Multiscale Analysis of CH_4 and CO_2 Conversion

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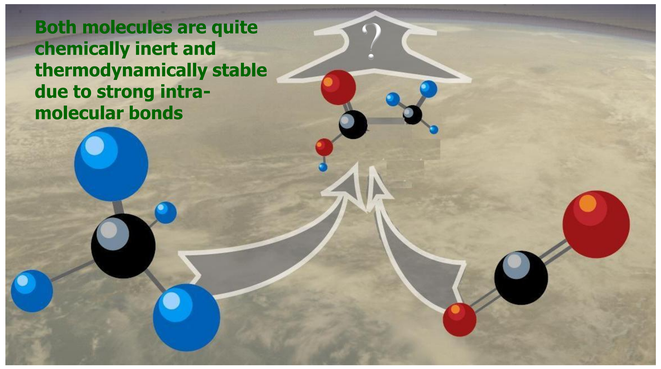
Direct conversion of CH_4 and CO_2

The goal of this research is to study direct conversion of these two greenhouse gases at mild conditions to obtain higher-value products.

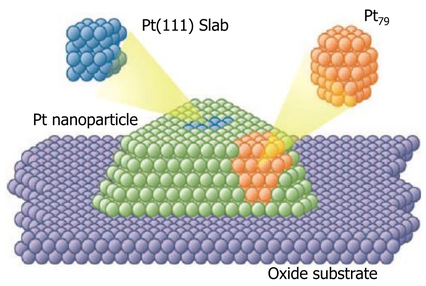


→ most of their reactions require significant energy inputs as well as properly designed catalytic systems that lower kinetic barriers in their direct conversion

• The abundance of these two gases makes them attractive raw materials for fuels and chemical synthesis

Structure sensitivity of methane activation

• It is considered that the rate limiting step is methane activation. The formation of CH_x fragments, via CH_4 dissociation, is structure-sensitive due to electronic and geometric constraints.

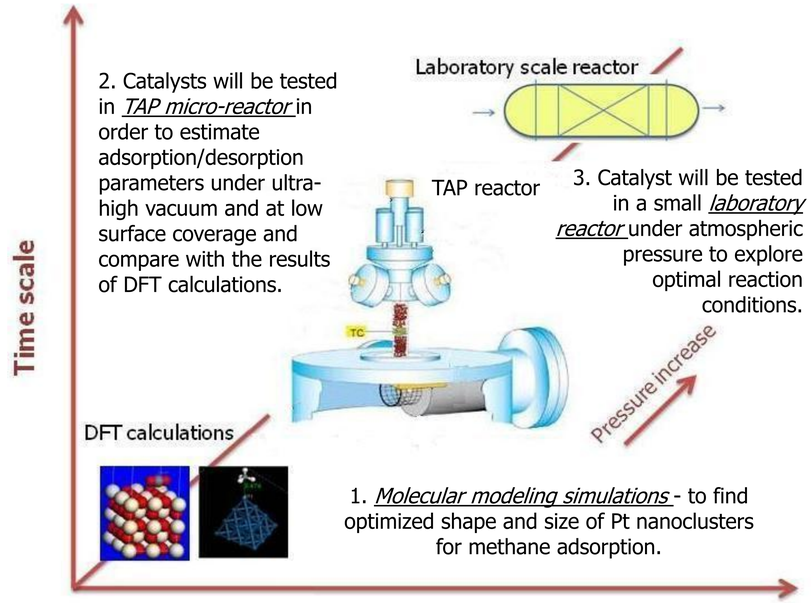


The presence of *low-coordinated sites, such as edge, corner*, and nearby sites on metal active centers has been shown to facilitate the conversion by reducing the energy barriers of every reaction step and stabilizing the reaction intermediates.

• Pt nanoparticles with cubic, tetrahedral and octahedral shapes contain numerous surface steps, edges and kinks which are expected to critically affect catalyst activity and selectivity.

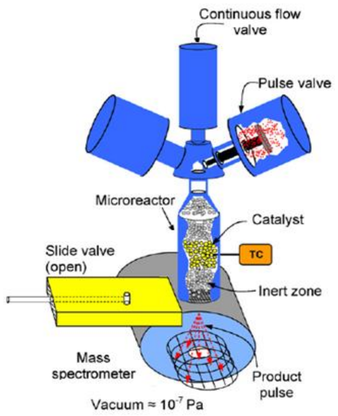
Fundamental understanding and quantification of diffusion and adsorption/desorption of the reactants on the different cluster morphologies, should help in determining the key features needed for catalyst design and process development and operation

Integrated experimental and theoretical approach



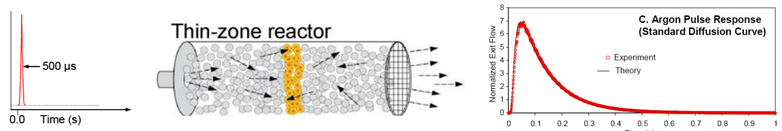
Early comparison of catalyst performance under high vacuum conditions and in an atmospheric pressure micro-reactor will lead to an improved systems approach to catalyst design.

Temporal Analysis of Product (TAP)



A narrow gas pulse (~10^{13} molecules) is injected into an evacuated TAP micro-reactor

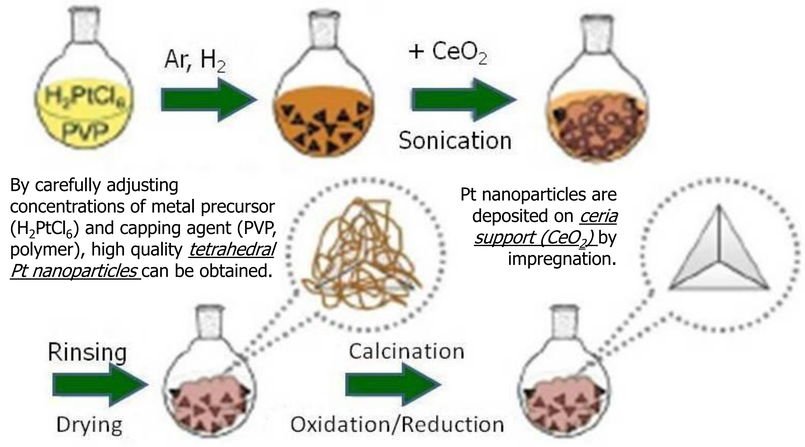
Molecules are transported through the reactor by Knudsen diffusion



The time-dependent response curves from the micro-reactor, interpreted by an appropriate theoretical model, will be used to determine the adsorption/desorption and apparent kinetic parameters which will be compared with the results from DFT calculations.

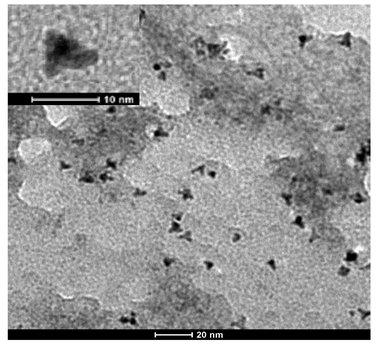
Shape-controlled synthesis

The independent control of particle size and shape during catalyst preparation could result in the design of highly selective catalysts

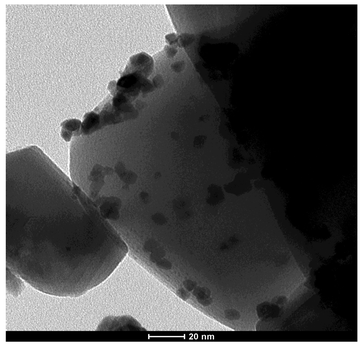


TEM images

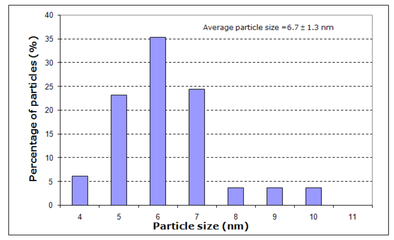
Tetrahedral Pt clusters

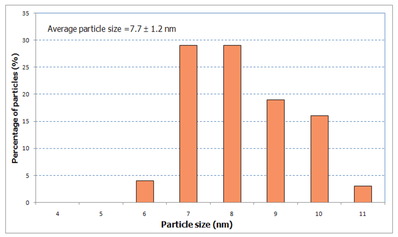


Amorphous Pt on ceria



Particle size distribution





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