Novel Annular Jet Reactor for Direct Conversion of Oligomers to Olefins and Aromatics with Pathway to Carbon Neutrality

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Highlights

- Reactor is capable of high heat transfer rates of 1 million K/s utilizing centrifugal acceleration up to 1 million g
- Oligomers are melted and sprayed directly into reactor as fine droplets (Sauter Mean Diameter $< 50 \ \mu m$)
- The oligomer feed can be derived from mixed plastic waste and thus provides a more direct way of converting plastic waste to high value chemicals
- The CO₂ by-product is produced at elevated pressure and high concentration, reducing capture cost compared with the flue gas in a conventional steam cracker

1. Introduction

Two sustainability challenges that are facing the petrochemical industry are plastic waste and carbon emissions. Currently the primary ways to address circularity are via mechanical and chemical recycling. Chemical recycling takes difficult to recycle mixed plastic waste and converts it to pyrolysis oil. This pyrolysis oil is hydrotreated and later introduced into liquid steam crackers to create circular olefins and polymers. This process has come under considerable scrutiny as significant carbon is lost along with high energy and cost penalties. Thermal insulating characteristic of plastics is at the core of the heat transfer limitation resulting in slow heating of plastics to produce pyoil and in the process over cracking the plastics to light gases and unsaturated oils. These oils are typically hydrotreated to make them suitable naphtha-like feeds for steam crackers. The process of making the pyoil also offers limited decarbonization opportunities as the light gases such as methane are used as fuel to heat the pyrolysis units. In addition, the steam crackers need to be decarbonized via CO₂ capture from flue gases, electrification of the furnace, or combustion of clean hydrogen. To address these issues, the annular jet reactor has been developed to process oligomers directly to olefins and aromatics with a pathway to capture CO₂ at elevated pressure. The oligomers (5,000 to 10,000 Molecular Weight, or MW) can be derived from partial cracking of mixed plastic waste with lower carbon loss, capital, and energy as compared with pyoil production.

Shtern and Hussain [1] reviewed the characteristics of high swirl flows such as in a tornado and developed a 2D axisymmetric analytical solution for the full Navier-Stokes equations. Thin annular high-speed jet near the wall with large recirculation region and intense mixing were recently leveraged for methane pyrolysis [2, 3] where the reactor features heat transfer with minimum mass transfer followed by rapid mixing between the combustion products and feed. The theory guided an optimal design of a vortex burner with a converging-diverging nozzle that provides a stable and robust anchored flame away from the walls. This reactor was recently extended to crack ethane [4] and served as the basis for further development to study of direct cracking of oligomers.

2. Methods

ANJEVOC is an acronym for ANnular JEt VOrtex Chamber. This device is a compact, integrated burner and a millisecond reactor with feed heating rates of a million K/s for endothermic and homogeneous reactions to convert a broad range of hydrocarbons to olefins and aromatics [4]. The fuel, oxygen, and reactants are fed through a series of volutes (conduits that have airfoil shaped passages) to create a high swirling flow with minimal drag. This in conjunction with the shape of the convergent divergent section of the reactor results in an annular jet with peak centrifugal acceleration of ~10⁵-10⁶×g (g=9.81m/s²). The feed system, the order of the hydrocarbons, fuel, and oxidizer, and the high swirling flow create hydrodynamic control to minimize mass transfer of hydrocarbons to the high temperature flame while permitting some heat transfer. In this process, heated polybutadiene oligomer (~175 °C, as a model oligomer) was introduced as a spray from top of the reactor using a Spray SystemsTM SUJ11 injector. The droplet size is controlled to completely vaporize the spray droplets along the axis with low swirl velocity and prevent the droplets from centrifuging to the walls. As these segregated streams (feed hydrocarbon, vaporized oligomers, and combustion products) pass the throat of the reactor, there is fast and violent gas-gas countercurrent mixing via large recirculation of the combustion products leading to rapid mixing. This fast mixing is critical to rapidly cool down the mixture gas to the moderate temperature (~1500 K) where formation of shorter-chain olefinic compounds dominate, while bypassing the high-temperature region where deep oxidation and formation of acetylenics dominate (>1800 K). This is one of the important features that distinguishes ANJEVOC reactor from previous partial oxidation reactors that suffered from low selectivity to high value chemicals and higher loss of C to CO and CO_2 .

3. Results and discussion

Fig. 1 shows representative results from cracking ethane compared to various circular co-feeds (two pyoils with various boiling points, whole range pyoil wax, and polybutadiene oligomer with MW \sim 3000). These proof of concept results demonstrate that the ANJEVOC reactor can be used to directly convert oligomers to olefins efficiently.



Figure 1. Exit composition (wt %) for co-cracking ethane with circular co-feeds (pyoil, pyoil wax, and oligomer). HVCs: High Value Chemicals

4. Conclusions

We have utilized a novel reactor to directly convert oligomers to high value chemicals. It has very high heat transfer rate and ability to convert a fine spray of oligomers to high value chemicals. Its novel design maximizes the high-value chemicals while minimizing undesired by-products. This pathway will provide an efficient way to convert mixed plastic waste oligomers to high value chemicals, and more details along with comparison to current chemical recycling routes will be presented at the conference.

References

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Keywords

high swirl flows; oligomer conversion, thermochemical conversion, olefin production.