Computational Investigation on CO$_2$ Adsorption in Titanium Carbide-Derived Carbons with Residual Titanium

Scientific Achievement
A new approach for modeling carbide-derived carbons (CDCs) with residual metal has been developed.

Significance and Impact
To date, no modeling of CDCs with residual metal have been developed. This tool can now provide important insights into the way in which residual titanium within the CDCs influences structural composition and gas adsorption.

Research Details
– Modified atomistic structures based on a silicon carbide derived-carbon (SiC-CDC) model to generate a model for studying titanium carbide derived-carbons.
– SiC-CDC structure modified by (i) removing carbon, (ii) adding carbon and (iii) adding titanium.
– TiC-CDC system with residual titanium is modeled as a weighted combination of pure carbon CDC structures, CDC structures with titanium and a TiC crystalline structure.
– TiC-CDC model is able to produce both structural properties and adsorption isotherms in agreement with experimental data.
– Analysis of modeling results suggests that titanium accessible to CO$_2$ at the transitional interface between bulk TiC and fully etched CDC may provide significant interaction sites for CO$_2$ and lead to more efficient gas interactions.

Modifications of base SiC-CDC model employed to generate TiC-CDC model

SiC-CDC model used as the original structural model (a)

Three models studied, based off of modifications of SiC-CDC base model:

- **Type 1** - removal of carbon atoms from the structure (b)
- **Type 2** - addition of carbon atoms to the structure (c)
- **Type 3** - addition of Ti atoms or clusters to the structure (d)

Bulk crystalline TiC model also used to reflect residual TiC remaining in etched materials.
Distinct Phases of Etched Carbides Requires Weighted Combination of Different Systems to Model TiC-CDC

Type 3 (TiC-CDC) model alone does not match experimental data → assume 3 regions of TiC-CDC system:

1. Unreacted TiC (bulk TiC)
2. Fully etched CDC (Type 1, Type 2)
3. Interfacial region (Type 3)
Weighted Models Produce Structural Properties and Adsorption Isotherms in Agreement with Experimental Data

The fraction of different models in the systems accurately reflects the structural differences in experimental TiC-CDC materials.
Simulation Data Allows for Extraction of Adsorption Contributions from Different TiC-CDC Regions

The interfacial region (Type 3) has higher adsorption loadings than the fully etched region (Type 1 and Type 2) per unit surface area → Suggests the interfacial region is more efficient for CO₂ gas adsorption.