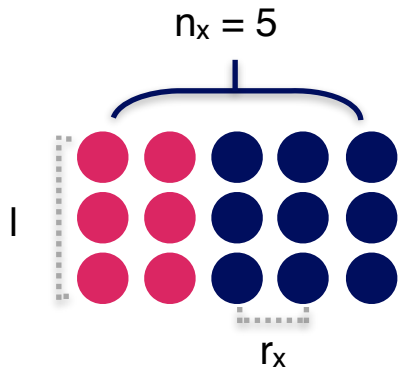

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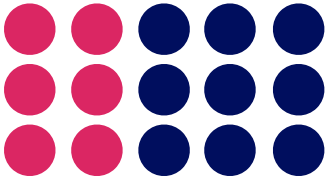

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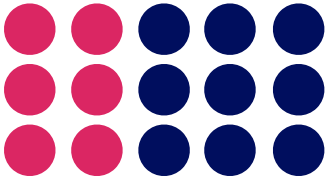

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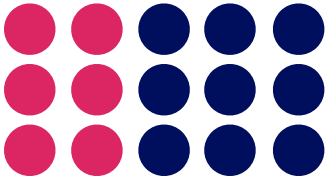

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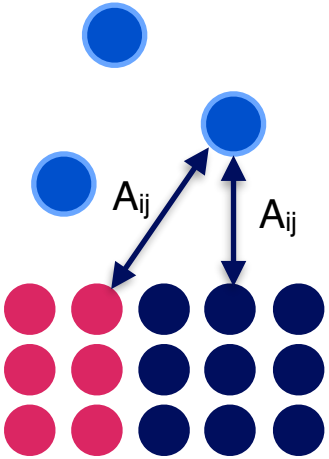
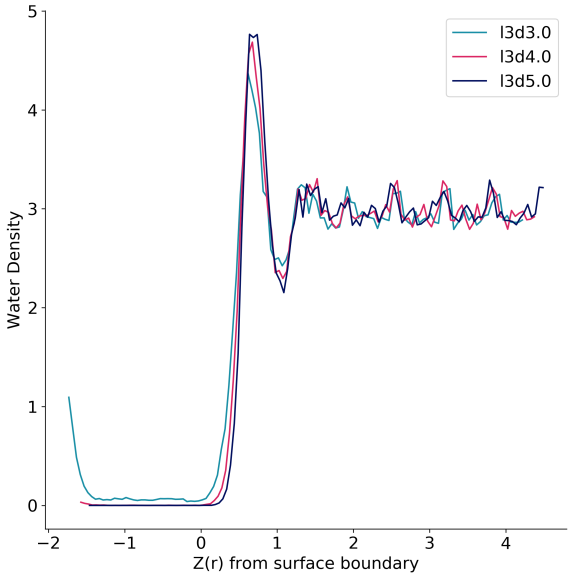
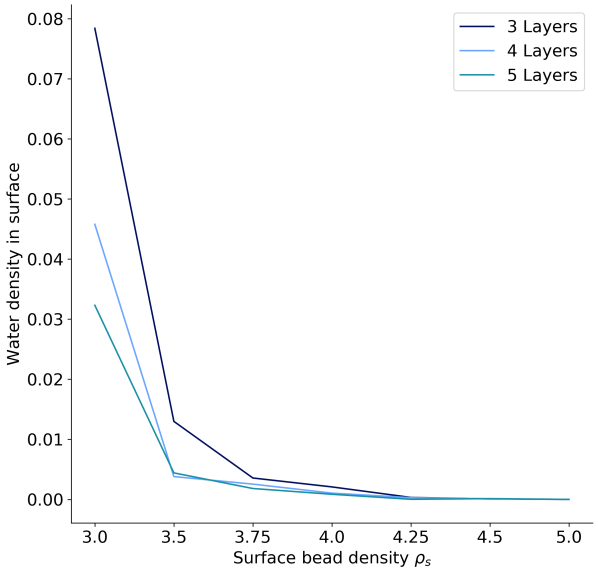


Surface Parameterisation

Investigated the effect of the input parameters

Want to avoid surface permeation and water ordering





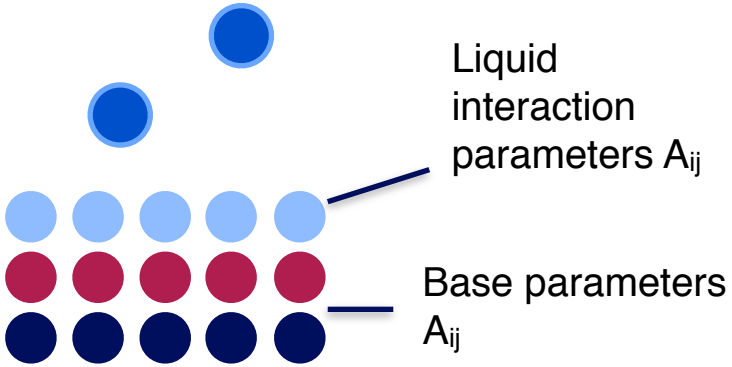
Selected defaults:

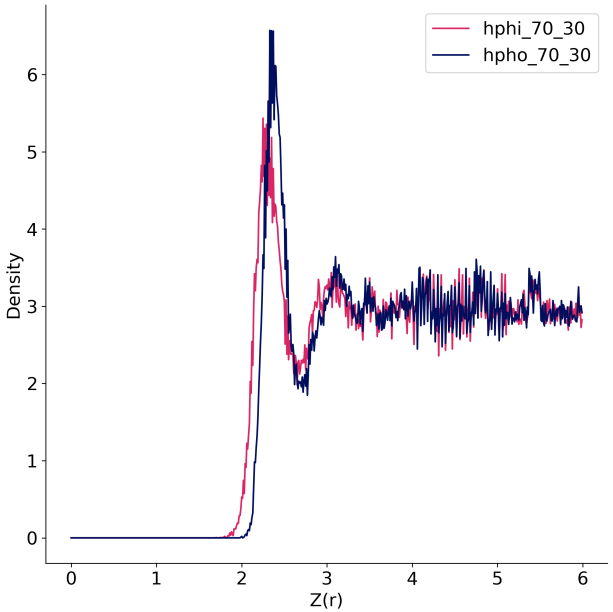
$\rho_s = 4$

$n_z = 3$

Surface Parameterisation

- Added definition of base surface layers
- Maintains a consistent base surface independent of the top layer parameters





Introducing Disorder

Initial surface specification:

Surface species
Surface fractions
Number of surface layers (l)
Surface bead density (ρ_s)

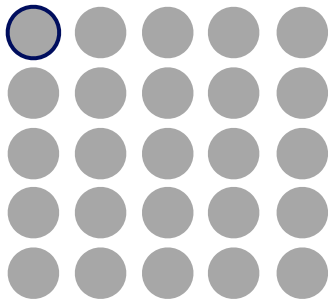
Order parameters (x, y)
Surface depth
Domain number (d_x, d_y)

Surface bead positions

Border beads of domain assigned by surface fractions

$\chi_a = 0.4$

$\chi_b = 0.6$



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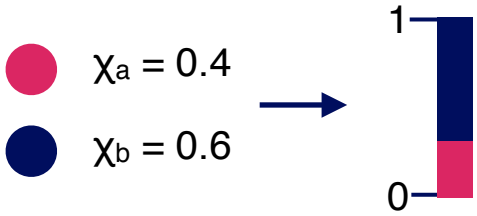

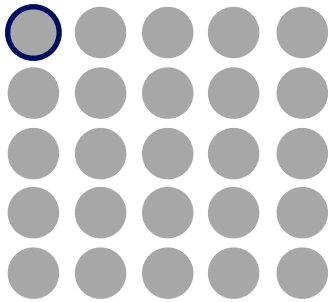

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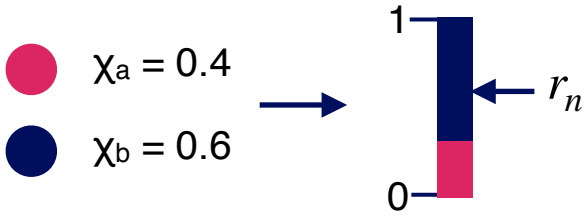

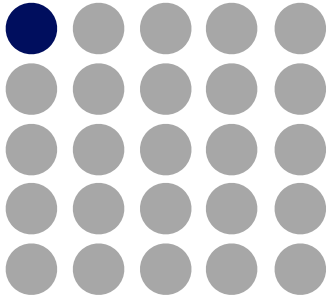

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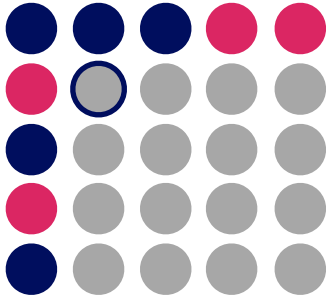
Assign rest of domain by order parameters and neighbouring species

$$r_n \begin{cases} r_n < x_{order} \\ r_n \geq x_{order} \end{cases}$$



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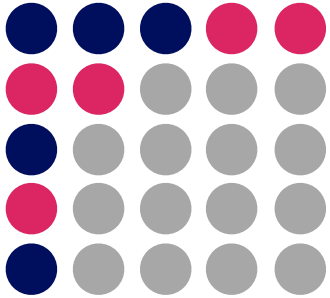
Bead = bead(x-1)



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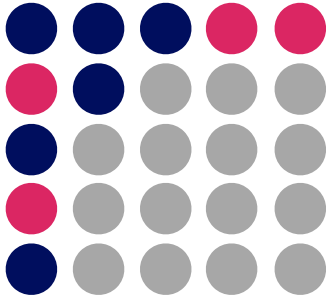
$$r_n \begin{cases} r_n < x_{order} & \text{Bead = bead(x-1)} \\ r_n \geq x_{order} & \text{Random assignment} \end{cases}$$



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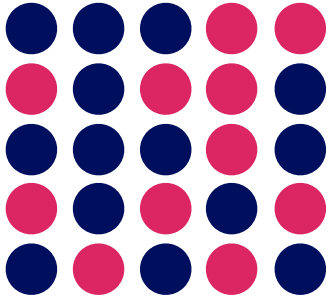
Surface populations

Surface CONFIG



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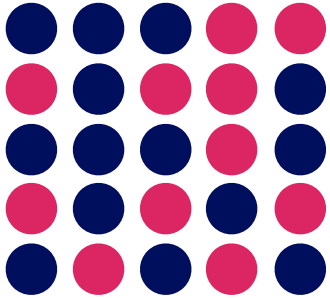


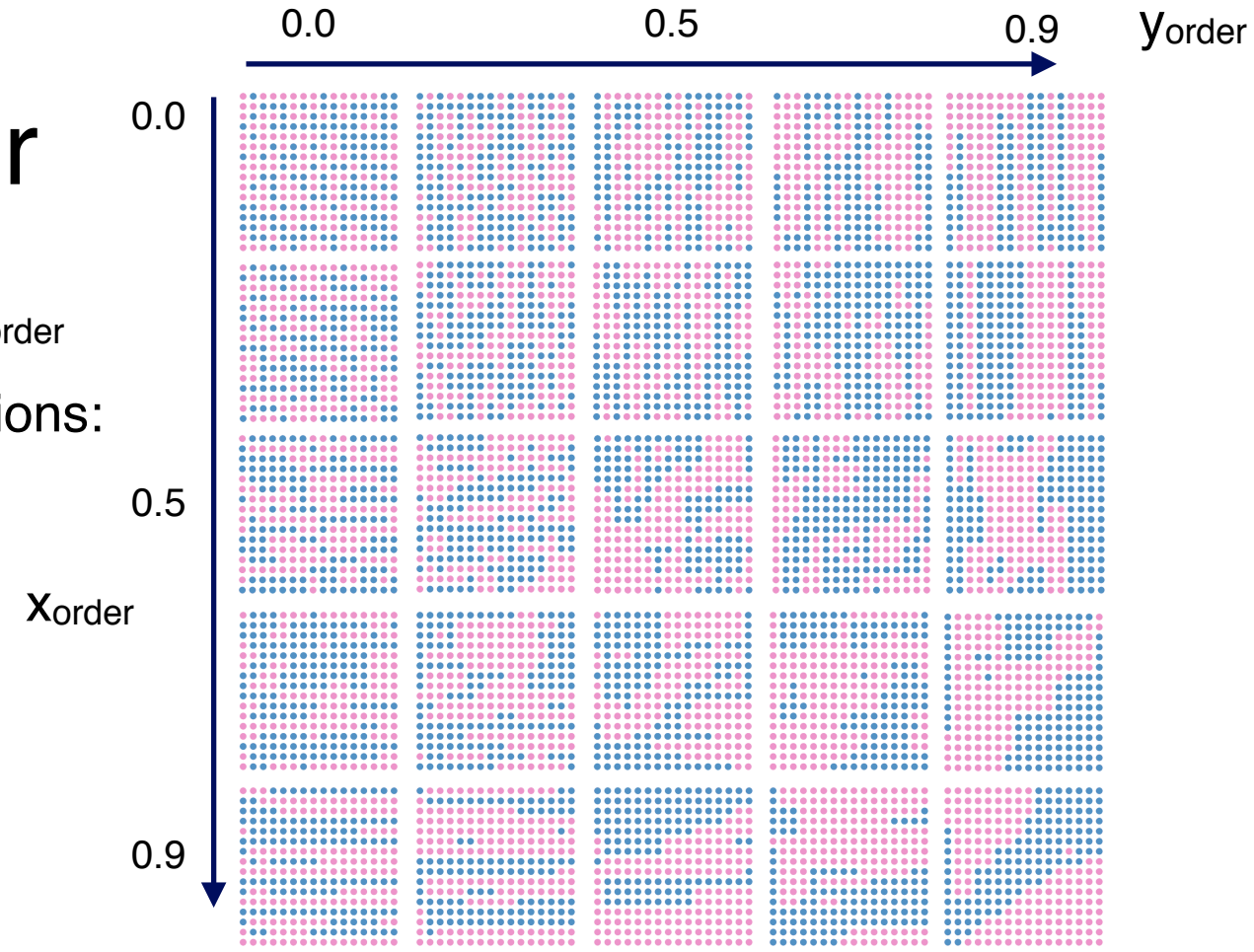
Order Parameter

Varying x_{order} and y_{order}

Target surface fractions:

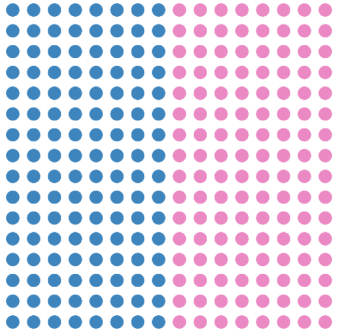
$\chi_a = 0.5$

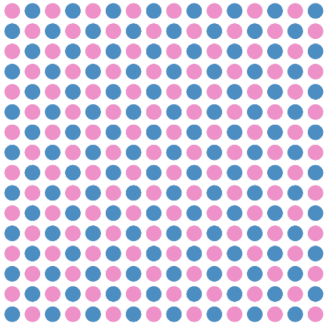
$\chi_b = 0.5$



Domain Number

Various ways of achieving full order:





Varying number of domains

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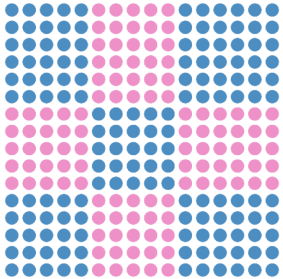
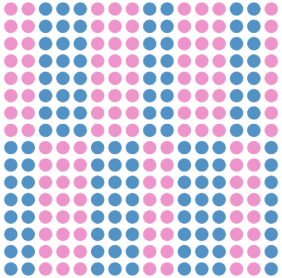
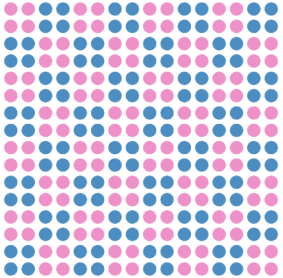
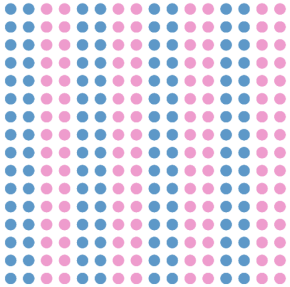




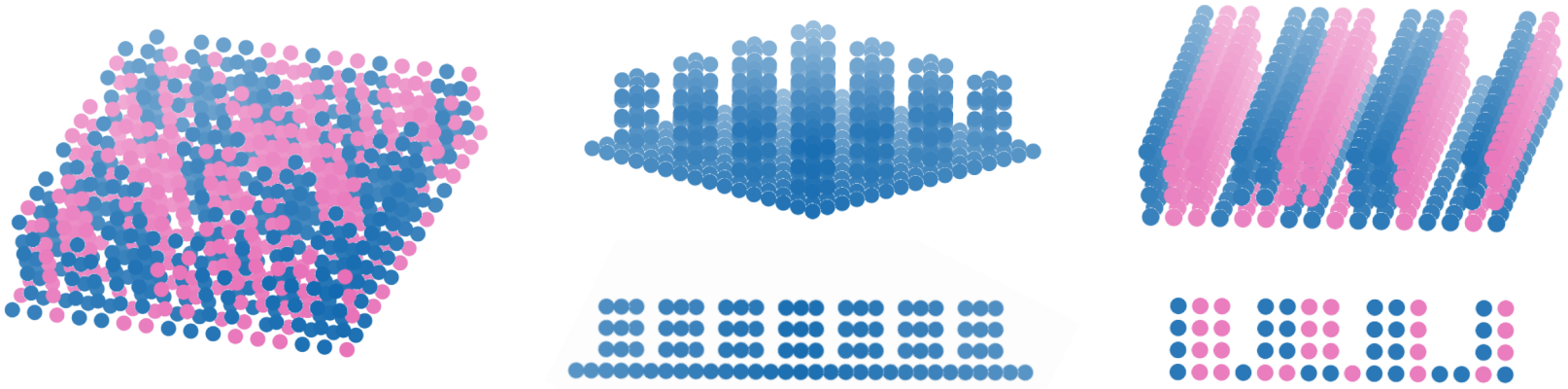




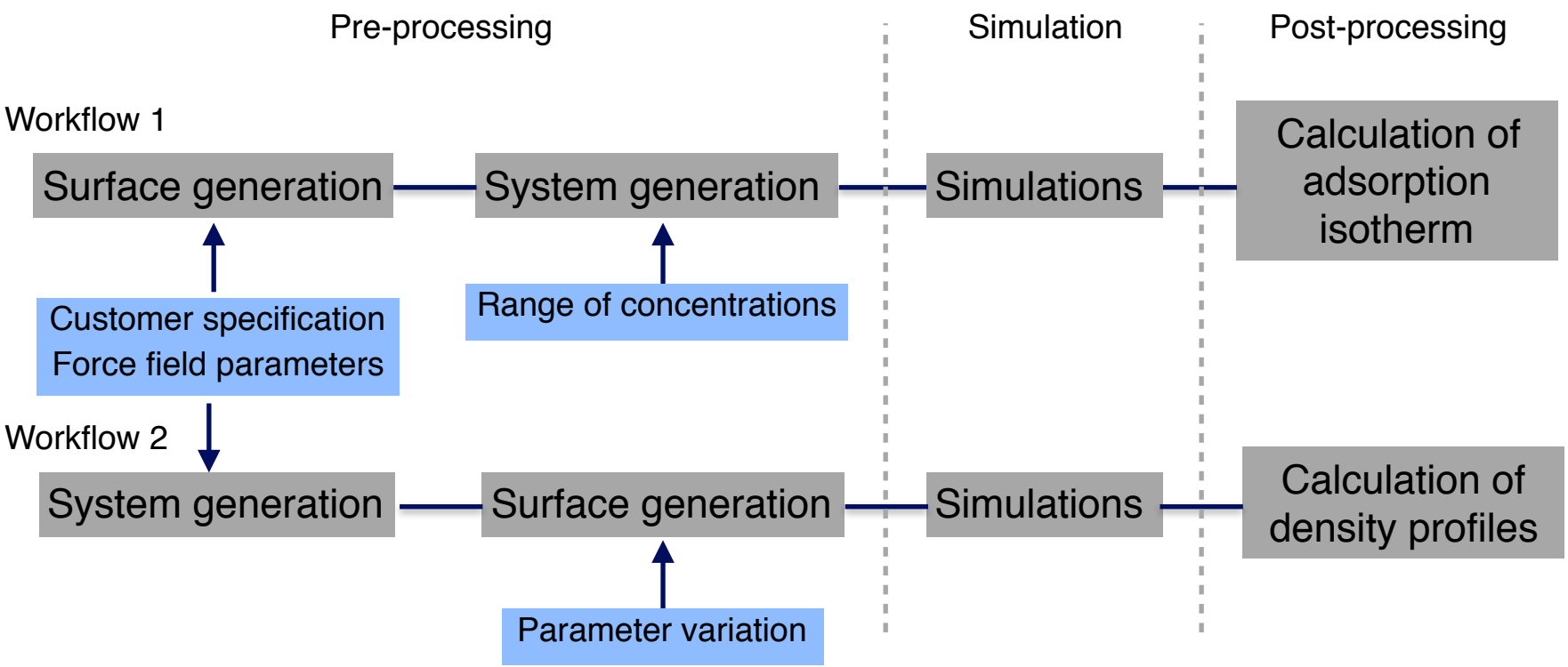
Physical Defects

Introduced a ‘vacancy’ type

Surface depth parameter allows trenches to be defined with a bottom layer



Experiment workflows



Conclusions

- Created a toolbox to generate surfaces with a range of heterogeneity
  - Inclusion of chemical and physical defects
  - Easy application of base parameters
  - Simple user text-based input
  - Produces input files for DL MESO
- Automated workflows to fit inside flow for surface investigation

Future Scope

- Code development:
  - Increase range of surface assignment schemes
  - Multiple layers
- Experiment workflows:
  - Post-processing calculation of adsorption isotherms and other observables
  - Pre-processing of inputs
- Customer use of the workflows to investigate surface adsorption

Acknowledgments

Chemistry Team

Breanndan O'Conchuir
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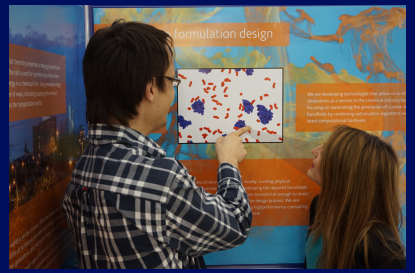
Tom Ridley

STFC

David Bray
Rick Anderson

IBM Internships



- 3 month research project
- 4 IBM Research groups: Chemistry, Life Science, Engineering and Enabling Technologies
- 3 cycles each year: Spring, Summer and Autumn
- For further information or to submit a CV and Cover letter: breanndan.conchuir@ibm.com

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