QUARTERLY PROGRESS REPORT

December 2015 – February 2016

PROJECT TITLE: Development and Evaluation of Contaminant Removal Technologies for Landfill Gas Processing

PRINCIPAL INVESTIGATOR(S):

John N. Kuhn
Department of Chemical & Biomedical Engineering
University of South Florida
Email: jnkuhn@usf.edu
Phone: (813) 974 - 6498

Babu Joseph
Department of Chemical & Biomedical Engineering
University of South Florida
Email: bjoseph@usf.edu
Phone: (813) 974 - 0692

PROJECT WEBSITE: http://www.eng.usf.edu/~jnkuhn/Hinkley2015.html

Work accomplished during this reporting period:

For the period outlined in this second report, the catalyst has been synthesized and several poisoning techniques have been tested to determine the most effective one in terms of the amount of silica loaded onto and into the catalyst.

For the synthesis, the catalyst support (Fig 1a) was synthesized through the co-precipitation method. It consisted of cerium oxide and zirconium oxide in a 0.6:0.4 weight percent respectively. Nickel, magnesium and platinum (Fig 1b/c) were all then deposited onto the support using wetness impregnation method. Nickel was deposited in a 1.34 weight percent while magnesium was deposited in a 1.00 weight percent and finally platinum in 0.16 weight percent.

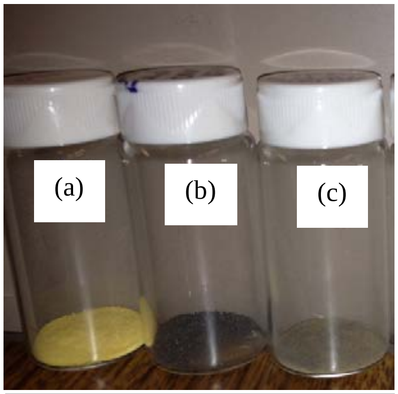


Figure 1: Catalyst support (a), support with Ni and Mg (b), support with Ni-Mg and Pt (c)

The siloxane amounts that were chosen as previously mentioned to poison the catalyst were based on a control of a clean sample (0 days), a lower limit (2.5 days), a middle limit (1 month) and finally a high limit (6 months). These values are based on the same concentration of siloxanes, which was determined as typical representative values from a literature survey.

The challenge faced at this stage in the experimental process is how to dissolve and load the siloxanes onto the catalyst and whether the full effect of the siloxanes will be realized.

In general, a wetness impregnation method was utilized to load the siloxanes. However it was determined that breaking down the siloxanes into silica is very challenging especially in the case of D4 which is a cyclic molecule. Initially, the siloxanes were dissolved in ethanol and were added dropwise through wetness impregnation to a crucible containing the catalyst (Figure 2) and left to dry in atmosphere at 80°C. This method ultimately proved to be ineffective because there was a lot of sample loss. In addition, the amount of silica deposited could not be accurately determined. Furthermore D4 did not readily break down at the chosen temperature.

Figure 2: Catalyst poisoned in crucible

With that in mind, a second synthesis method was utilized using a closed autoclave. The catalyst was loaded and weighed into an autoclave (Figure 3), then the siloxanes (pre-dissolved in ethanol) were added to the catalyst. The siloxane-loaded catalyst was left to dry at 200°C in the closed (air-tight) autoclave for 45 min increments. Although there were slightly better results with this method, however once again D4 did not fully break down and the amount of silica that ended up on the catalyst was much lower than expected indicating that there was still a lot of sample loss or that the siloxanes did not fully break down as expected.

A third method was attempted using the autoclave, however this time, after the final amount of siloxanes was added, the autoclave-containing sample was left for 24 hours in the oven at 200°C. However, again the attempt was not fully successful at breaking down the entire siloxane amount as indicated by the presence of non-decomposed siloxanes.

In addition, we have initiated process simulations for the removal of siloxanes. A student team has also been grouped to design a reactive adsorbent bed for the removal of siloxanes via decomposition. The student team of 4 senior students designed a reactor system for the deposition of siloxanes over a bed of abundant oxide materials.

Figure 3: Catalyst poisoned in autoclave

Future Tasks: The future direction would be to load the siloxanes onto the catalyst using different methods and characterize them to determine the best method to use for the remaining portion of the study. Starting with siloxanes may not be the ideal initial step, attempting to poison the catalyst using other silica-containing starting materials will be used. Then, it is essential to characterize the materials. The catalyst will be characterized using several different techniques such as temperature programmed reduction (TPR), x-ray diffraction (XRD), and identifying the surface area using BET. Those characterization techniques will help to show the reducibility of the catalyst, the crystal structure and the surface area of the catalyst and the effect of the siloxanes on the catalyst if any.

TAG Meetings:
A TAG meeting was not held during this reporting period.

Metrics:

1. List research publications resulting from **THIS** Hinkley Center project.

   None

2. List research presentations resulting from (or about) **THIS** Hinkley Center project.

   - A poster at the USF COE Research Day (see bottom picture).
   - Abstracts have also been submitted the USF Undergraduate Research and Arts Colloquium

3. List who has referenced or cited your publications from this project.

   None

4. How have the research results from **THIS** Hinkley Center project been leveraged to secure additional research funding? What additional sources of funding are you seeking or have you sought?

   PI: Ergas, co-PIs: Kuhn, Joseph and Zhang. "Sustainable Bioenergy Production from the Organic Fraction of Municipal Solid Waste" Preproposal submitted to EREF.

   PI: Kuhn, co-PIs: Ergas, Joseph and Zhang. "Flexible Process for Thermochemical Conversion of Biogas to Fuels and Chemicals" Concept paper submitted to DOE EERE.

5. What new collaborations were initiated based on **THIS** Hinkley Center project?

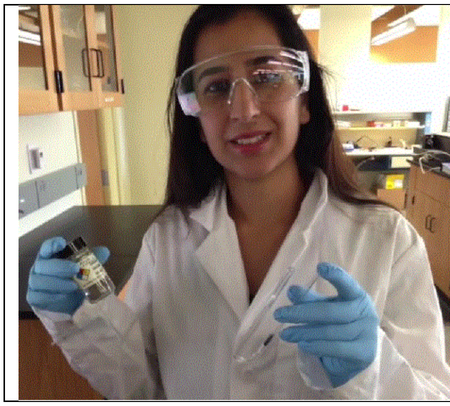
   No change.

6. How have the results from **THIS** Hinkley Center funded project been used (not will be used) by the FDEP or other stakeholders?

   None

Pictures:

The primary student researcher on this project is Nada Elsayed. Anthony Elwell is an undergraduate researcher also assisting with this research.



Nada Elsayed is seen in the pictures above. On the right, she is with the USF COE Dean (Robert Bishop) during the award of a plaque for her USF GSS fellowship.

Tony is a junior Chemical Engineering student working on this project. He recently presented a poster on this project.