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# Design and Integration of Bio-Oil Co-Processing with Vacuum Gas Oil in a Refinery

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Due to the climate change and the strong dependence on fossil fuels, biomass-derived liquid transportation fuels and energy products have been proposed to deal with such situations. Usually, the prices of the bio-diesel and bio-gasoline are much higher than those of fossil fuels. The co-processing of bio-oil and vacuum gas oil (VGO) in a fluid catalytic cracker of a refinery has been proposed to utilize the existing refinery infrastructures and decrease the prices of bio-fuels. However, the integration between the bio-oil production and the existing refinery is not clear. In this work, a superstructure model is built to design and optimize the co-processing process with minimum total annual cost which features to select the optimal bio-oil production technology (fast pyrolysis or catalytic pyrolysis) and obtain the optimal integration scheme. Furthermore, the impacts of the prices of biomass and hydrogen, the bio-oil co-processing ratio on the economics of the co-processing system are also investigated. The optimal integration scheme of the co-processing of bio-oil and VGO can be obtained by solving the proposed model.

## 1. Introduction

Due to the shortage of fossil resources and greenhouse effects, it is significant to develop renewable fuels with less greenhouse gas emissions for the sustainable development of mankind. The bio-fuels have been proposed to deal with the resource dearth and lower the environmental burden. More attention have been paid on the development of bio-fuels which contain lower sulfur impurity due to the heavier and inferior of crude oil and the increasing demand of ultra-low sulfur diesel and gasoline. However, the prices of the bio-fuels are usually higher than those of petroleum-based fuels. There are several reasons for this, the biomass cost is much higher than the cost of crude oil, large capital investment is needed for the construction of a bio-refinery and further treatments are needed for the bio-fuels which only contain certain components compared to the petroleum-based fuels.

The co-processing of bio-oil and crude oil in an existing refinery has been proposed to lower the prices of biofuels by utilizing the existing refinery infrastructures to decrease the capital investment of a bio-refinery (Bezergianni et al., 2018). In addition, the mature fuel distribution system of the refinery can be also adopted for the distributions of the bio-fuels (Espinoza Pérez et al., 2017).

The co-processing of vacuum gas oil (VGO) and bio-oil in a fluid catalytic cracker (FCC) has shown its high technical feasibility for the production of diesel and gasoline with renewable carbon (Valle et al., 2019). Generally, there are two production technologies to make bio-oil, fast pyrolysis and catalytic pyrolysis (Kan et al., 2016). The two technologies of making bio-oil were both investigated for the co-processing with VGO in an FCC. For the co-processing of fast pyrolysis bio-oil and VGO, Pinho et al. (2015) directly co-processed the VGO with pinewood bio-oil without hydrotreating (HDT) in an FCC to make gasoline and diesel. The product yields were slightly changed when the bio-oil co-processing ratio was 10 %. When the ratio is more than 20 %, the coke yield was remarkably increased. Jarvis et al. (2018) used the hydrotreated bio-oil co-processing ratio was less than 20 %.

While for the catalytic pyrolysis oil, it can be directly co-processed with VGO due to its higher heating value and less oxygen and water contents. Wang et al. (2016) co-processed 90 % VGO and 10 % catalytic pyrolysis oil in

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an FCC and the diesel and gasoline yields were not affected. There was more than 7 % renewable carbon in the FCC gasoline by using <sup>14</sup>C analysis. Lindfors et al. (2015) compared the differences of the co-processing of fast pyrolysis oil, HDT fast pyrolysis oil and catalytic pyrolysis oil with VGO in an FCC. The results shown that no matter which bio-oil was co-fed with VGO, its co-processing ratio should be less than 20 % to avoid the increase of coking yield.

Therefore, the bio-oil from fast pyrolysis or catalytic pyrolysis can be both co-processed with VGO in an FCC to make gasoline and diesel with renewable carbon and the product yields are slightly changed if the bio-oil coprocessing ratio is less than 20 %. However, the integration of the bio-oil production and the existing refinery is not clear. The selection of the optimal bio-oil production technology and designing the optimum integration scheme are the priorities for the industrial application of the co-processing of bio-oil and VGO.

In this work, a superstructure model is proposed to obtain the optimal bio-oil production technology and integration scheme while minimizing the total annual cost of the co-processing system. Furthermore, the effects of the prices of biomass and hydrogen and changing bio-oil co-processing ratio on the integration of the co-processing system are also discussed.

## 2. Problem statement

The goal of this work is to design and optimize the co-processing system by choosing the optimal the bio-oil production technology (fast pyrolysis or catalytic pyrolysis) while minimizing the total annual cost of the system. The superstructure is shown in Figure 1. The optimal bio-oil production technology and the optimal integration scheme of bio-oil and VGO co-processing system can be obtained if the product yields of main processes, utility consumption factors per unit feed flowrate, prices of raw materials and utilities and the costs of catalyst and equipment in the reference case are known. In addition, the effects of the prices of biomass and hydrogen and the changing bio-oil co-processing ratio on the selection of the bio-oil production technology can be also discussed.



Figure 1: Superstructure of the co-processing of bio-oil and VGO

## 3. Mathematical model

## 3.1 Objective function

The total annual cost (TAC) of the co-processing system is adopted as the objective function. The optimal biooil production technology and the operation scheme can be obtained by minimizing the TAC. Generally, TAC contains the operating cost and capital cost, which is shown in Eq. (1).

$$TAC = Af \sum_{i} Z_{i}CC_{i} + OC$$
<sup>(1)</sup>

where *TAC* is the total annual cost, in  $y^{-1}$ ; *Af* denotes the annual factor, in  $y^{-1}$ ; *z* is a binary variable, 0 or 1; *CC* represents the capital cost, in  $y^{-1}$ ; *OC* is the operating cost, in  $y^{-1}$ .

## **Capital cost**

According to Figure 1, the capital cost of the co-processing system can be divided into two parts, the bio-oil production and the existing refinery processes. The capital cost of the existing processes, like crude oil distillation, VGO HDT, FCC, diesel HDT and gasoline HDT, are ignored. As for the bio-oil production, the capital cost mainly contains the costs of equipment in the fast pyrolysis and its bio-oil hydrotreating (HDT) process (NREL, 2014), and the catalytic pyrolysis (Vasalos et al., 2016). They are grinder, drier, screw feeder, air

compressor, heater, pyrolysis reactor, cyclone and quench in fast pyrolysis. Besides these equipments, the catalytic pyrolysis also contains the riser and the catalyst regenerator. The HDT process of fast pyrolysis oil has feed oil pump, heat exchanger, compressors of make-up and recycling hydrogen, HDT reactor, flashes and fractionator.

The costs of the above-mentioned equipment can be calculated according to the reference cost in the reference case, the Chemical Engineering Plant Cost Index (CEPCI) and the size or flowrate of the corresponding equipment (Zhang et al., 2014).

$$CC_{i} = CC_{i}^{ref} \left(\frac{CEPCI}{CEPCI_{ref}}\right) \left(\frac{m_{i}^{feed}}{m_{i}^{ref}}\right)^{sf}$$
(2)

where  $CC_i^{ref}$  is the reference cost, in \$;  $m_i^{feed}$  and  $m_i^{ref}$  denote the feed flowrates in this case and in reference case, in t·h-1; *sf* represents the scale-up factor.

The related data to calculate the capital cost are shown in Table 1.

Table 1: Related data to calculate capital cost

| Parameter | CEPCI | CEPCIref | sf  | Af   |
|-----------|-------|----------|-----|------|
| Value     | 533.9 | 567.3    | 0.7 | 0.23 |

## **Operating cost**

The operating cost contains the costs of biomass, electricity, water, hydrogen, catalysts for catalytic pyrolysis and bio-oil HDT processes. The revenue of bio-gas and bio-char should be subtracted from the operating cost.

$$OC = \left(C_{biomass} + C_{elec} + C_{water} + C_{H_2} + C_{cat} - R_{bio-gas} - R_{bio-char}\right) AOT$$
(3)

$$C_k = m_k \rho_k \tag{4}$$

where  $C_{biomass}$ ,  $C_{elec}$ ,  $C_{water}$ ,  $C_{H_2}$ ,  $C_{cat}$  and  $C_{biomass}$  denote the cost of biomass, electricity, water, H<sub>2</sub> and catalyst, in \$·h<sup>-1</sup>;  $R_{bio-gas}$  and  $R_{bio-char}$  mean the revenue of bio-gas and bio-char, in \$·h<sup>-1</sup>; AOT is annual operating time, in h·y<sup>-1</sup>; *m* represents the consumption of each item, in t·h<sup>-1</sup>, kW or Nm<sup>3</sup>· h<sup>-1</sup>; *p* is the price of utilities, raw materials and by-products shown in Table 4, in \$·t<sup>-1</sup>, \$·kWh<sup>-1</sup>, \$·GJ<sup>-1</sup>or \$·Nm<sup>-3</sup>; Subscript *k* denotes the utilities or the raw materials consumed in the co-processing system.

The consumptions of electricity, water (NREL, 2014; Vasalos et al., 2016) and hydrogen (Gueudré et al., 2017) can be calculated according to the consumption factor of utility and the flowrate of the bio-oil product.

$$m_k = m_{\text{bio-oil}} f_k \quad \forall k \in \mathcal{K} \tag{5}$$

where  $m_{bio-oil}$  is the flowrate of bio-oil, in t·h·1; *f* denotes the consumption factors of electricity, hydrogen and water, in kWh·t<sup>-1</sup>, Nm<sup>3</sup>·t<sup>-1</sup> or t·t<sup>-1</sup>; *K* contains electricity, hydrogen and water. The related data to calculate the operating cost are shown in Table 2.

Table 2: Related data to calculate operating cost (NREL, 2014, Vasalos et al., 2016)

|             | Electricity<br>kWh·t <sup>-1</sup> bio-oil | /Water /<br>kWh·t <sup>-1</sup> bio-oil | Hydrogen /<br>Nm <sup>3.</sup> t <sup>-1</sup> bio-oil | Base capacity/<br>t·day <sup>-1</sup> | ′Base catalyst<br>cost / \$·y⁻¹ |
|-------------|--|---|--|---------------------------------------|---------------------------------|
| FP* and HDT | 360  | 0.3                                     | 190  | 2000                                  | 6141656                         |
| CP*         | 447.6                                      | 0.3                                     | 0  | 500                                   | 2368501                         |

Note: FP means fast pyrolysis; CP denotes catalytic pyrolysis.

## 3.2 Constraints

The mass balance of each equipment and process should be satisfied.

$$\sum_{j} F_{j}^{in} = \sum_{l} F_{l}^{out}$$
(6)

where *F* is the mass flowrate of an equipment, in t $h^{-1}$ ; Superscripts *in* and *out* denote inlet stream and outlet stream, respectively; Subscript *j* and *l* are the sets for in the inlet streams and the outlet streams, respectively.

$$F_i^{out} = \sum_j F_j^{in} y_i$$

where *y* is product yield, in %.

## 4. Case study

## 4.1 Basic data

An FCC of a refinery with crude oil processing capability of 8 Mt/y in China is adopted as the co-processing site. The main scheme of the refinery is that the VGO is obtained from the crude distillation unit and cracked in the FCC with annual processing capability of 1.2 Mt to produce gasoline and diesel which are then upgraded in gasoline and diesel HDT processes, respectively. 20 % bio-oil from the fast pyrolysis (including the following HDT process) or the catalytic pyrolysis is mixed with 80 % VGO and co-processed in the FCC to produce gasoline and diesel with renewable carbon.

Pulpwood is used as the biomass feedstock to make bio-oil. The product yields for the related processes are shown in Table 3. The prices for biomass, utilities and by-products are listed in Table 4. The biomass price contains the costs for the planting, harvest, collection, pretreatment, storage and transportation.

| Yield/%  | FP   | FP oil HDT | CP   | FCC  | Gasoline | HDT Diesel HDT |  |
|----------|------|------------|------|------|----------|----------------|--|
| Bio-oil  | 52.5 | /          | 33   | /    | /        | /              |  |
| Bio-gas  | 26   | /          | 53   | /    | /        | /              |  |
| Bio-char | 21.5 | /          | 12.5 | /    | /        | /              |  |
| HDO oil  | /    | 66         | /    | /    | /        | /              |  |
| Gasoline | /    | /          | /    | 48.1 | 99.5     | 7.6            |  |
| Diesel   | /    | /          | /    | 23   | /        | 91.2           |  |

Table 3: Product yields of pulpwood pyrolysis, FCC and HDT processes (Kan et al., 2016) (Wu et al., 2017)

| Table 4: Prices | for biomass, | utilities and | by-proa | lucts |
|-----------------|--------------|---------------|---------|-------|
|-----------------|--------------|---------------|---------|-------|

| ltem  | Biomass/\$·t <sup>-1</sup> | Electricity/\$·kWh <sup>-1</sup> | Water/\$·t <sup>-1</sup> | Hydrogen/\$·Nm <sup>-</sup> | <sup>3</sup> Bio-gas/\$·GJ <sup>-1</sup> | Bio-char/\$·t <sup>-1</sup> |
|-------|----------------------------|----------------------------------|--------------------------|-----------------------------|--|-----------------------------|
| Price | 67.5                       | 0.07                             | 0.065                    | 0.893                       | 4.7                                      | 20                          |

## 4.2 Optimal results

The proposed model is solved in GAMS 24.1 with solver CONOPT to obtain the optimal bio-oil production technology and integration scheme by minimizing TAC of the system. The effects of the biomass and hydrogen prices and the bio-oil co-processing ratio on the selection of bio-oil production technology are also investigated. According to the optimal results from GAMS, the optimal bio-oil production technology and integration scheme are shown in Figure 2. The details of the co-processing cost when integrating different bio-oil production technologies are presented in Table 5.



Figure 2: Optimal integration scheme and its main mass balance

According to Figure 2, the optimal integration scheme of the co-processing of bio-oil and VGO is that the bio-oil is obtained by the catalytic pyrolysis, which is then mixed with VGO and co-fed to the FCC to produce the gasoline and diesel which are then upgraded in their HDT processes, respectively. In this case, the existing

infrastructures in the refinery like FCC, diesel HDT and gasoline HDT are all utilized to lower the capital investment of the bio-refinery. The main mass balance of the co-processing system is also given in Figure 2. According to Table 5, the TAC of the integrated process with fast pyrolysis and the following HDT process is \$100.59 M, while for the one with catalytic pyrolysis it is \$99.5 M/y. The cost of the integrated process with fast pyrolysis. Therefore, the catalytic pyrolysis should be chosen as the bio-oil production technology. The main differences of the two bio-oil production technologies are the costs of biomass, hydrogen and the capital investment. The biomass cost of the integrated process with fast pyrolysis, while the hydrogen cost of the process with fast pyrolysis, while the hydrogen cost of the process with fast pyrolysis, while the hydrogen cost of the process with fast pyrolysis is \$47.35 M/y more than the one with catalytic pyrolysis. The capital cost of the integrated process with catalytic pyrolysis is \$47.6 M more than the one with fast pyrolysis.

Table 5: Comparison of cost of different pyrolysis processes to be integrated in a co-processing scheme

|    |                                     |             |          | , ,   |          |        |                      |                  |          |
|----|-------------------------------------|-------------|----------|-------|----------|--------|----------------------|------------------|----------|
|    | Operating cost/M \$·y <sup>-1</sup> |             |          |       |          |        |                      | Conital cost/M C | TAC/     |
|    | biomass                             | electricity | water    | $H_2$ | catalyst | gas    | char Capital cost/iv |                  | M \$∙y⁻¹ |
| FP | 29.76                               | 7.032       | 0.005442 | 47.35 | 3.709    | 0.2632 | 2.116                | 65.70            | 100.59   |
| СР | 59.35                               | 7.520       | 0.004680 | 0     | 11.41    | 2.149  | 2.690                | 113.3            | 99.50    |

#### 4.3 Sensitivity analysis of feedstock cost and co-processing ratio

#### **Biomass price**

Due to the uncertainty of biomass harvest, which would affect the biomass price, the effect of the changing biomass price on the integration is discussed in this section.

According to Figure 3, the operating costs of the integrated processes for co-processing with fast pyrolysis and catalytic pyrolysis are both increased with the increase of biomass price, while the capital costs are not changed. As a result, TAC is increasing. As the biomass consumption of fast pyrolysis is much lower than the one of catalytic pyrolysis, the increase of biomass cost in catalytic pyrolysis is much higher than the one in fast pyrolysis. When the biomass price is  $70 \cdot t^{-1}$ , the TACs of the co-processing schemes with two alternative pyrolysis technologies integrated are similar. Therefore, when the price is higher than  $70 \cdot t^{-1}$ , the fast pyrolysis should be chosen as the bio-oil production technology. Otherwise the catalytic pyrolysis is the optimal one.



Figure 3: Effect of biomass and hydrogen prices and bio-oil co-processing ratio

## Hydrogen price

It is known that the hydrogen cost is the second largest cost of the raw materials in a refinery. Thus, the effect of changing hydrogen price on the integration should be discussed.

Similar to the effect of biomass price, the changing hydrogen price only affects the operating cost. The higher hydrogen price makes a higher operating cost. When the hydrogen price is \$0.87 \Nm<sup>-3</sup>, the TACs of the processes with different bio-oil production technologies are similar. That is to say, when the hydrogen price is higher than \$0.87 \Nm<sup>-3</sup>, the catalytic pyrolysis should be chosen as the bio-oil production technology. Otherwise the fast pyrolysis should be adopted.

## **Bio-oil co-processing ratio**

The bio-oil co-processing ratio, i.e. bio-oil and VGO feeding ratio, will strongly affect the consumptions of biomass, hydrogen and utilities and the capital investment of the pyrolysis process. The effect of bio-oil co-processing ratio should be discussed.

According to Figure 3, the operating cost and capital cost are both increased with the increase of bio-oil coprocessing ratio, which then causes the TAC to increase. The critical ratio of bio-oil and VGO is about 12 % when the TACs of the different bio-oil production technologies integrated with the VGO are similar. When the ratio is more than 12 %, the catalytic pyrolysis should be adopted. Otherwise the fast pyrolysis is the optimal bio-oil production technology.

## 5. Conclusions

A superstructure model is proposed to design and optimize the co-processing process and obtain the optimal bio-oil production technology and integration scheme. The results show that the catalytic pyrolysis and the fast pyrolysis can both be used to produce bio-oil. The optimal bio-oil production technology in the proposed case is catalytic pyrolysis. The catalytic pyrolysis should be chosen as the bio-oil source if the hydrogen price or the bio-oil co-processing ratio are relatively high. When the biomass price is high, the fast pyrolysis should be adopted.

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