Development of a Tool for Determining and Evaluating the Most Optimal Reaction Pathway Systems: Converting Carbon Dioxide to Methanol and Dimethyl Ether

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Abstract

An integral cog in the design of new chemical plants is the identification of promising, feasible reaction pathways, thereby necessitating that an extensive amount of resources be focused on this developmental stage. Despite the substantially expanded quantity of reaction pathway possibilities in the chemical and fuel industries, it is exceedingly difficult to build an economically and environmentally sound manufacturing processes. Additionally, this task has proven to be time consuming. Novel catalytic pathways that exhibit feasibilities at laboratory scale, may not hold true when scaled to an industrial level, leaving potential plant application with much uncertainty. One of the major concerns, is the cost of unit operations required to achieve the reaction and separation conditions, as well as the separation units needed in achieving a marketable purity of the desired product. To increase feasibility, products from a single reactive system can be treated as an intermediate, thereby producing a secondary product with greater economic value. Whilst this is not guaranteed, the possibility cannot be negated. This would involve employing two reactive systems consecutively, in a chemical plant, potentially optimizing the economic and environmental gains of the overall process system.

The Automated Systematic Synthesis Framework tool (ASSF tool) presented herein, is a novel, rapid screening tool which bridges the gap between the scale up of laboratory systems, process design, and economic and environmental analysis, by (1) providing generic design structures, cost and environmental implications associated with the scale up of a reactive system, (2) systematically searching for, and identifying, the possibility of pairing reactive systems as

consecutive reaction mechanisms in a chemical plant, attempting to increase overall feasibility of the process structure. This, however, does not imply that the ASSF tool is able to identify new reaction pathways, but rather uses existing reaction pathways in the inputted database to look at extending the boundaries of the chemical plant to incorporate a secondary and/or tertiary reactive system. This search space reduction-based strategy aids in identifying promising reactive systems, and their likely bottlenecks, prior to the utilization of extensive time and design resources. The potential and novelty in the ASSF tool are demonstrated through its application to a case study focused on the production of methanol (MeOH) and dimethyl ether (DME) via carbon dioxide (CO_2) hydrogenation.

Existing engineering software systems require prior training, and an in depth understanding of multiple engineering concepts, including widely utilized platforms such as Aspen ® Plus. One of the novel aspects of the ASSF tool, however, lies in its generic design and level of detail required to analyse a reactive system, only needing the balanced stoichiometric equation and its operating conditions. Once entered as a database, the ASSF tool searches for potential reaction pathway combinations. Each reactive system undergoes a basic, generic process design, costing the unit operations required in, and leading up to, the reaction and separation systems. These plant structures are then analysed using economic and environmental criteria, with the final, reduced search space being ranked in order of its expected industrial feasibility.

It is important to note, however, that the ASSF tool is not designed to replace existing engineering design and simulation software systems, such as Aspen ® Plus. Instead, the ASSF tool is a novel, precursory platform with the ability of automatically narrowing large reaction pathway search spaces in a fraction of the time than would be needed if individually simulated in the existing software platforms. Once the search space has been reduced and ranked, as per the ASSF tool, the user can focus their design resources on the intricate design of those potentially feasible reactive systems, necessitating the use of extensive engineering software. Additionally, the simple interface of the ASSF tool allows it to be utilized without prior training, providing reliable results with limited/streamlined inputs.

To ensure reliability of the results obtained from the ASSF tool, the code was validated and verified against data published when using existing simulation platforms, i.e., CHEMCAD ® and W-EcoMP, noting minor deviations (< 6%) in capital costs. This is within the ± 30% range for

preliminary design procedures. Previously a reaction pathway database (RPD) developed and analysed by Jugmohan, *et al*. (2020) noted an analysis period of 10 months, owing to the manual design and transfer of data between four intricate software platforms. The use of the ASSF tool on the same RPD results in a complete analysis in under 5 hours, concluding an identical final search space of 5 potentially feasible reactive systems – using economic and environmental criteria as the crux of this feasibility. This reiterates the use and uniqueness of the ASSF tool in identifying promising reactive systems at the early onset of the design process, allowing for future design and research resources to be more focus driven.

Whilst not designed for the purpose of analysing experimental data, the ASSF tool can be expanded to provide a quick, preliminary analysis of experimental data, determining the economic practicality of attaining operating conditions in a scaled up industrial setting. When practical, the ASSF tool provides the user with an indicative range of operating conditions likely to provide the greatest chance of success in future experimental work and scale up ventures, allowing for research resources to be concentrated within this narrow scope.