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Sustainable Biofuel Production for Compatibility with Conventional Refining Technologies

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Abstract

The text describes the exploration of various process schemes for producing second-generation biofuels, focusing on reducing the high investment costs associated with gasification followed by Fischer–Tropsch synthesis. Three pilot-scale approaches are presented: Catalytic pyrolysis of biomass using fluid catalytic cracking zeolitic catalysts at moderate temperatures, with results showing the impact of the catalyst on yields of coke, gas, and liquid products-processing gas oil mixed with hydrotreated biomass pyrolysis liquids in FCC units, demonstrating technical viability depending on the biomass liquid concentration. Co-hydro processing vacuum gas oil with sunflower oil to produce mid-distillate biofuels such as gasoline and diesel, with discussion on catalysts and process conditions. These approaches aim to produce high-quality biofuels compatible with existing fossil fuel infrastructure but with potentially lower investment costs compared to traditional gasification and Fischer–Tropsch routes. If you would like, I can help summarize, explain further, or assist with specific questions on this topic.

Keywords: Biofuels, biomass, pyrolysis, FCC

Introduction

This text discusses the current state and challenges of biomass as a renewable energy source for biofuel production: Biomass is the primary renewable energy source today, convertible into liquid, solid, and gaseous fuels via biological and thermochemical processes. First-generation biofuel technologies use only a small portion of biomass and their CO2 reduction benefits are debatable. Second-generation biofuels use lignocellulosic biomass, which requires high investment costs, resulting in expensive fuels. An alternative for affordable biofuels is co-processing low-cost bio-based liquids (such as bio-oil or vegetable oils) within existing petroleum refining infrastructure, which demands minimal capital investment. This paper explores the feasibility of such co-processing via catalytic cracking. The text also provides a list of abbreviations commonly used in this field, such as FCC (fluid catalytic cracking), VGO (vacuum gas oil), HDO (hydrodeoxygenation), and others related to pyrolysis liquids and refining processes. This passage focuses on bio-oil, also called biomass flash pyrolysis liquids (BFPL), as a bio-based liquid feedstock for co-processing in conventional petroleum refineries. Key points include:

Bio-oil is produced in high yields through biomass fast pyrolysis. The quality and yield of bio-oil depend strongly on process variables such as temperature, heating rate, and rapid vapor quenching to avoid secondary cracking into lighter products. Various reactor types have been developed to maximize bio-oil yield, including bubbling fluidized beds, circulating fluidized beds (CFB), transported beds, and rotating cone reactors. Conventional pyrolysis liquids have been used for heat and power generation in boilers, furnaces, and sometimes diesel engines. Bio-oil is a valuable source of renewable chemicals but cannot be used directly as a transportation fuel due to:

High oxygen content (40–50%) High water content (15–30%) Limited stability High acidity. Therefore, bio-oil requires upgrading before it can be used as a transportation fuel. This passage discusses two main downstream upgrading routes for bio-oil and also the use of vegetable oil as a feedstock for biofuel production through hydro processing:

Bio-oil upgrading: Two primary routes are investigated: hydrodeoxygenation (HDO) and catalytic cracking (FCC). HDO involves hydrotreating bio-oil over heterogeneous catalysts (CoMo-, NiMo-based) at moderate temperatures (330–450 °C) with high-pressure hydrogen. FCC upgrading uses mainly zeolite Y and ZSM-5 catalysts in micro-activity test (MAT) reactors. Both methods face significant challenges such as reactor plugging, rapid catalyst deactivation, and low liquid yields, largely due to bio-oil's tendency to polymerize even under mild heating. The paper suggests a process scheme for co-processing bio-oil in refineries and presents experimental results.

Vegetable oil as co-feedstock: Vegetable oil is renewable and commonly used for biodiesel production via transesterification, but biodiesel economics depend heavily on glycerol by-product pricing. The paper investigates biofuel production by hydrotreating vegetable oil, which parallels petroleum feedstock processing. Hydro processing is a familiar petrochemical technology for upgrading heavy hydrocarbons and is emerging as a biofuel production method. Hydro treating vegetable oils produces straight-chain alkanes (n-C15 to n-C18) with high cetane numbers and good cold flow properties. Neste Oil has commercialized diesel production from vegetable oil via modified hydrotreating. Scale-up challenges for co-hydro processing include managing acidity and corrosion in industrial reactors.

This section describes the biomass catalytic pyrolysis (CP) experimental unit used in the study: The CPERI biomass catalytic pyrolysis pilot plant is a small-scale, fully automated system. It consists of:

Biomass feed section: Includes a 4-liter cylindrical hopper and a screw feeder to control biomass flow into the reactor.

Solid feed section: Has a 30-liter fluidized bed regenerator vessel.

The regenerator regenerates spent catalyst and supplies heat for the pyrolysis reactions.

It is connected to the reactor via a heated transfer line with a slide valve to control catalyst flow.

Reactor: Comprises an injector and a riser section.

The injector promotes direct mixing of hot catalyst with biomass particles.

The riser is a vertical tube 5 meters long with 6.2 mm internal diameter.

Gases and solids exiting the riser enter the cyclonic head of a stripper, which removes solids.

This setup is designed to study catalytic pyrolysis of biomass, focusing on efficient catalyst regeneration, heat supply, and product separation. If you want, I can explain how this design impacts the pyrolysis process or provide more details on any unit component.

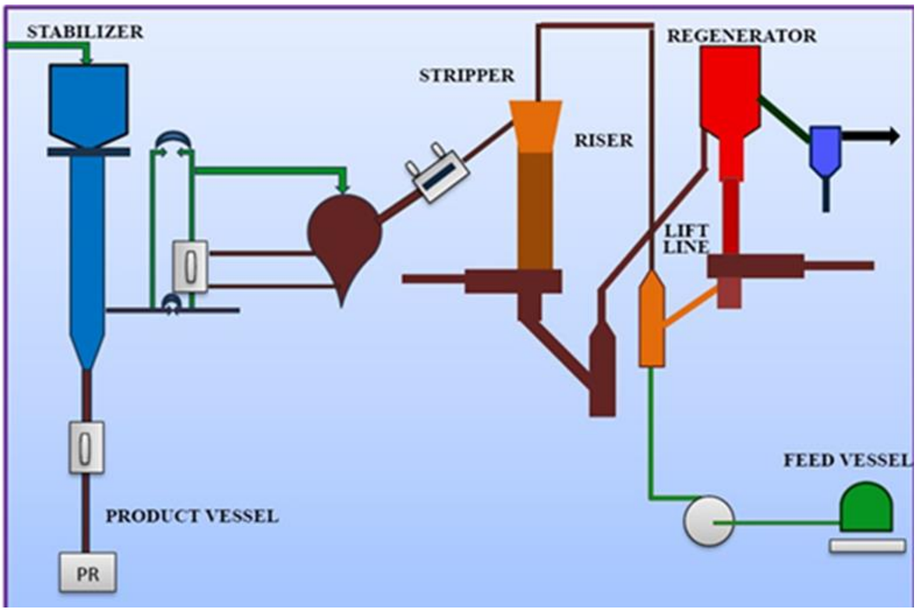

Figure 1: CPERI FCC small scale pilot Plant

Fig.1. This section describes the CPERI FCC small-scale pilot plant unit: The unit operates in full catalyst circulation mode with continuous catalyst regeneration.

Key components include: Riser reactor: 7 mm internal diameter (ID) and about 9 meters in height. Stripper vessel. Lift-line. Fluidized bed regenerator system with 78 mm ID. Catalyst circulation is controlled by two slide valves, similarly to commercial FCC units. Independent temperature control is maintained over multiple reactor zones, allowing isothermal operation.

Steam is injected at the bottom of the stripper to aid catalyst stripping. Gas and liquid products are separated using a specially designed refrigerated stabilizer: Liquid products (mainly C5 and heavier hydrocarbons) are condensed and collected. The pilot plant is fully automated with a process control system based on industrial standards. Key measurements during FCC tests include yields of coke, flue gas, cracked gases, and total liquid products. Liquid products are further characterized into gasoline, light cycle oil (LCO), and heavy cycle oil (HCO) yields using GC simulated distillation (ASTM D2887 method).

The analysis also includes characterization of produced cracked gases and flue gases.

This detailed setup mimics commercial FCC operation while allowing controlled experimental investigation of catalyst performance and product distribution. Let me know if you want more details on any aspect or about the FCC process itself.

This section describes the CPERI hydro processing small-scale pilot plant unit used for hydrotreating (HDS, HDN) and hydrocracking of various feedstocks, from light gasoil to heavy vacuum gas oil (VGO):

The unit consists of three main parts:

Feed system: Maintains constant feed quality and hydrogen-to-oil ratio using a liquid feed pump and gas flow controllers.

Reactor system: Features a single fixed-bed reactor (70 cm length, 14.7 mm internal diameter) with six independent heating zones to maintain the desired temperature profile.

Product separation system: The reactor effluent is first cooled. Then it passes through a high-pressure low-temperature (HPLT) separator where gas and liquid products are separated. Gas product flow is controlled by a pneumatic valve to regulate system pressure. Liquid product flow is controlled by a level control system and a second pneumatic valve after the HPLT separator. Liquid products are collected for analysis in the CPERI laboratory. Important liquid product properties analysed include density, total sulphur, total nitrogen, carbon, hydrogen, bromine number, and simulated distillation (SimDist). Gas products undergo offline gas chromatography (GC) analysis to monitor gas quality consistency.

This hydro processing unit is designed for flexible testing of feedstocks and precise control of reaction conditions, enabling detailed characterization of both liquid and gaseous products. Let me know if you want more explanation on any part or the analysis techniques used.

Experimental results and discussion

This section details the materials used for the biomass catalytic pyrolysis experiments:

Biomass feedstock: Forestry residue from beechwood. Composition on a dry basis (weight %): Carbon (C): 49.41% Hydrogen (H): 6.73% Nitrogen (N): 0.16% Oxygen (O, by difference): 42.96% Ash content: 0.54% Moisture content: 8.25% Gross heating value (GHV): 18.22 MJ/kg Heat carrier medium for non-catalytic pyrolysis: Silica sand Particle size: 100–250 µm Bulk density: 1.56 g/ml Catalyst for catalytic pyrolysis: Equilibrium FCC catalyst Particle size: 50–180 µm Bulk density: 0.96 g/ml Total surface area (TSA): 178.4 m²/g Zeolite area (ZA): 58.5 m²/g Unit cell size (UCS): 24.26 Å Nickel (Ni) content: 150 ppm Vanadium (V) content: 367 ppm

This information provides the foundational materials characterization important for understanding the pyrolysis process and catalyst performance. Let me know if you want further explanation on any of these parameters or their relevance.

Table No.1: Biomass fast pyrolysis results with two different heat carriers (S/B = 16)

S.N.		T = 450ºC Silica sand	FCC	T = 500ºC Silica sand	FCC
1.	Raw product yields (wt% on biomass)				
	BIO-oil yield	73.5	49.2	74.1	46.6
	Gas yield	11.2	17.6	13.5	23.5

	Char yield	10.5	23.1	7.2	22.5
	Raw mass Balance	95.2	90	95	92.3
2.	Individual liquid yields (Wt% on biomass)				
	Organic	56	18.4	57.4	16.2
	Water	17.9	30.9	16.6	30.4
3.	Individual gas yield (Wt% on biomass)				
	CO_2	7	8.6	7.2	9.9
	CO	3.8	8.0	5.8	11.1
	C_1+C_2s	0.4	0.6	0.9	1.3
4.	Wt% on organic bio-oil				
	Hydrocarbons	5.1	7.5	5.4	7.6
	Heavy oxygenates	36.1	18.2	30.3	11.1
	Unidentified	23.4	12.8	19.9	20.1

This section presents the results of biomass catalytic pyrolysis experiments: Experiments were conducted at two pyrolysis temperatures: 450 °C and 500 °C. The solid-to-biomass ratio was about 16, meaning the heat carrier feed rate was 16 times the biomass feed rate. Nitrogen flow rate in the riser reactor was 4 litres /min, giving a vapor residence time of about 1 second. Biomass feed rate was 0.6 kg/h.

Key Findings

Using silica sand as heat carrier (non-catalytic pyrolysis), total liquid product yields ranged from 65% to 78% (wt% on biomass feed basis), among the highest reported for fluidized bed pyrolysis units.

High liquid yields are attributed to: Rapid mixing of biomass with heat carrier. Short vapor residence time in the riser. Rapid vapor quenching in the product recovery section.

Comparing conventional pyrolysis (silica sand heat carrier) with catalytic pyrolysis (using FCC catalyst as heat carrier): Liquid production decreases when FCC catalyst is used. Gas and coke/char production increase significantly with the FCC catalyst. These trends hold true at both 450 °C and 500 °C.

The presence of FCC catalyst strongly promotes secondary reactions that cause bio-oil decomposition, leading to less liquid product and more gas and solid residues.

If you want, I can help analyse the implications of these results or assist with interpreting specific data from Table 1

This detailed analysis from Table 1 and related discussion highlights the effects of temperature and catalyst type on biomass pyrolysis product yields and bio-oil characteristics:

At 450 °C with silica sand as heat carrier: Gas yield: 11.1 wt% (on biomass basis) Char yield: 10.4 wt% Increasing temperature to 500 °C: Gas yield increases Char yield decreases This is mainly due to higher conversion rates of primary pyrolysis reactions producing less char, rather than secondary cracking.

With FCC catalyst present: Temperature increase effects are more pronounced, especially for gases. Oxygen-containing gases dominate: CO and CO2.At 450 °C with silica sand, CO2 yield is highest (6.9%), CO second (3.8%). At 500 °C, CO2 remains about the same, but CO increases significantly to ~6% with silica sand, and up to ~10% with FCC catalyst.

Explanation

CO2 mainly comes from decomposition of carboxyl groups, which happens at lower temperatures.CO is formed from cracking more stable carbonyl groups, requiring higher temperatures. FCC catalyst enhances decarbonylating reactions, favouring CO production. Catalysts strongly affect bio-oil quality: GC/MS analysis shows FCC-catalysed bio-oil contains more hydrocarbons and fewer heavy oxygenates than thermal bio-oil. This is due to cracking of heavy compounds into lighter ones via catalyst acidity and deoxygenation. However, these reactions produce mainly water, so catalytic bio-oil has much higher water content (~60 wt%) compared to thermal bio-oil (~22–24 wt%).

Water content strongly influences physical properties: Density: thermal bio-oil ~1.19 g/cm³, catalytic bio-oil ~1.0 g/cm³.Micro-carbon residue (MCRT) is lower with catalyst, indicating fewer heavy oxygenates. Higher water content lowers the gross heating value (GHV).

Analysis

FCC catalyst changes both yield and quality of bio-oil, favouring water and coke formation. Because FCC catalyst is very active and produces high water and coke, it is likely not the ideal catalyst for bio-oil production by catalytic pyrolysis. More suitable catalysts need to be explored for optimized bio-oil yield and quality.

If you want, I can help summarize this or assist with more details on catalyst development or bio-oil upgrading challenges.

Table 2Physical properties of bio-oils produced from silica sand and FCC catalyst (T = 450 8C, solid/biomass = 16)

S.N.	Property	Silica sand	FCC
1.	Density at 15.5 ^{0}c (g/cm^3)	1.19	1.0
2.	Viscosity at 50 ^{0}c (cSt)	6.4	1.1
3.	Viscosity at 50 ^{0}c after heating for 6 h at 80 ^{0}c (cSt)	8.6	2.6

4.	Viscosity at 50 ^{0}c after heating for 24 h at 80 ^{0}c (cSt)	10.6 3.6	10.6 3.6
5.	Conradson carbon residue (MCR) (wt%)	16.3 1.7	16.3 1.7
6.	Pour point (^{0}C)	-34	20
7.	Flash point (^{0}C)	63	38
8.	HHV (Mj/kg)	60.4	8

Table 3Properties of the two fractions produced after thermally hydrotreating the bio-oil

S.N.	Propertyfraction	Light	Heavy
1.	Element analysis (wT%)		
	C	82.3	84.5
	H	10.8	9.3
	S	0.02	0.02
	N	1.16	0.41
	O	6.5	5.0
	H2O (wt%)	0.98	-
	Density (g/cm3)(15 0C)	.932	1.036
2.	Distillation (0C, wt%)		
	<250	28	
	250-400	55.4	24
	400-550	17.6	65
	>550		12

This section discusses the upgrading of biomass fast pyrolysis liquids (BFPLs) into transportation fuels using hydrotreating technology studied by CPERI in collaboration with vebaoil. Key points include:

Two hydrotreating modes were tested: Catalytic hydrogenation using conventional hydrotreating catalysts. Thermal hydrogenation without any catalyst.

Catalytic hydrogenation: Achieved very high de-oxygenation conversion (>85 wt%). Faced significant operational problems such as catalyst bed plugging.

Thermal hydrogenation: Proven feasible without operational issues. Achieved up to 85 wt% de-oxygenation conversion. Produced hydrotreated bio-oil with oxygen content around 6.5 wt%. Yield of hydrotreated bio-oil was about 42 wt% (based on original bio-oil).

The thermally hydrotreated bio-oil (with low oxygen content) can be separated by distillation into two fractions:

Light fraction (LBFPL): Contains components mainly in the gasoline and diesel range and can be directly blended with corresponding petroleum fractions.

Heavy fraction (HBFPL): Has characteristics similar to conventional vacuum gas oil (VGO); can be co-fed with VGOs in FCC units, acting as a resid component in refinery feedstocks. CPERI proposes a bio-oil co-processing technology incorporating these principles, schematically shown in Fig. 4.

This passage describes experimental work in the CPERI FCC small-scale pilot plant to test co-processing of the thermally hydrotreated heavy bio-oil fraction (HBFPL) with conventional vacuum gas oil (VGO): The heavy fraction of thermally hydrotreated bio-oil (HBFPL) was used as co-feed with VGO in the FCC unit. A main challenge with such feedstocks is nozzle plugging in the pilot plant. To prevent plugging, HBFPL was first diluted with light cycle oil (LCO) in a 15:75 weight ratio. This LCO+HBFPL mixture was then blended with conventional FCC feed VGO, making up 15 wt% of the total feed. For comparison, baseline tests used VGO + 15% LCO (without bio-oil). Experiments were conducted under: Isothermal riser operation at 520 °C. Partial pressure of hydrocarbons in the riser: 12 psia. Nitrogen flow rate: 3500 cc/min. Vapor residence time in riser: 0.8 seconds.

The catalyst used was an equilibrium catalyst (Ecat) from a Greek refinery with: Total surface area (TSA): 158 m^2/g. Rare earth content (RE): 0.65 wt%. Nickel (Ni): 163 ppm. Vanadium (V): 362 ppm. Catalyst circulation and operational performance were satisfactory, with no nozzle plugging observed during bio-oil injection. Overall mass balances for tests ranged between 98% and 101%, indicating good material accounting.

This shows that co-processing the heavy bio-oil fraction in FCC, with appropriate dilution, can be operationally feasible at pilot scale without plugging issues

The experimental results from co-feeding thermally hydrotreated heavy bio-oil fraction (HBFPL) with vacuum gas oil (VGO) in the FCC pilot plant reveal the following key points:

Product yields: The VGO/HBFPL co-feed produces about 1 wt% more gasoline and more light cycle oil (LCO) compared to VGO alone. This indicates that components from the hydrotreated bio-oil favour cracking into gasoline and diesel-range products. Coke yield increases by about 0.5 wt% with the co-feed, which is considered manageable within typical FCC operation. Conversely, liquefied petroleum gas (LPG) yields decrease when bio-oil is added.

Gasoline quality: PIONA analysis (not shown) indicates that gasoline from bio-oil co-feed contains more aromatics and fewer paraffins and olefins than gasoline from VGO alone.

Feedstock crackability: The presence of HBFPL reduces overall feedstock crackability, resulting in about 1 wt% lower conversion at the same catalyst-to-oil (C/O) ratio. This is attributed to heavy bio-oil components that are harder to crack and require more catalyst.

Overall assessment: Despite a slight reduction in conversion and increased coke, the selectivity towards valuable liquid products improves. Co-feeding thermally hydrotreated bio-oil with petroleum VGO is technically feasible in FCC units processing high-quality feedstocks with sufficient coke-burning capacity.

This section describes the study of co-hydro processing vacuum gas oil (VGO) and sunflower oil mixtures as a potential biofuel production technology:

Feedstocks: Four feedstocks were tested:

Pre-hydrotreated VGO (HDT-VGO) with sunflower oil at 70/30 volume radiopure-hydrotreated VGO with sunflower oil at 90/10 ratio. Straight run (not hydrotreated) VGO with sunflower oil at 70/30 ratio. Straight run VGO with sunflower oil at 90/10 ratio. Properties of these feedstocks are summarized in Table 4. For feedstocks containing hydrotreated VGO, additives dimethyl-disulfide (DMDS) and tetra-propyl-amine (TPA) were added to regulate catalyst activity, compensating for low sulphur and nitrogen levels after pre-hydrotreatment.

Hydro processing conditions: Experiments were run under identical conditions simulating commercial hydrotreating: Temperature (T): 350 °C, Pressure (P): 2000 psig, Liquid hourly space velocity (LHSV): 1.5 h^{-1}, Liquid feed (FL): 20 ml/h, Gas flow (FG): 0.75 scfh. A noble metal mild-hydrocracking catalyst was used for all experiments.

Product analysis: Once steady state was reached, the liquid products were analysed for: Density Carbon, hydrogen content Total sulphur, total nitrogen Bromine number Simulated distillation

Oxygen content was estimated by difference (100% minus C, H, S, and N percentages).

This setup allows evaluation of the effect of sunflower oil blending and feedstock pre-treatment on hydro processing performance and product quality.

The passage explains that co-hydro processing of VGO–sunflower oil mixtures leads to several key chemical changes, including saturation of unsaturated bonds, removal of heteroatoms (mainly sulphur, nitrogen, and oxygen), as well as isomerization and cracking reactions.

Specifically, the hydrogen-to-carbon (H/C) ratio changes observed (shown in Fig. 8) reveal: Feedstocks containing pre-hydrotreated VGO (HDT-VGO) mixed with sunflower oil have higher H/C ratios initially because the fossil component (HDT-VGO) was already hydrotreated. The increase in H/C ratio during co-hydro processing is more pronounced in mixtures with higher sunflower oil content (70/30 ratio), suggesting that the hydro processing primarily affects the heavy hydrocarbon molecules present in the sunflower oil.

In essence, the bio-component (sunflower oil) undergoes significant upgrading during co-hydro processing, improving saturation and reducing heteroatoms more notably when present in higher proportions.

This passage summarizes heteroatom removal results from co-hydro processing VGO–sunflower oil feedstocks:

Feed properties: All four feedstocks have similar properties except oxygen content, which is higher in the two feedstocks with more sunflower oil (70/30 ratio).

Heteroatom removal effectiveness: Oxygen removal is more efficient in feedstocks containing pre-hydrotreated VGO plus sunflower oil compared to those with non-treated VGO.

Sulphur removal occurs only in the pre-hydrotreated VGO + sunflower oil feedstocks. In contrast, sulphur removal is negligible in non-treated VGO + sunflower oil feedstocks. This is expected because sulphur in non-treated VGO exists as mercaptans, sulphides, benzothiophenes, and thiophenes, which require specialized catalysts for hydrodesulfurization (HDS) and hydrodenitrogenation (HDN).

Nitrogen removal remains relatively unchanged across all feedstocks.

This passage summarizes the hydrocracking conversion results from the four co-hydro processing experiments (shown in Fig. 10): The hydrocracking conversion of VGO–sunflower oil mixtures is lower than the typical hydrocracking conversion of conventional VGO alone, which is around 60–80%.

Higher sunflower oil content (70/30 ratio) in the feedstock further decreases hydrocracking conversion, indicating that sunflower oil inhibits hydrocracking.

Hydrocracking conversion is higher for feedstocks containing pre-hydrotreated VGO mixed with sunflower oil compared to those with non-treated VGO.

In essence, sunflower oil reduces hydrocracking efficiency, but pre-hydrotreated VGO helps improve conversion relative to non-treated VGO in these mixtures.

Conclusions

Biomass catalytic pyrolysis with FCC catalyst: The FCC catalyst lowers bio-oil yield but improves bio-oil quality. The process shows significant potential if a more suitable catalyst is developed. Further research is needed to optimize catalyst selection and performance.

Co-processing gas oil with thermally hydrotreated bio-oil: The presence of bio-oil enhances gasoline and diesel production. Coke yield increases but remains within manageable limits. This approach is technically viable for FCC units operating with high-quality feedstocks and sufficient coke-burning capacity.

Co-hydro processing of VGO with sunflower oil: Sunflower oil reduces hydrocracking conversion, especially when non-hydrotreated VGO is used. Despite this, the process is feasible and produces good-quality diesel.

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