Molecular Modelling and Machine Learning-Based Prediction of MOF Materials for Hydrogen Adsorption

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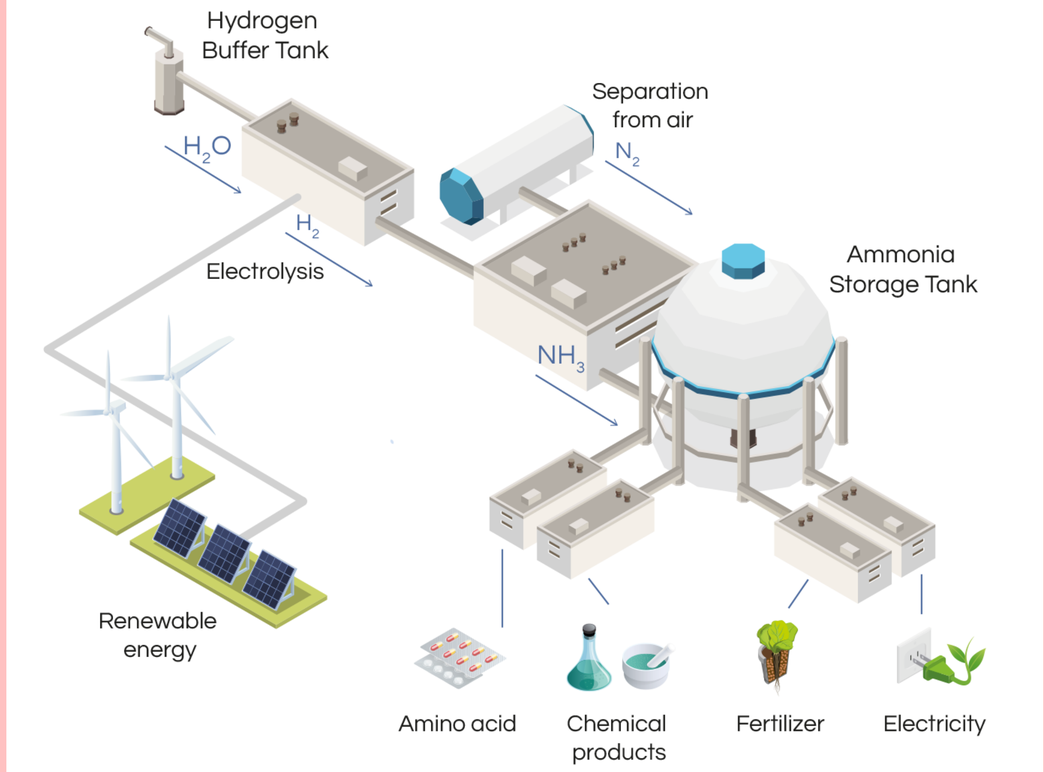
HySTrAm[1]: Hydrogen Storage and Transport using Ammonia

- Green Ammonia production plant based on an improved Haber–Bosch process
  $N_2 + 3\ H_2 \rightarrow 2\ NH_3$ (ΔH = -91.8 kj/mol)
- A short-term hydrogen vessel with new ultraporous material identified and optimized through machine learning technology

Specific objectives

- Development of functional catalyst/sorbent materials for ammonia synthesis;
- Development of new ultra-porous materials with high H_2 capacity;
- Realisation of a lightweight composite vessel for physical-adsorption hydrogen storage;
- Design, construction, optimisation and demonstration of dynamically operated packed bed reactors for ammonia synthesis;
- Demonstration of the overall HySTrAm solution at TRL5;
- Validation of a business case.



Machine Learning based approach to the modelling of short-term hydrogen storage

MOF Descriptors

MOF Chemistry | **Pore Geometry**

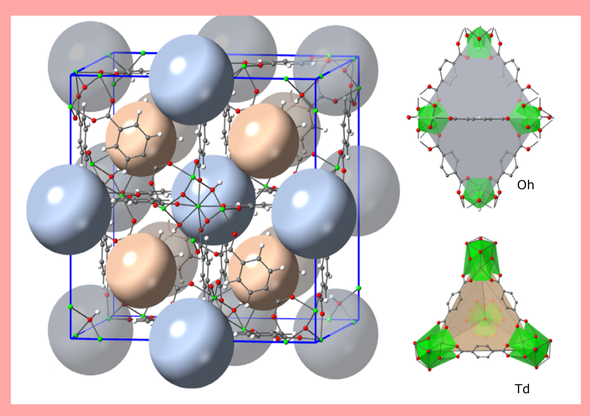
Revised Autocorrelations (RACs)[2]: discrete correlations between heuristic atomic properties of atoms on a graph.

$$ {}^{start}_{scopt}P_d^{diff} = \sum_i^{start} \sum_j^{scope} (P_i - P_j)\delta d_{i,j},d $$

$$ {}^{start}_{scopt}P_d^{prod} = \sum_i^{start} \sum_j^{scope} (P_i\ P_j)\delta d_{i,j},d $$

Molecular Graph Modification Strategy:
Atoms → Node (Z, I, T, S, χ)
Bonds → Edges (BO, d, I)
Molecule → Graph (q, M_W, N_{atoms}, μ, α)

AABBA (Atom Atom Bond Bond Atom) Code[3,4]



UiO-66 Pore structure

Largest included sphere,
Largest free sphere,
Crystal density,
Volumetric surface area,
Gravimetric surface area,
Pore volume, *etc.*

Calculated using Zeo++ software

H_2 adsorption Capacity

- Grant-Canonical Monte Carlo (GCMC) simulations to obtain the adsorption isotherms
- μVT ensemble (number of particles, P and E are allowed to fluctuate)
- Atoms in MOF are held fixed at their crystallographic positions.
- Force field to describe interactions (UFF+Dreiding Lennard-Johns potential)
- H_2: 3-site rigid model
- Moves particles stochastically; generate configurations according to Boltzmann probability
- RASPA software package

Machine Learning Study

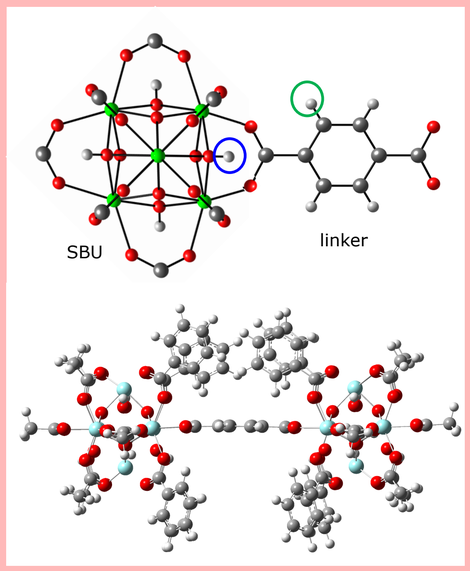
A data-driven approach can significantly accelerate the **materials screening and prediction** of new materials for efficient hydrogen storage. By training machine learning models such as **graph neural networks (GNNs)** on the gas adsorption data using geometry and graph-based MOF representations, we aim to identify efficient MOF materials for high **hydrogen adsorption capacity at low pressure (<100 bar) and room temperature conditions**.

Periodic and Cluster Model

Electronic structure calculation of potential MOF candidates

MOF-H_2 interaction study using UiO-66 sbu/linker functionalized variants

DFT calculations using CP2K and Turbomole softwares



References

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3. David Balcells and co-workers; *Digital Discovery*, 2023,2, 618-633
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