Development of a Novel Supersonic Hybrid Non-Equilibrium Plasma Reactor for Efficient and Tunable Co-Production of Hydrogen and Value-Added Solid Carbons

With the availability of excess renewable electricity at peak hours, an environmentally benign plasma synthesis method for hydrogen and valuable carbon production is very attractive. The plasma synthesis method is free from pollutant emissions and its theoretical yield can reach 100% by using a non-equilibrium kinetic process. However, due to the poor understanding of the plasma properties and plasma chemistry in the plasma synthesis method, the yield of existing plasma reactors is far below the theoretical limit. Therefore, a novel and controlled plasma synthesis method is urgently needed to increase the yield and the carbon value. The objective of this project was to develop and optimize an innovative supersonic hybrid non-equilibrium plasma reactor for efficient and tunable co-production of hydrogen and value-added solid carbons with a negative CO₂ footprint.

Project Description:
A new plasma-chemical reactor based on the flow of products of methane decay through a supersonic nozzle was designed and constructed. The formation of the plasma zone in the critical section of the nozzle, the dynamics of the discharge development in pure methane, and methane conversion were investigated. The main characteristics of the discharge were measured by advanced laser spectroscopy methods. Experimental data on hydrogen concentration in the products of methane decomposition in plasma were obtained, and the energy efficiency of this process was analyzed. Analysis was carried out on the solid products of methane decomposition in plasma by scanning electron microscope imaging and energy dispersive X-ray spectroscopy. Data on the size of solid carbon particles formed in the process of methane decomposition and their composition were also obtained.

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$193,000

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Technology Line:
Fuel Cell Technology/Natural Gas Conversion to Hydrogen and Solid Carbon Products

ns-pulse discharge modeling. Methane, U = 2 kV, P = 410 Torr, h = 1.3 mm.
f = 5, 13 and 20 ns.
Accomplishments:
A new supersonic non-equilibrium plasma reactor for controlled methane reforming and H\textsubscript{2}/carbon synthesis was assembled and tested. Non-equilibrium plasma was generated in a supersonic methane flow. Measurements of discharge parameters showed that a pulsed spark plasma can reach $T \sim 4000$ K for transient chemical reforming. Detailed 2D plasma modeling demonstrated the geometry and dynamics of the plasma channel. Estimated hydrogen production rate is $G$(H\textsubscript{2}) = $4.3 \times 10^{18}$ molec/s for $P$ = 3.5 W discharge. Hydrogen production was measured using gas chromatography. It was shown that the energy efficiency of hydrogen production in a supersonic expanding flow by pulsed spark is 28 kWh/kg hydrogen. Theoretical estimation is 19 kWh/kg hydrogen. Simultaneous measurements of rotation-vibration non-equilibrium in 1-D from pure rotational fs/ps CARS were demonstrated in a CH\textsubscript{4}/N\textsubscript{2} nanosecond-pulsed pin-to-pin discharge. A plasma instability theory was developed and validated.

NETL Collaboration:
Collaborations with NETL on microwave plasma aided fuel pyrolysis and oxidation were established. One graduate student, Christopher Burger, visited NETL and conducted collaborative experiments on microwave assisted methane pyrolysis and oxidation at different pressures and plasma frequencies.

Relevant Publications:


Benefits:
The results showed that hydrogen production by non-equilibrium plasma is significantly increased by using a supersonic nozzle with shifting chemical equilibrium. These results show promising potential to use non-equilibrium plasma to enhance hydrogen and carbon production from methane. A new method for plasma property diagnostics using fs/ps CARS was also developed. 1D distribution of vibrational and rotational temperatures were measured. This new method will significantly simplify the diagnostic system for time and spatial resolved non-equilibrium diagnostics. In addition, a new plasma thermal-chemical instability theory was developed and validated. It provides new insights to control plasma stability and reactivity.