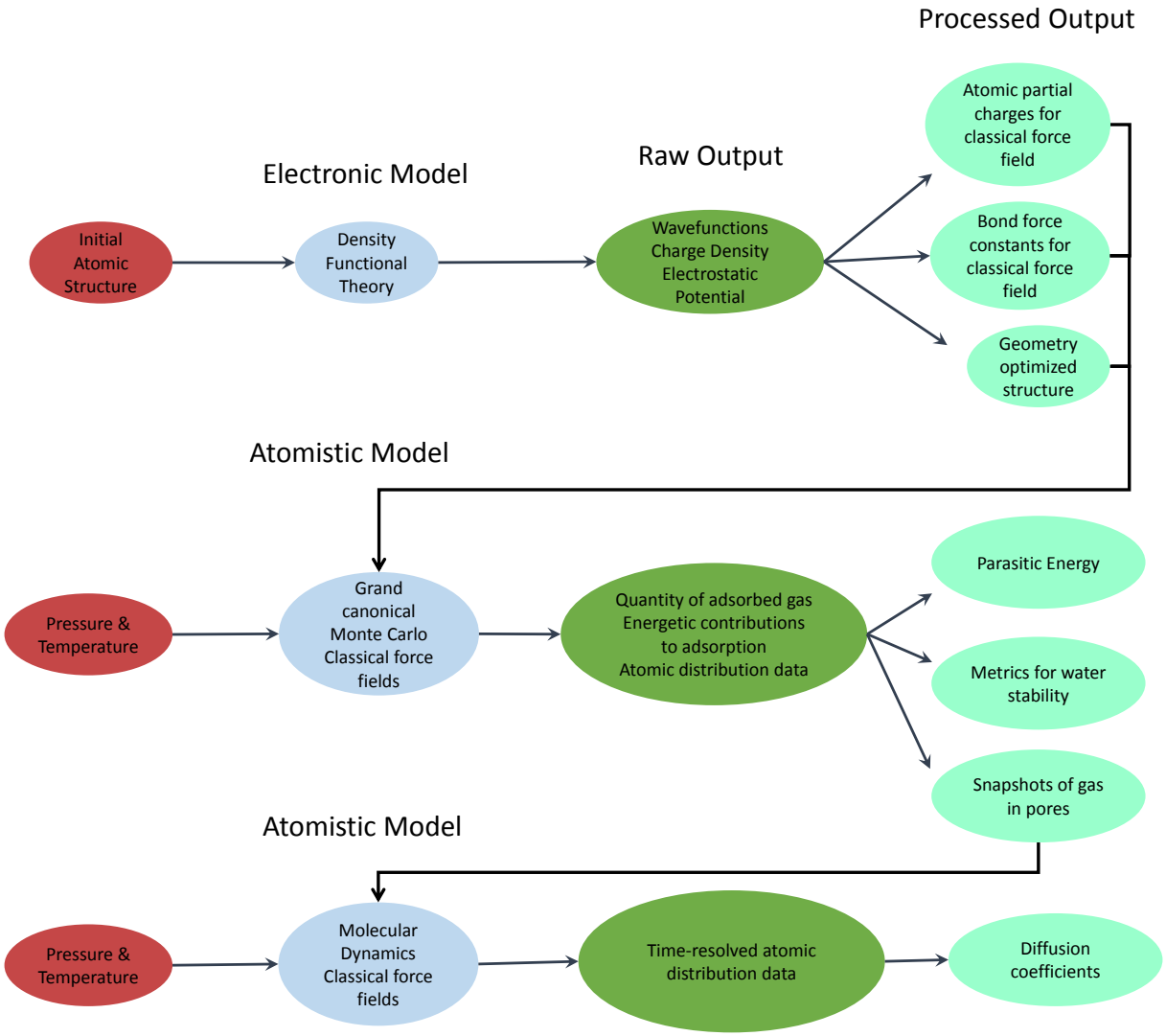
Please fill in the boxes and delete all explanatory text in italics

MODA for <*user-case*>
***Simulated* in project <GENESIS>**

OVERVIEW of the SIMULATION			
1	**USER CASE**	*Metal Organic Frameworks are porous materials that exhibit unique void space. These materials can be tuned for a range of applications by adjusting the chemical composition of the lattice structure.* *Due to this feature, several of these materials demonstrate selective preference for adsorbing CO_2 over other, less environmentally harmful gases. We aim to systematically study the performance of these materials towards the selective capture of CO_2 and related separations in dry and humid conditions.*	
2	**CHAIN OF MODELS**	**MODEL 1**	*Electronic*
		MODEL 2	*Atomistic*
		DATA-BASED MODEL	*None*
3	**PUBLICATION PEER-REVIEWING THE DATA**	*Witman, M.; Ling, S.; Anderson, S.; Tong, L.; Stylianou, K. C.; Slater, B.; Smit, B.; Haranczyk, M. In silico design and screening of hypothetical MOF-74 analogs and their experimental synthesis.* ***Chem. Sci.****, 2016,* ***7****, 6263-6272*	
4	**ACCESS CONDITIONS**	*Open source*	
5	**WORKFLOW AND ITS RATIONALE**	*While there are many modellers that estimate gas adsorption using simplified equations such as the Langmuir and Brunauer-Emmet-Teller models, they provide little insight into the atomic features of adsorption in nanoporous materials.* *The workflow provided is designed to obtain the most accurate atomic-level detail of gas adsorption (CO_2, H_2O, N_2, etc.) in these materials given the current constraints in computational power. The purpose for this is to extract atomic-level design criteria with respect to pore shape and chemistry in order to rapidly develop the next-generation of MOF materials.* *The gas adsorption calculations require millions of MOF-gas energy calculations, negating the possibility of using electronic-structure based calculations. Thus we turn to classical force fields to do the 'heavy lifting'. Because a force field is only as good as its parameters, we intend to extract these parameters from preliminary electronic structure calculations, such that we obtain a close match between DFT and classical energies.*	

Workflow picture



Each physics-based model used in this simulation is to be documented in four chapters:

1. *Aspect of the User Case or system simulated with this model*
2. *Model: Please make sure the notions Physics Equation and Materials Relation are properly understood.*
    - *Tightly coupled models can be written up collectively in one set of four tables. To solve tightly coupled PE one matrix is set up and solved in one go.*
    - *For continuum models the PE is often the conservation equations coded up in bought software packages.*
    - *Often the MR is established by the modeller.*
3. *Computational aspects include also a documentation of how the user case specifications are translated into computer language.*
4. *Post processing documents how the raw output of one simulation is processed into input for the next simulation. This information given under 4.1 in the first model will be the same as the "simulated input" information under 2.4 for the next model. This is the essence of model inter-operability!*
5. *Pre-processing before the first model can be depicted in pink as it is considered to be part of the user-case.*

Each data-based model in this simulation is to be documented in three chapters:

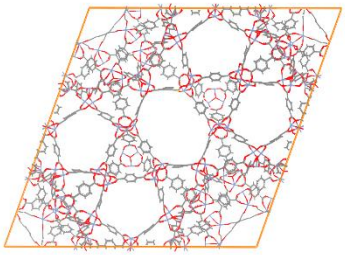
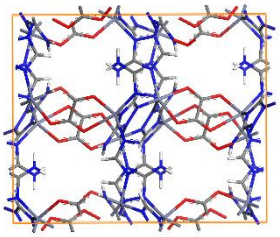
1. *Aspect of the User Case or system simulated with this data-based model*
2. *Data-based Model*
3. *Computational detail of the datamining operation*

MODA

Physics-based Model

MODEL 1

<Please name the single materials model in the chain you will now document in 4 chapters>

1	ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED	
1.1	**ASPECT OF THE USER CASE TO BE SIMULATED**	*The MOF materials are provided as an irreducible repeating unit representation of their crystalline structure (much like a single tile can represent a tiled floor). This consists of a bounding 3-D cube (or parallelpiped), with the atomic data resolved inside. This is called the 'unit cell' of the crystal, and if one were to replicate the unit cell in all directions many times, one would recover the MOFs macroscopic crystal.* *The chemical composition will be preprocessed to select the appropriate pseudopotentials for the electronic structure calculations.*
1.2	**MATERIAL**	*MOFs contain both organic (C,H,N,O,S) and inorganic (Cu,Mg,etc.) components, each with its own unique composition.*
1.3	**GEOMETRY**	*As with their chemistry, MOFs can vary in size from ~1 nm^3 to 10 nm^3. Their atom count and density exibit an extreme range, and hold the record for lowest density in nanoporous materials.*  *MIL-101 – consisting of Cr, C, H, and O atoms. The unit cell dimensions are ~9 nm^3*  *CALF-15 – consisting of Zn, N, C, H, and O atoms. The unit cell dimensions are ~1.5 nm^3*
1.4	**TIME LAPSE**	*1 month*
1.5	**MANUFACTURING PROCESS OR IN-SERVICE CONDITIONS**	*Not applicable (the electronic structure calculations will be used for downstream properties computed with atomistic models)*
1.6	**PUBLICATION ON THIS DATA**	*Sumida, K.; Rogow, D. L.; Mason, J. a; McDonald, T. M.; Bloch, E. D.; Herm, Z. R.; Bae, T.-H.; Long, J. R. Chem. Rev. 2012, 112 (2), 724.*

2	GENERIC PHYSICS OF THE MODEL EQUATION		
2.0	MODEL TYPE AND NAME	*Quantum mechanics DFT*	
2.1	MODEL ENTITY	*electrons*	
2.2	MODEL PHYSICS/ CHEMISTRY EQUATION PE	**Equation**	*Kohn-Sham (derived from Schrödinger)*
		Physical quantities	*Wavefunction, charge density, electrostatic potential*
2.3	MATERIALS RELATIONS	**Relation**	*Geometric and electrostatic environment*
		Physical quantities/ descriptors for each MR	*Electrostatic potential, atomic distributions*
2.4	SIMULATED INPUT		

3	SOLVER AND COMPUTATIONAL TRANSLATION OF THE SPECIFICATIONS		
3.1	NUMERICAL SOLVER	*Iterative (self-consistent) matrix diagonalization*	
3.2	SOFTWARE TOOL	*Quantum Espresso*	
3.3	TIME STEP	*Not applicable*	
3.4	COMPUTATIONAL REPRESENTATION	PHYSICS EQUATION, MATERIAL RELATIONS, MATERIAL	*Not applicable*
3.5	COMPUTATIONAL BOUNDARY CONDITIONS	*Unit cell boundary conditions to simulate an infinite domain.*	
3.6	ADDITIONAL SOLVER PARAMETERS	• *Electronic cut-off ~80 Ry* • *Electronic convergence criteria 10^{-3} Ry*	

Post processing

The "raw output" calculated by the model consists per definition of values for the physics variable in the PE(s). This variable is already specified in 2.2 and this raw output will appear in your dark green circle in the workflow picture.

This raw output is often processed

- *to calculate values for physics variables for different entities of the next model. E.g. the output can be homogenised for larger volumes*
- *in the form of a MR for the next model*
- *into a Descriptor Rule that is the final output of the total simulation.*

This processed output will appear in your light green circle in the workflow picture and also in 2.4 of the next model (if there is one).

The methodology (often including physics) used to do this post processing calculation is to be documented in 4.2.

4	POST PROCESSING	
4.1	THE PROCESSED OUTPUT	*The partial charges computed in this post processing will be used as classical force field parameters in the atomic simulations (next model in chain).*
4.2	METHODOLOGIES	*A linear solver, which provides a set of classical coulombic charges (centered on each atom of a MOF) that best reproduce the quantum electrostatic potential defined on a volumetric grid.*
4.3	MARGIN OF ERROR	*In reproducing the quantum electrostatic potential with classical point charges, we are introducing a roughly 10% error in the potential. Please keep in mind that this is, to-date, the most accurate representation of the electronic electrostatic potential one can obtain in a classical simulation.*

MODA

Physics-based Model

MODEL 2

Grand Canonical Monte Carlo model.

1	ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED	
1.1	**ASPECT OF THE USER CASE TO BE SIMULATED**	*A thermodynamic state point is defined that represents a specific industrial processing condition (i.e. adsorption and regeneration of a material will be performed at 298 Kelvin and 313 Kelvin, respectively). This will be used to evaluate if the material can capture and release a significant quantity of purified CO_2 using mild thermodynamic conditions.*
1.2	**MATERIAL**	*See above*
1.3	**GEOMETRY**	*See above*
1.4	**TIME LAPSE**	*2 months*
1.5	**MANUFACTURING PROCESS OR IN-SERVICE CONDITIONS**	*Temperatures of 298 – 313 Kelvin, pressure ranges from 0 – 2 bar. Typical conditions of exhaust plumes from coal-fired power plants.*
1.6	**PUBLICATION ON THIS DATA**	*Samanta, A.; Zhao, A.; Shimizu, G. K. H.; Sarkar, P.; Gupta, R. Ind. Eng. Chem. Res. 2012, 51 (4), 1438.*

2	GENERIC PHYSICS OF THE MODEL EQUATION		
2.0	MODEL TYPE AND NAME	*Grand canonical Monte Carlo*	
2.1	MODEL ENTITY	*Atoms*	
2.2	MODEL PHYSICS/ CHEMISTRY EQUATION PE	**Equation**	*Newtonian classical mechanics*
		Physical quantities	*Energy, partition function, Boltzmann weighted averages.*
2.3	MATERIALS RELATIONS	**Relation**	*Observed adsorption quantities of CO_2, H_2O and N_2 gas in each material*
		Physical quantities/ descriptors for each MR	*Boltzmann averaged distribution of particles at a given temperature & pressure. Specific and averaged gas particle distributions in the pores.* *The averaged heats of adsorption (energy) for each gas particle.*
2.4	SIMULATED INPUT	*The input is provided from the DFT structure in the previous model. In addition the force field parameters provided by the DFT calculations are used as input into the classical model used in this step.*	

3	SOLVER AND COMPUTATIONAL TRANSLATION OF THE SPECIFICATIONS		
3.1	NUMERICAL SOLVER	*Monte Carlo*	
3.2	SOFTWARE TOOL	*RASPA*	
3.3	TIME STEP	*Not applicable*	
3.4	COMPUTATIONAL REPRESENTATION	PHYSICS EQUATION, MATERIAL RELATIONS, MATERIAL	
3.5	COMPUTATIONAL BOUNDARY CONDITIONS	*A multiple of the unit cell (typically) to simulate an infinite domain.*	
3.6	ADDITIONAL SOLVER PARAMETERS	• *short-range interaction cutoffs of 1.3 nm,* • *roughly 20 million Monte Carlo steps*	

Post processing

The "raw output" calculated by the model consists per definition of values for the physics variable in the PE(s). This variable is already specified in 2.2 and this raw output will appear in your dark green circle in the workflow picture.

This raw output is often processed

- *to calculate values for physics variables for different entities of the next model. E.g. the output can be homogenised for larger volumes*
- *in the form of a MR for the next model*
- *into a Descriptor Rule that is the final output of the total simulation.*

This processed output will appear in your light green circle in the workflow picture and also in 2.4 of the next model (if there is one).

The methodology (often including physics) used to do this post processing calculation is to be documented in 4.2.

4	POST PROCESSING	
4.1	THE PROCESSED OUTPUT	*Atomic snap shots – for use in diffusion calculations in the molecular dynamics chain.* *Adsorption per unit mass (and volume) of the material will be computed, this can be directly compared with macroscopic experimental data.*
4.2	METHODOLOGIES	*Thermodynamics quantites computed at this step require the conversion of mass from atomic units to macroscopic units (grams or kilograms)*
4.3	MARGIN OF ERROR	*The error typically observed in adsorption quantities is on the order of 10% (over-estimate). The reasons for this are mainly because we simulate a perfect crystal, while in reality defects and impurities in the pores of these materials can hinder the adsorption performance.*

MODA

Physics-based Model

MODEL 1
Molecular Dynamics model

1	ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED	
1.1	ASPECT OF THE USER CASE TO BE SIMULATED	*A thermodynamic state point is defined that represents a specific industrial processing condition (i.e. adsorption and regeneration of a material will be performed at 298 Kelvin and 313 Kelvin, respectively). This will be used to evaluate the diffusion behaviour of gases such as CO_2, N_2, and H_2O.*
1.2	MATERIAL	*See above*
1.3	GEOMETRY	*See above*
1.4	TIME LAPSE	*2 months*
1.5	MANUFACTURING PROCESS OR IN-SERVICE CONDITIONS	*Temperatures of 298 – 313 Kelvin, pressure ranges from 0 – 2 bar. Typical conditions of exhaust plumes from coal-fired power plants.*
1.6	PUBLICATION ON THIS DATA	*Samanta, A.; Zhao, A.; Shimizu, G. K. H.; Sarkar, P.; Gupta, R. Ind. Eng. Chem. Res. 2012, 51 (4), 1438.*

2	GENERIC PHYSICS OF THE MODEL EQUATION		
2.0	MODEL TYPE AND NAME	*Molecular Dynamics*	
2.1	MODEL ENTITY	*Atoms*	
2.2	MODEL PHYSICS/ CHEMISTRY EQUATION PE	**Equation**	*Newton's classical equations of motion*
		Physical quantities	*Force, velocity, momentum*
2.3	MATERIALS RELATIONS	**Relation**	*Measurements of diffusion of gas particles in the MOFs.*
		Physical quantities/ descriptors for each MR	*Mean squared displacement of gas particles.*
2.4	SIMULATED INPUT	*Snapshots of MOFs loaded with CO_2 N_2 and H_2O will be obtained from grand canonical Monte Carlo simulations from the previous chain. This will guaranty that the quantity of gas loaded in these pores will be at the correct thermodynamic conditions of the process. The specific loadings of gas particles can impede or enhance motion, thus affecting gas diffusion in these materials.*	

3	SOLVER AND COMPUTATIONAL TRANSLATION OF THE SPECIFICATIONS		
3.1	NUMERICAL SOLVER	*Molecular dynamics (finite difference time integration)*	
3.2	SOFTWARE TOOL	*LAMMPS*	
3.3	TIME STEP	*1 femtosecond*	
3.4	COMPUTATIONAL REPRESENTATION	PHYSICS EQUATION, MATERIAL RELATIONS, MATERIAL	
3.5	COMPUTATIONAL BOUNDARY CONDITIONS	*Multiple of the unit cell to simulate an infinite domain.*	
3.6	ADDITIONAL SOLVER PARAMETERS	• *1.3 nm short range cutoffs,* • *Typically 1 nanosecond simulation time* • *Leapfrog integrator*	

Post processing

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The methodology (often including physics) used to do this post processing calculation is to be documented in 4.2.

4	POST PROCESSING	
4.1	THE PROCESSED OUTPUT	*Diffusion coefficients for CO_2 N_2 and H_2O.*
4.2	METHODOLOGIES	*The diffusion coefficients are determined from a linear fit to the mean-squared displacement of gas particles measured during the simulation.*
4.3	MARGIN OF ERROR	*We can expect a 10% margin of error in these calculations as well. As with the previous model, this is due to the fact that the material we simulate is free from defects and impurities.*