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Establishing a Quantitative Structure-Property Relationship for Porous Carbons in CO_2 Capture Applications

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CO_2 capture is an important technology for climate change mitigation.

Climate change is caused by anthropogenic CO_2 emissions, most of which originate from the energy and industrial sector [1]. Thus, one of the main ways to mitigate climate change is by preventing emissions from those sectors. To do this, we can ***capture CO_2 from the exhaust gases of conventional power plants and factories.***

Porous carbons can make CO_2 capture cheaper and easier.

Typically, CO_2 is captured by absorbing them on liquid amines, but that method has *low working capacity (<0.45 mmol/g)* [2] and *uses a large amount of heat* for desorption, which also leads to the *degradation* of the amines. One solution is to use physical adsorption on high surface area **porous carbons** instead. This method has the advantages of **needing less energy for desorption, being more stable, and having larger capture capacities (>3 mmol/g)**.

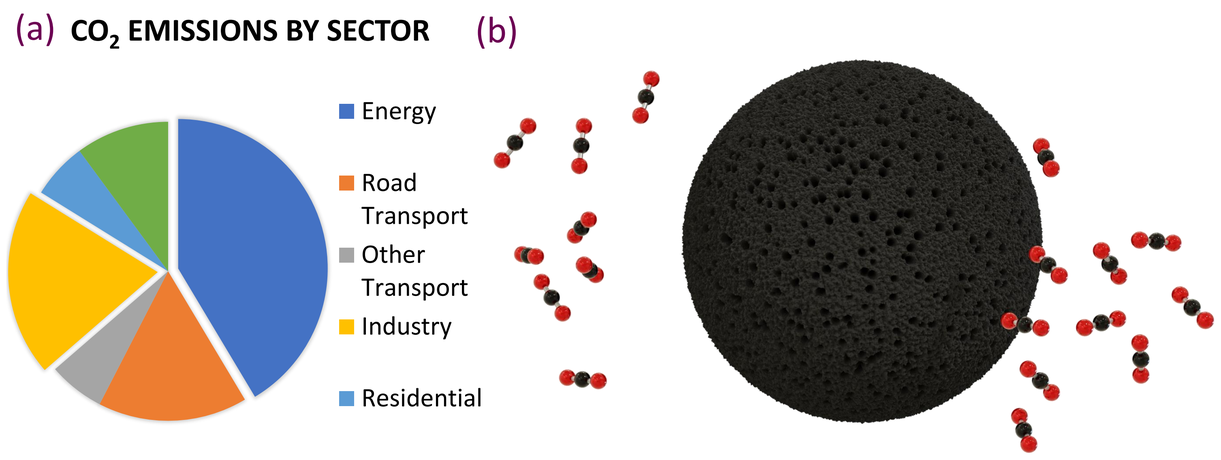

Figure 1. (a) A pie chart based on data from the International Energy Agency [1] showing CO_2 emissions by sector. (b) Schematic illustration of CO_2 adsorption on carbon

The Research Challenge

Determining the variables that make a carbon material suitable for CO_2 capture is **hard**:

- Low sample size per study
- Non-random samples
- Multicollinearity within study

For example, in our previous work [3]:

N-doped Carbon Foam (NCF)
- **>2600 m²/g**
- **Micropores only**
- **Nitrogen-doped (8 at%)**

Carbon Foam (CF)
- **>3400 m²/g**
- **Micro-mesoporous**
- **No doping**

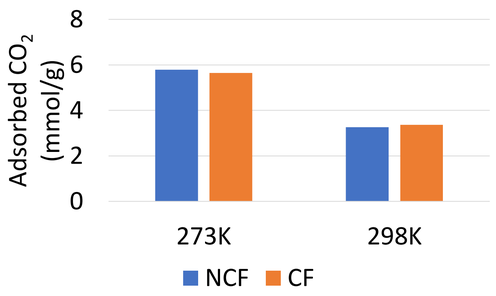

Figure 2. A bar plot comparing the CO_2 capture capacity of NCF and CF at 100 kPa & two different temperatures.

Very different properties but *the end results look the same*. Did NCF's doping help it compensate for the lower surface area, or was it the pore size distribution?
How do we know what variables are important?

No clarity on variable importance →
No rational design →
Trial-and-error → Slow research!

Our solution? Gather a vast amount of data from the scientific literature and develop models that can *predict the CO_2 adsorption isotherm* of each material based on their porosity and chemical composition using **statistics**. In other words:

Establish a clear, quantitative structure-property relationship.

Data Collection

To date, we collected a dataset from:
- **19 studies**
- **74 carbon samples**
- **~3900 data points at different pressures**

The input variables: chemical composition (wt%), pore size distribution, pore volume, and surface area.

The output variable: excess CO_2 adsorbed amount, in mmol/g.

Results and Discussion

We are at the stage of model and variable selection.

- Try ***different isotherm equations*** (equations that describe adsorbed amounts of CO_2 at different pressures)
- Try ***different combinations of the input*** variables to predict the parameters of those equations using **a generalized Bayesian linear model**

Here is our best model-variables combination so far:

Model based on Dubinin-Astakhov Micropore Filling Equation [4]

$$Q_{ads} \sim Lognormal(\mu, \sigma)$$

$$\mu = \log Q_{max} - (A/E)^n$$

$$A = RT \log \frac{P}{P_0}$$

$$\log Q_{max} = \sum x_i \alpha_i$$

$$\log E = \sum y_i \beta_i$$

$$\log n = \sum z_i \gamma_i$$

- Q_{ads} is the amount of CO_2 adsorbed on the carbon
- R is the universal gas constant
- T is temperature
- P is pressure and P_0 is the saturation pressure of CO_2 at temperature T
- Q_{max} is the maximum adsorption capacity of the carbon materials
- α, β, and γ are linear regression coefficients
- x_i includes N wt% & O wt% and their squares, surface area divided by pore size, and temperature
- y_i includes N wt% & O wt% and their squares, percentage of ultramicropores and micropores, and temperature
- z_i includes percentage of ultramicropores and micropores, and temperature

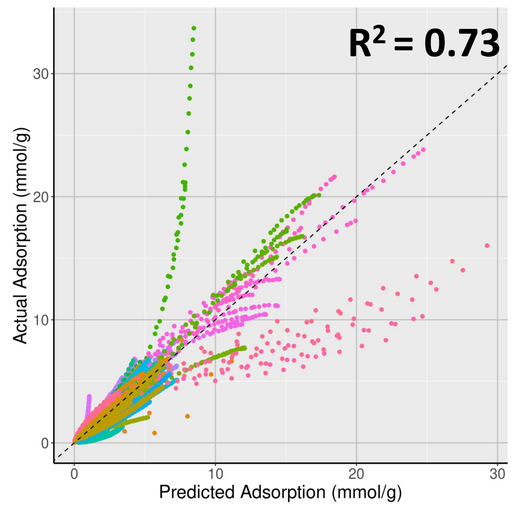

Figure 3. A parity plot comparing model predictions and reality in leave-one-study-out cross validation.

The model only uses linear regression, so the accuracy is not perfect yet.

Future Work

Improve accuracy using non-parametric fitting

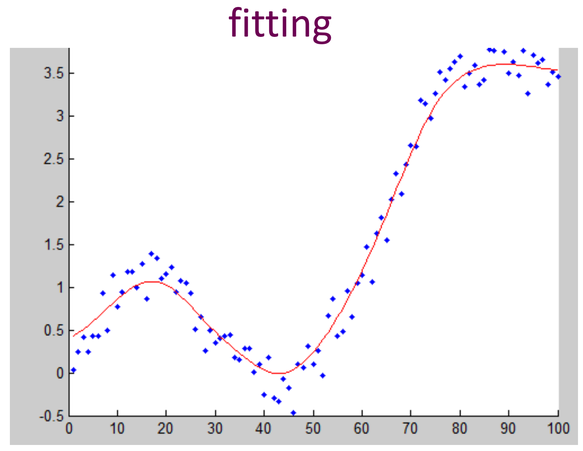

Figure 4. Non-parametric models like Gaussian Process regression can automatically find a best fit for a non-linear relationship. (Image Source: Yunyoungmok, CC BY-SA 3.0, via Wikimedia Commons)

References

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