Process Design of Integrated Biorefinery in Pulp and Paper Industry for Sustainable Development

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Abstract
Integrated biorefinery to the existing pulp mill has been considered as long-term development for the sustainable of both biorefinery and pulp and paper industries. Among many alternatives of promising technologies, the sustainable integrated biorefinery network is developed by a 3-stage systematic methodology supported by computer-aided tools involving synthesis, design, and innovation. Process design and innovation are further performed to improve integration of the biorefinery to an existing pulp and paper process from superstructure-based process synthesis. According to results of synthesis task, the implementations of succinic acid production and black liquor gasification with DME production with the existing Soda pulp mill provide maximum profit based on the generated superstructure and have potential to improve the profitability of Kraft pulp mill. The simulation of the integrated succinic acid and DME process with the existing pulp mill was performed to obtain rigorous configuration as base case; the process is evaluated in energy, economic and environmental aspects. Improvement targets are then defined to generate alternatives in the innovation stage. The alternatives are evaluated to project a practical and sustainable biorefinery network with pulp and paper industry as a system of biofuel, biochemical and bioenergy production.

Keywords: Biorefinery, Pulp and paper industry, Process synthesis, Process design

1. Introduction
The global warming has been an environmental concern driving study of alternative resources to replace fossil resources. In addition, rapidly reduction of petroleum reserves was reported (Singh and Singh, 2012). Biomass has drawn a great attractive as a carbon-neutral resource and abundant quantity overcoming the conventional fossil fuels. Biorefinery is has been of considerable interest from the globally industrial and scientific community as sustainable alternative that utilize renewable biomass to generate green chemicals, fuel and energy replacing oil refinery (Yuan et al., 2013). An integrated biorefinery design for reliable and sustainable system plays an

important role to obtain positive impacts on economics, environment and society as well as providing useful bioproducts to the markets.

Lignocellulosic biorefinery is defined as the second generation has been developed to solve the problem on food-feed competition concerned in the first generation biorefinery utilizing crop feedstock. Due to their low price and abundance, non-food lignocellulose such as agricultural residue and forestry biomass is a possible greener alternative as raw material. Lignocellulosic biomass mainly composes of cellulose, hemicellulose and lignin. Cellulose and hemicellulose are polysaccharide that make up 60 – 90 wt% of terrestrial biomass. They can be hydrolysed into sugar and converted into diverse bioproducts via both biochemical and thermochemical techniques (Choi et al., 2015). Regarding to lignin, it can be a potential building block of a various aromatic compound. However, it typically is burnt as solid and liquor biofuel in pulp and paper industry or utilized as a biomaterial (Chio et al., 2019). Pretreatment and fractionation step is a limitation on cost-effectiveness and efficiency of lignocellulosic biorefinery.

The pulp and paper industry is a global system of lignocellulosic utilization for bioenergy and biomaterials production mainly paper and packaging products. However, the printing paper demand has been decreasing since digital breakthrough was advanced replacing paper use such as tablet, computer, laptop computer and smart phone. Moreover, the pulp and paper industry has been concerned about low innovation system, mature markets of several core products, low margin business and high price volatility. The traditional pulp and paper industry need to adapt for the current market change. The key challenge of the pulp and paper industry development is to materialize a low-carbon bioeconomy by the implementation of the necessary new green innovations for enhancing performance towards profitability and environmental impact. The integrated lignocellulosic biorefinery to an existing pulp and paper industry has received much attention as an opportunity not only to enhance profitability but also supply marketable multi-products of bioenergy, biofuels, and biochemicals apart from paper with the mature supply chain and facilities for significant efficiency and environment advantages. The transformation of today's pulp and paper mill into modern biorefinery system has been considered as a sustainable growth of both biorefinery and pulp and paper industry by many benefits; higher efficiency of material and energy utilization, value-adding of traditional production lines, generating new domestic and international markets for biofuel, bioenergy and biochemical and promoting the sustainable bioeconomy.

2. Methodology

3-stage approach was applied for the integration of biorefinery technologies into pulp and paper industry. The systematic framework is decomposed hierarchically into three design problems (Babi et al., 2015);

<u>Process Synthesis</u>

The process synthesis stage aimed to define the optimal processing routes of integrated biorefinery network by superstructure-based approach which is an effective approach of an integrated business and engineering framework for synthesis and design of chemical and biochemical process (Quaglia et al., 2013). The synthesis scopes are established based on data availability by literature review. The list of implemented biorefinery technologies are collected to generate the superstructure as the network of alternatives that including the existing pulp mills as reactors. The optimal feedstock, product and processing route is obtained by superstructure optimization via generic mathematical model. The promising process based on problem statement was forwarded to further design and improve in the next stages. The process synthesis problem with formulated MI(N)LP model which is huge and complex needs support by computer-aided tools for systematic data management and problem solution. Super-O is employed as user-interface software to systematically collect the technical and economic data for superstructure generation and interface with GAMS solver for superstructure optimization.

Process Design

The promising networks from process synthesis are established as base case. Process design and evaluation of base case was performed in this stage. Process flowsheet of defined base case is constructed by base case simulation for rigorous process configuration and performance. The rigorous information is obtained to analyse and evaluate the base case on economic, energy consumption, environmental impacts so that process bottlenecks are addressed. Improvement targets can further be identified to minimize the process hot-spots or debottlenecks.

Process Innovation

Finally, the improvement targets are achieved in this innovation stage involving two main tasks; alternative generation and alternative evaluation. Innovative alternatives are generated to be the better processes than base case. Sustainable developments are expected depending on the targets from process analysis in the previous stage. The generated alternatives can be created by process improvement strategies that typically are process integration and/or process intensification. The alternatives relying on available technologies and development purposes are evaluated in term of assessments relating to process hot-spots to confirm the improving achievement. Effective alternatives as well as the best sustainable pathway are obtained achieving the improvement targets overcoming the performance of base case. Although process innovation can be omitted, this stage plays a key role to enhance sustainability of the base case to get the better systems with economic and/or environmental advantages.

3. Process Synthesis

For the superstructure generation, Kraft pulp mill with eucalyptus wood feedstock and Soda pulp mill with sugarcane bagasse feedstock are considered as alternatives of the pulping processes for integration of biochemical production via sugar platform and black liquor utilization via thermochemical conversion. Pulp is alternatively utilized for high value-added biochemical production, including (1) succinic acid, (2) lactic acid and (3) ethanol process, comparing to conventional paper production. In case of black liquor utilization, the alternative technologies consist of (1) black liquor gasification combined cycle (BLGCC) for bioenergy production, (2) black liquor gasification with dimethyl ether production (BLG/DME) for biofuel and energy production and (3) lignin extraction apart from conventional Tomlinson boiler. The objective function for superstructure optimization is set to maximize profit that is calculated by product sale, raw material cost, chemical added cost, utility usage and capital costs. Performance criteria, process constraints as well as economic data that are required for superstructure optimization are systematically collected by super-O in-house software. From superstructure optimization solved by super-O interface with GAMS, Integrated succinic acid and black liquor gasification for DME production in the Soda pulping process is obviously the most promising network that provide high profit with multiple bioproducts replacing petrochemicals (Mongkhonsiri et al.,2018). The overview of generated superstructure with the optimal process is shown as Fig. 1.

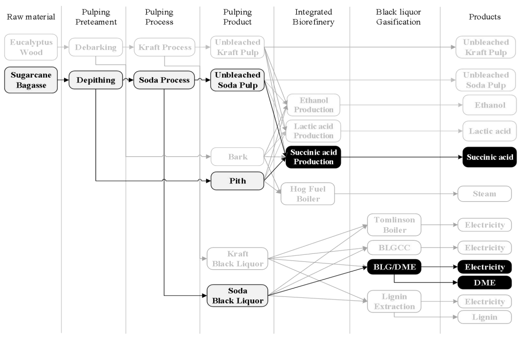

Fig.1 Superstructure overview with the optimal processing route

4. Process Design

Integrated succinic acid and DME production into the existing Soda pulp mill was considered as the base case to be further simulated rigorous process flowsheet and evaluated process performance. Base case was designed to be an energy self-sufficiency system by integration of biomass gasification with imported hog fuel as supplementary energy. Configuration of base case includes the soda pulping process, black liquor gasification with DME synthesis, biomass gasification and succinic acid production (Fig. 2).

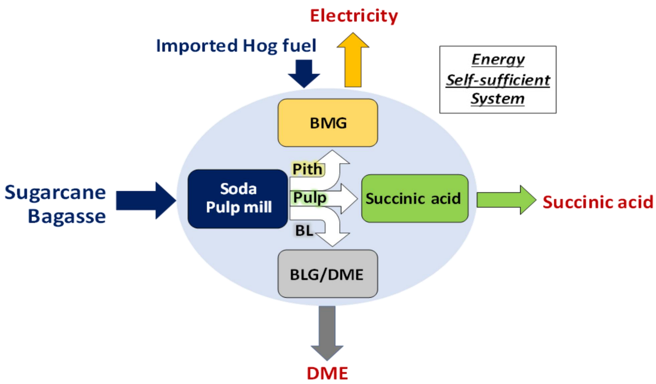

Fig.2 Base case design

As the results of process evaluation shown in Table 1, base case was considered on energy, economic and environmental assessment comparing to conventional Tomlinson boiler and BLGCC. It has higher profit up to 48 M$/year. Although base case shows negative Net CO_2, it generates high CO_2 emission from higher operation of biomass gasification because of extra energy consumption of succinic acid production. Therefore, more CO_2 reduction was set as the improvement target.

Table 1 Results of process evaluation

Info/Process		Tomlinson	BLGCC	Base case (DME+SA)	Innovation (DME+SA:CO2-MeOH)
Raw material cost	$/year	2,300,000	2,323,773	3,661,699	3,661,699
Bioproduct sale	$/year	9,217,290	9,217,290	47,613,854	65,647,466
Power export/import	$/year	-3,369,737	992,724	11,634,970	7,941,740
Capital cost	$/year	194,790	343,547	3,074,024	4,344,862
Chemical cost	$/year	-	-	954,245	10,244,624
Extra utility cost	$/year	-	-	-	4,344,862
Profit	**$/year**	**3,352,763**	**7,542,694**	**51,558,857**	**51,242,361**
Total CO_2 Emission	ton/year	68,632	70,044	298,699	300,611
Total CO_2 Reduction	ton/year	115,899	143,334	331,860	347,941
Net CO_2	**ton/year**	**-47,266**	**-73,290**	**-33,161**	**-47,330**

5. Process Innovation

Due to the improvement target, the designed biorefinery system has opportunity for more sustainability via innovation design. To reduce CO2 emission, large CO2 generated by biomass gasification combined with power plant can be captured from flue gas by employing gas cleaner in black liquor utilization process with extra feed of flue gas from power island. After that, 95% purity of CO2 can be converted to methanol by integrated methanol synthesis via CO2 hydrogenation process (Frauzem, 2017). More DME synthesis from produced methanol leads to more income of extra biofuel as well as less CO2 emission. Following Table 1, the implemented methanol synthesis for more DME process achieves the target for CO_2 reduction around 14,000 tons of CO_2 per year comparing to the base case. However, profit of the innovation design is decreased by 0.6% with expanded chemical, utilities, capital cost of additional methanol process. Comparison of process evaluation is illustrated in Fig.3.

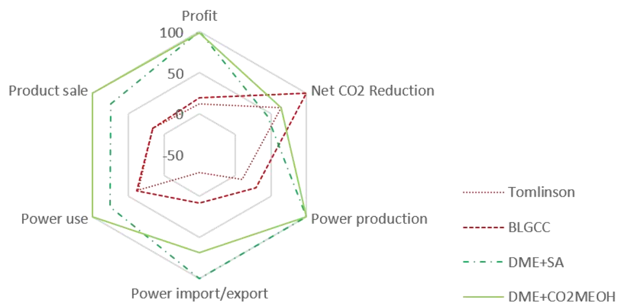

Fig.3 Alternative evaluation by performance factor

6. Conclusions

The systematic framework with computer aided tools is effective approach for developing integrated biorefinery network into the existing pulp mill to improve profitability and environmental impact. Biochemical, biofuel and bioelectricity can be generated to supply the market that promoting a green innovative system with the robust pulp and paper business. Integration of succinic acid and DME production into the existing Soda process provides maximum profitability defined by superstructure optimization. The optimal process set as base case is obtained rigorous information with maximum material efficiency and energy self-sufficiency by process simulation. Then, process evaluation is performed to specify the improvement target that is to reduce CO_2 emission. Due to integrated succinic acid and DME production, external biomass fuel must be imported to meet heat requirement. Consequently, it provides more income from electricity export. As the economic results, integrated biorefinery obviously improve profitability comparing to the conventional pulping process and BLGCC. Though the integrated network shows negative CO_2 emission when CO_2 reduction from replacement of conventional process and biomass growth is involved, process CO_2 emission can be reduced by innovation design to get more sustainable process. Integrated methanol synthesis via CO_2 utilization take place to more DME production as well as reduction of CO_2 emission. CO_2 emission is reduced y 42% whereas profit drop only 0.6%. Comparing to base case and conventional process, innovative process provides environmental benefit and supplies more biofuel as DME to the market demand.

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