

Catalytic Pyrolysis of Fuel Oil Blended Stock for Bio-Oil Production: A Review

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Catalytic pyrolysis is a combination process of fast pyrolysis in the presence of catalyst. The process capable to break the complex petroleum hydrocarbon into smaller compounds. It is known as a promising technology to convert waste from fuel oil blended stock (FOBS) into useful products such as bio-oil. FOBS could serve as one of the potential sources to produce fuel. FOBS known as a blend of residue oil, mixed-oil and incomplete oil forms a complex structure featuring toxic and hazardous compound creating risk towards human and environment. However, it still contains certain extend of precious hydrocarbon. In order to tackle the problem, thermochemical conversion was suggested and catalytic pyrolysis is hypothetically a reliable process for producing bio-oil from waste such as FOBS at temperature between 400-650 °C. The reaction is oxygen-free process as the process operate at elevated temperatures in the absence of oxygen and is cost-effective and at the same time able to produce high yields of valuable products by turning waste into valuable hydrocarbon range bio-oil. High quality bio-oil produces in C₄-C₂₀ range will be mainly consisting of gasoline and diesel like product. In this array, bio-oil has the potential to serve as a perfect alternative fuel source, lowering the need for fuels derived from unrenewable petroleum. The aim of this review is to discuss the state of art transformation of FOBS by catalytic pyrolysis to produce bio-oil from recent literatures based on the reviewed the presence of bi-metallic catalyst gives a high yield of bio-oil and in the range of valuable fuels. A variety of metals need to be studied in the future in order to get good results, highlighting the study's deficit.

1. Introduction

The exhaustion of fossil resources and the rapid growth of global populations contributed to a negative vibe on the energy supply chain. This has led to an increase in the demand for energy in our day-to-day living, and it is anticipated that this need will expand by 50 % by the year 2050 (Anahas and Muralitharan, 2018). Due to the high cost of technology, the utilisation of renewable energy is still limited, especially in countries with low incomes (Teo et al., 2021). There is a serious demand to produce alternative energy source. In Malaysia, petroleum refineries with capacity almost 400,000 litre per day produced FOBS and oily sludge approximately 900 kg per year which mostly contains oil, water and inorganic sediments (Ling and Isa, 2017). In order to combat the depletion and demand for non-renewable fuel as well as to manage the surplus of undesired FOBS, recycling of FOBS has gained attention because it has could be transformed into the energy fuel (Kuan et al., 2020). FOBS is an alternative energy source that is required to substitute fossil fuels in order to maintain environmental sustainability and human civilization. Clean chemical process methods are needed to remove pollutants from hazardous substances. Normally, thermochemical method such as gasification, combustion and pyrolysis is employed to produce value-added products from waste (Jahirul et al., 2012). Perceivably, pyrolysis has replaced combustion and gasification due to its environmental benefits. In the absence of oxygenated chemicals or oxygen, high-temperature dioxin production is precluded (Singh and Ruj, 2016). However, the pyrolysis process was upgraded by the presence of catalyst which is known as catalytic pyrolysis. Catalytic pyrolysis attempts to increase overall process and conversion efficiency by reducing activation energy and process temperature, as well as maximising end product production It is also can create high yields of low-

molecular-weight liquid fuel with high quality by using less energy (Oyeleke et al., 2021). Herein it can say that catalytic pyrolysis is the best way to treat FOBS. The process yields three principal products which are bio-oil, bio-gas and bio-char. The review focus to present the yield of bio-oil from FOBS by using catalytic pyrolysis.

2. Fuel oil blended stock (FOBS) and composition

FOBS is any unfinished oil or product that is mixed with other unprocessed oils to make a finished product, but because it is hard to process further, it is often seen as an undesirable product (Teo et al., 2021). Due to the high levels of hydrocarbons and other carbonaceous components that are included in FOBS, it is considered to be a harmful waste in several nations (Huang et al., 2014). The appearance of FOBS is in black colour due to its high viscosity and high level of asphaltenes, resin and heavy metal (Speight, 2017). Based on the properties it will form a complex structure and consequently difficulty to be processed (Sun et al., 2021). However, the recycling of FOBS and oily sludge received increased attention as it has the quality as potential substitute for petroleum energy (Kuan et al., 2020). Basically, around 30 % of the petroleum residue is oil and 40 % is water. Solid particles are the remaining content (Huang et al., 2015). Indeed, FOBS is recognised as a fuel replacement, and heavy fuel oil (HFO) is one of its most basic parts in oil refineries (Goldsworthy, 2006).

The composition of FOBS's physical and chemical properties are vary by origin, depth, and age. The chemical composition of heavy fuel oil is highly complex, as it is a mixture of many different sorts of components. A comprehensive characterization of HFO is both impracticable and unattainable. Separation by column chromatography is a typical technique for breaking down heavy fuel oil into smaller fractions. SARA analysis, which represents for Saturates, Aromatics, Resins, and Asphaltenes fractions provides the data of a compound class characterization that is prominent in FOBS (Rahimi and Gentzis, 2007). This complex structural combination and its fractions were analysed for heteroatom content by using GC-MS. Recent study highlight the polar portions of the HFO typically have the greatest heteroatoms (S and N) in the resins and asphaltene fractions and the least in the saturates (Garaniya et al., 2018).

The SARA analysis provides a practical way for estimating the heteroatom content of the constituents. Table 1 showed SARA and elemental analysis of HFO and its fractions. The saturates fraction is mostly carbon and hydrogen (98.5 %). The reaction's H/C ratio is less than 2, which suggests condensed cyclic structures may be essential (Garaniya et al., 2018). Elemental composition of feedstocks gives an early indicator of quality and the quantity of molar hydrogen needed for upgrading when paired with molecular weight (Speight, 2017).

Table 1: Elemental and SARA analysis of HFO and its respected fractions (Garaniya et al., 2018)

| SARA Analysis | | | | Elemental Analysis | | | |
|-----------------|----------|-------|-------|--------------------|------|-------|-------|
| Components | Weight % | C % | H % | S % | N % | Total | H/C |
| Aromatics | 55.81 | 83.83 | 9.92 | 4.28 | 0.20 | 98.23 | 1.420 |
| Saturates | 24.08 | 85.32 | 13.17 | 0.48 | 0.05 | 99.02 | 1.852 |
| Asphaltenes | 7.86 | 83.49 | 8.03 | 7.07 | 0.75 | 99.34 | 1.154 |
| Resins | 6.66 | 80.03 | 10.55 | 2.78 | 0.82 | 94.18 | 1.581 |
| Loss & Unknowns | 5.59 | 85.17 | 9.16 | 4.42 | 0.24 | 98.99 | 1.290 |

In general, thermochemical process has been adopted as process to convert waste into value-added product. Under thermochemical process are gasification, pyrolysis and combustion (Jahirul et al., 2012). Gasification and combustion are exothermic processes that take place in an oxygenated environment. While pyrolysis is an endothermic method that degrades waste in an oxygen-deficient and inert atmosphere (Zaman et al., 2017).

Pyrolysis is the most practicable technique for generating small-molecular-weight compounds in an oxygen-free atmosphere. This approach has low manufacturing costs as thermal consumption at optimum condition around 500 °C instead of gasification and combustion that can be more than 800 °C (Bulushev and Ross, 2011), simple processing, and high productivity efficiency (Uyar and Aydın, 2021). However, there are some drawback of the process which is the oil produced by this process contains more contaminants and residues because pyrolysis is essentially a thermal process (Ma et al., 2020) and also coke can easily formed (Ochoa et al., 2020). As a result, pyrolysis process is upgraded with the presence of catalyst in order to solve the problem appeared.

3. Catalytic pyrolysis

Pyrolysis is a remarkable technology that can transform local, abundant, and waste into valuable products with little capital investment (Dickerson and Soria, 2013). Pyrolysis process convert the waste into valuable products which are; bio-oil, bio-gas and bio-char products. A portion of the volatiles are transformed into tar, bio-oil, or pyrolysis oil. Tar production during pyrolysis process resulted to toxic aerosols, condensation, and equipment blockage. Some of the key concerns that arises during the process include (i) bio-oil appeared in a dark brown

liquid indicating it contains corrosive, viscous of heavy inorganic and organic compounds (Li and Suzuki, 2009); (ii) pyrolysis results in the development of char which is a solid residue rich in carbon (Guan et al., 2016). Numerous efforts have been made to improve the qualities of bio products produced by pyrolysis (Zarnegar, 2018). In spite of progress made in the production of bio-oil through pyrolysis, bio-oil cannot replace conventional fossil fuels in a systematic manner because of its high oxygen concentration, high thermal instability, enormous viscosity and caustic nature (Zhang et al., 2007). The existence of water and oxygenated organic molecules has reduced the effectiveness of bio-oil from pyrolysis process (Trane et al., 2012). Bio-oil should be free of by-products, especially oxygen. The aim to increase the hydrogen-to-carbon (H/C) proportion and a decrease the oxygen-to-carbon (O/C) fraction, which reflect an excellent liquid product quality (Dickerson and Soria, 2013). These flaws can be remedied by integrating efficient catalysts into the pyrolysis reactor to enhance bio-oil generation. As a result of this upgrade, bio-oil has increased thermal stability and heating value by reducing the amount of oxygen and polymerization by-products (Carlson et al., 2011). The inclusion of catalyst is an excellent alternative for char conversion because it not only lowers the solids but also influences the oil production. A significant decrease in the hazardous compounds and tar yields can also be achieved using the catalyst (Zarnegar, 2018). The adaptability of the raw material, accessibility of the infrastructure, chemical homogenization, and low operating costs all endorse the utilization of the catalyst for the pyrolysis (Hassan et al., 2020). Catalytic pyrolysis has spectacular advantages where it can reduce the activation energy for breaking C–C bonds, decrease reaction temperature, improve selectivity of product by forming lower molecular weight species in gasoline and diesel range and also decrease the residence time in the reactor (Kassargy et al., 2018). The selected catalyst must be highly active, strongly immune to deactivation, recyclable, and economical (Dickerson and Soria, 2013).

Catalytic improvement can change the properties of bio-oil by removing oxygenated compounds with water and carbon dioxide, reducing the size of the molecules, and changing the chemical properties to be like those of petroleum products through cracking, dehydrogenation and deoxygenation process (Dickerson and Soria, 2013). Carbon-carbon bonds are broken during cracking, which results in the formation of lighter molecules. Dehydrogenation reduces the quantity of hydrogen atoms by breaking C–H bonds, while carbon atoms remain unchanged. The condensation of two hydrocarbons produces a product with a larger molecular weight (Mahinpey et al., 2007). Deoxygenation is a term used to describe the process of removing several oxygenated molecules from bio-oil. There are three types of reactions in the deoxygenation process which are (i) dehydration (ii) decarbonylation and (iii) decarboxylation (Banks and Bridgwater, 2016).

3.1 Types of catalyst used and their yield in catalytic pyrolysis

The selection of a catalyst affects product dispersion and chemical selectivity and product yield. In this reviewed, the focussed is on the catalyst that able to enhance bio-oil yield and decrease the gas and coke formation. According to the recent studies, metal-oxide and zeolite catalysts are favoured in catalytic pyrolysis as it can inhibit the development of stable chemical structures in hydrocarbons, hence hastening their decomposition (Duan et al., 2014). Various sorts of catalyst have unique characteristics including surfaces acidity, morphology, pore diameter, and pore size dispersion that affect yield and selectivity (Banks and Bridgwater, 2016). Meanwhile, by support of the process conditions such as temperature, ratio of catalyst and feedstock as well as reaction time, it is able to improve the production of main products (Foong et al., 2021). Table 2 summarized the recent findings on conversion of FOBS to produce bio-oil by catalytic pyrolysis.

ZSM-5 has exhibited in activating the oil sludge pyrolysis products, which enhanced the subsequent aromatization reactions (Hou et al., 2022). Then, the results showed that the sole bi-metallic catalyst (Kumar et al., 2019) performed better at deoxygenating all oxygenated compounds than the combined mono-metallic catalysts (Liang et al., 2017) and favoured the production of aliphatic and aromatic hydrocarbons. In this case the combination of monometallic catalysts produced a high yield of biogas. Dolomite generates hydrogen-rich syngas from petroleum waste. Increased catalytic temperature resulted in an increase in H₂ production (Huang et al., 2015). The addition of metal oxides such as CaO, MgO, KOH, Fe₂O₃ and Na₂CO₃ to the framework of the catalyst has resulted to good effect on pyrolytic oil by producing more hydrocarbons and less oxygen heterocycles. However, vast fraction of yields from pyrolysis are produced (Jin et al., 2021).

For the synthesis of benzene, toluene, and xylene, ZSM-5 has shown outstanding performance due to its porous acidic properties (He et al., 2021). The catalytic capabilities of zeolite catalysts are intrinsically tied to pore size, distribution, structure, and acidity. By virtue of its molecular shape-selectivity properties, such catalysts are able to transform waste oil into an aromatics-rich high-octane fuel that is comparable to gasoline (Milne et al., 1990). Insight, aromatic hydrocarbons can be produced more effectively with the help of unique pore channels of ZSM-5 (Wang et al., 2021). Theoretically short residence time, low reaction temperature, large ratios of catalyst to oil; and large steam to oil ratios are good catalytic pyrolysis conditions for heavy oil transformation to light hydrocarbon; and various catalysts give different effects on the product distribution (Xiang-Hai et al., 2003). Temperature and catalyst condition play a role in determining the amount of liquid product yield. The efficiency

of gasoline and diesel increases as temperature, catalyst percentage, and heating rate increase (Demirbas et al., 2017). Despite the benefits of pyrolysis and catalytic reforming and their high productivity, there still room for further research on this theme.

Table 2: Recent finding on conversion of FOBS to bio-oil production under catalytic pyrolysis condition.

| Raw Material | Catalyst | Conditions | Bio-oil yield % | Bio-oil Composition | References |
|--------------------------------|--|--|-----------------------------|---|-------------------------|
| Oil sludge | ZSM-5 | 650 °C 1 h | 60 % | Higher production of aromatic compounds (C6 and above) | (Hou et al., 2022) |
| Oil based drill cutting (OBCD) | CaO MgO | 450-500 °C 1 h | 105.49 % 82.28 % | Alkanes, alkenes, aromatics, and heterocyclic compounds | (Lv et al., 2022) |
| Oily sludge | Fe ₂ O ₃ KOH + CaO | 30 g / sample 450-850 °C 15-60 min | 53.59 % 76.84 % | Toluene, O-xylene, Azulene | (Jin et al., 2021) |
| Pyrolytic oil | Cu- Ni/ZSM-5 | 500 °C 0.5 h | 70 % | Hydrocarbon range C ₆ -C ₁₃ | (Kumar et al., 2019) |
| Pyrolytic oil | Ni/ZSM-5 | 500 °C 8 min | 20 % | High yield of gas | (Liang et al., 2017) |
| Oily sludge | KOH | 600 °C 1: 2 | 60.1 % decline to 53.5 % | Adding KOH saturate and aromatics composition increase | (Lin et al., 2017) |
| HFO | Na ₂ CO ₃ | 160–350 °C 0.5 h 2-10 wt. % | 82.66 % | Gasoline-like-product; 21.5-39.1% in 5 % and 32.5-42.5% in 10 % Diesel-like-product; 9.3-29.8 % in 5 % and 15.5-33.7 % in 10 % | (Demirbas et al., 2017) |
| Petroleum sludge | Dolomite | 600-1000 °C 30 s | - | Hydrogen rich syngas | (Huang et al., 2015) |

3.2 Yield of pyrolytic products from FOBS

Bio-oil yield are thick, dark brown liquid with a pungent smoky stench. Organic compounds contribute up to 40 % of the normal bio-oil makeup, while tars such as naphthalene, toluene, and benzene make up the remaining 25% (Bulushev and Ross, 2011). Bio-oil cannot be distilled because the chemical changes it goes through make it susceptible to heat. It has an approximate heating value of 17 MJ/kg. The properties of bio-oil depend on the feed, the structure of the reactor, and the process parameters. The selection of pyrolysis reactor depends on feedstock particle size, which affects operation and scale (Uddin et al., 2018).

From Table 2, it can be summarized that bi-metallic catalyst is capable to produce high bio-oil and obtain large percentage of aliphatic hydrocarbons, having high quality bio-oil within the range of diesel and gasoline. The cumulative impact between the active bimetals that combine their different physicochemical characteristics to create new properties was successfully proven to catalyze the reaction synergistically. High metal dispersion and small particle size, lead to high rate of reduction of catalytic and minimal carbon production (Mohd-Nasir et al., 2021). By using metal oxides's catalyst such as CaO, MgO, KOH, Fe₂O₃ and Na₂CO₃, the result gives high yield of bio-oil in the range of C₁₂-C₂₂. The most smaller range is in the scale of diesel fuel (Lv et al., 2022). In the study of using KOH, asphaltenes were reduced by half, while oil product quality improved in terms of average molecular weight, viscosity, heating value, asphaltenes, and straight chain hydrocarbons (Lin et al., 2017). It can be highlighted that with the presence of suitable catalyst, FOBS can be transformed into oil in the range of gasoline and diesel fuel. For usage in gasoline engines, a light fraction should be gathered. For heavy fraction oil, it will be employed in diesel engines (Kassargy et al., 2018).

4. Conclusion

The generation of bio-oil is possible with the aid of appropriate catalyst with good yield. Catalytic pyrolysis performance can increase up to 50 % in aromatic yields when compared to conventional pyrolysis. Few researchers had promoted ZSM-5 catalyst as it has a very great performance and also introducing ZSM-5 to mono- and bi- metallic catalyst to degrade complex structure to smaller hydrocarbon fraction. Mono-metallic catalyst has been upgraded to bi-metallic catalyst to give high yield bio-oil due to more deoxygenation activity being favoured which resulted to increase in bio-oil production. Further study needs to be conducted on the variants of bi-metallic catalyst for catalytic pyrolysis process of FOBS. To enhance aromatic yield further, there are few key factors that must be studied which are high catalyst to feed ratio, rapid heating rates and appropriate

catalyst selection by including consideration of the catalyst's active sites and pore structure. Reaction parameter such as temperature, heating rate and carrier gas flow rate are crucially important to support high bio-oil yield.

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