"Advanced Chemical Manufacturing Ecosystem for Agriculture and Transportation Sectors"

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1. Project Summary

Our capability to produce fertilizers and fuels over the last century has resulted in sustainable growth of human civilization and their respective dominance on the planet earth. The socio-economic impact of our innate ability to manufacture these precious commodities is so immense that it cannot be justified by any dollar amount. However over past few decades we have exhausted our technical advances in manufacturing of these chemicals, and the current industry relies on catalysts and the respective processes that are over 30-70 years old thus leading to huge inefficiencies in the system. The current catalysts/processes typically have low conversion, high energy demand (typically high temperature and pressure), short life span, and more often than not severe environmental impact. As outlined clearly by the *"Kentucky's advanced manufacturing ecosystem",* it is essential to address the problem as a whole, where scientific and technical advances are desired at every step, specifically with key emphasis on materials and their properties, development of the novel processes and finally our capability to predict and analyze the huge amount of data resulting from the proposed experimental advances. Here, in this proposal we directly address these issues with emphasis on (a) catalyst by design and (b) novel plasma based processes and (c) utilize the big data platform to understand the role of each component in materials and processes. The primary targets are agriculture and transportation sector, and we will study ***(i) Conversion of nitrogen and hydrogen to ammonia for fertilizers, (b) conversion of methane to higher fuels and plastics, (c) CO_2 conversion, dry reforming and fuel production.*** Moreover, it is absolutely crucial to study these processes now as we have a ten-fold increase in natural (shale) gas production over last decade, (from 4-45 bcf a day in **US** along with significant production in lower Appalachian basin, **KY** – see Figure 1) as well as increased oil production. Therefore, Kentucky is well poised to contribute to the National Economy in a timely manner.

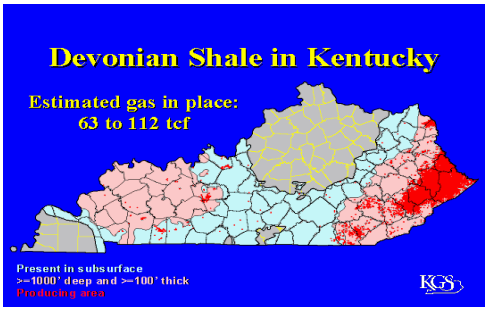
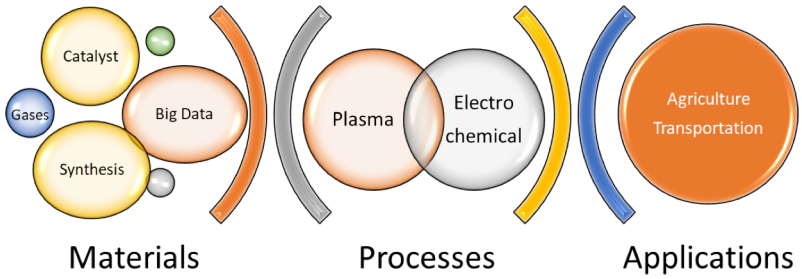


Fig. 1. Shale gas availability in the state of Kentucky



2. Proposed Research Infrastructure & Project Directions

To achieve our goals, we have assembled an interdisciplinary team of researchers and scientists with expertise in catalysis, materials sciences, physicist and computational science. The key members from university of Louisville include M. Sunkara (lead), J. Spurgeon, G. Gupta, W. Paxton, G. Sumanasekera, J. Jasinski, J. Liu, and from university of Kentucky include M. Winter and M. Menon**.** We have a strong collaboration track record in the past with several funded federal research projects and publications. Specifically, we have tremendous expertise in heterogeneous catalysis, electrocatalysis and plasma processing of materials. In the case of plasmas, UofL and UK researchers have developed several tools toward materials processing. These tools range from low pressure and atmospheric plasmas using both radio-frequency and microwave discharges. The technology of nanoscale materials processing using atmospheric plasma discharges has been translated and scaled up by Advanced Energy Materials, LLC toward production of advanced catalysts.

3. Proposed Research

The key scientific question that resides at the heart of this research is ***how a catalyst/process/reactor design can decrease thermodynamic, kinetic and transport losses in a system.*** The proposed research will result in economically viable, high efficiency conversion of natural gas/H_2 to products of choice and will provide fundamental insight into the reaction mechanisms of numerous chemical reactions enabled via plasma catalysis. Furthermore, the advances with plasma catalysis will have direct impact on the existing chemical processing and catalyst industry and chemical startups within the state of Kentucky. In addition to being a net producer of natural gas, Kentucky is home to chemical manufacturing units such as Momentive Plastics, Monumemt Chemicals, Dupont, Dow-Corning and Zeon Chemicals, etc along with catalyst development companies such as United Catalysts (now Clariant) and had several startups in the recent times (Advanced Energy Materials, LLC and PyroChem Catalysts, LLC). a research infrastructure on Plasma-catalysis will have a broad impact on workforce development and associated curricular activities at statewide Universities.

The specific goals of the project include:

a. Plasma assisted heterogeneous catalysis for ammonia: We propose to investigate the use of Ga and its alloys *(Materials by design)* for their synergistic action with plasma excited gas phase for ammonia and others. *(Advanced Processing)*

b. Plasma-assisted Electrochemical Synthesis of Ammonia: The primary objective of this area is to investigate and develop a novel plasma-assisted approach to the electrochemical synthesis of ammonia directly from gaseous nitrogen and water. We will investigate the mechanistic pathways for the reaction and the effect of plasma power and the distribution of excited nitrogen species. *(Advanced Processing)*

c. Theoretical Studies on Reaction Mechanisms: We will design special KMC algorithms to overcome the challenge of extreme computational complexity when simulating catalytic systems for long timescales by focusing on the timescale of the barrier crossings, while treating the wandering motion around a potential energy surface (PES) minimum in a coarse-grained sense. The comprehensive image will appear with experimental and theoretical correlation. *(Big Data)*