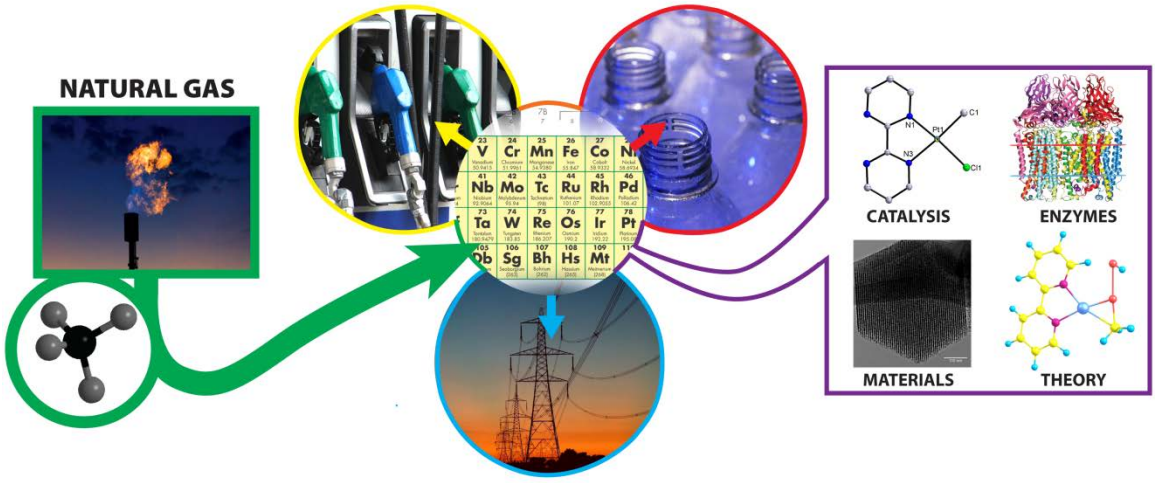
Center for Catalytic Hydrocarbon Functionalization (CCHF)
EFRC Director: T. Brent Gunnoe
Lead Institution: University of Virginia

Mission Statement*: To develop, validate, and optimize new methods to rearrange the bonds of hydrocarbons, implement enzymatic strategies into synthetic systems, and design optimal environments for catalysts that can be used to reversibly functionalize hydrocarbons, especially for more efficient use of natural gas including low temperature conversion to liquid fuels.*

Fossil fuels, the dominant energy source in the United States, are primarily composed of hydrocarbons. The current high temperature processes that convert these raw materials into useful products lead to low efficiency, excessive emissions (including carbon dioxide) and a disproportionate dependence on imported petroleum. Due to the increasing demand for energy, current fossil fuel combustion processes will not adequately meet future energy needs. To provide for a secure energy future in the United States and worldwide, new technologies that revolutionize hydrocarbon utilization, particularly methane from natural gas, will be necessary.

Catalysts for the low temperature and selective functionalization of hydrocarbons are central to the development of advanced technologies that can provide dramatic improvements in the utilization of energy resources. For example, such systems could reduce our dependence on petroleum through the use of abundant natural gas reserves as our source of liquid fuels and materials, provide cleaner and more energy efficient routes for hydrocarbon processing and lead to the development of high efficiency methane fuel cells. The Center for Catalytic Hydrocarbon Functionalization (CCHF) brings together a broad based collaborative team with the expertise to accelerate advances in fundamental aspects of catalyst technologies required for selective hydrocarbon functionalization.

A major focus of the CCHF is the development of catalysts for the selective and scalable functionalization of methane, with formation of methanol, a versatile liquid fuel, as one desirable target. Another focus is development of anodic catalysts for low temperature direct methane fuel cells, in which methane is oxidized to carbon dioxide and water at low overpotentials and high power. Research in the CCHF is organized into three subgroups: 1) the C–H Activation Subgroup, 2) the Oxygen Activation Subgroup, and 3) the Methane Fuel Cell Subgroup. The unifying element of research in the three subgroups of the CCHF is the development of molecular catalysts for the selective conversion of C–H bonds of hydrocarbons, especially methane, to C–O bonds. All three subgroups employ a combination of computational and experimental methods to rationally design new catalysts.



C–H Activation Subgroup

Research in the C–H Activation Subgroup focuses on development of molecular, single-site transition metal catalysts that activate hydrocarbons (e.g., methane) through reactions that generate a metal-carbon species, which is then functionalized to generate the alcohol or other functional products such as halogenated compounds. Key challenges in catalyst development include: minimizing inhibition and decomposition of catalysts, integration of the C–H activation and metal-carbon functionalization reactions into efficient catalytic cycles, use of dioxygen or air-recyclable oxidant, and minimizing the cost of the catalysts. In addition to designing molecular catalysts for use in solution there is also a focus on supporting these catalysts on nanoparticles.

Oxygen Activation Subgroup

The key distinction between this and the C–H activation subgroup is the underlying mechanism of catalysis. The Oxygen Activation Subgroup focuses on catalysts that activate oxygen to form a reactive metal oxo complex, which can react with hydrocarbons (e.g., methane) to generate functionalized products. Cleavage of C–H bonds by metal oxo complexes often forms a carboradical, and attenuating the lifetime of this radical is critical to selective catalysts. Multiple routes for generation of the reactive metal oxo complex are possible, and work in the CCHF is exploring different pathways, including oxygen atom transfer, electrochemical and photoelectrochemical methods.

Methane Fuel Cell Subgroup

The application of methane fuel cells for electricity generation is currently limited, and the use of methane fuel cells in vehicles is not possible. Key to enabling increased use of methane fuel cells is operation at lower temperatures (<250°C) with high efficiency. This requires the development of stable, efficient electrocatalysts with low overpotentials for the anodic oxidation of methane. Work in the CCHF combines expertise in homogeneous C–H functionalization, electrocatalysis, fuel cells and computational chemistry to design new catalysts for application in direct methane fuel cells.

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