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Characterization of light diesel fraction obtained from upgraded heavy oil

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ABSTRACT

In this study, the role of a low-cost catalyst (activated molasses soil) over the hydrocarbon distribution of the light diesel (or distillate) fraction of the pre-upgraded heavy crude oil (Bati Raman) was examined in detail. The low-cost catalyst showed a strong impact on the hydrocarbon distribution of the light diesel fuel fraction. The physicochemical characteristics such as hydrocarbon group distribution, density and kinematic viscosity of the light diesel fuels obtained from the upgraded heavy oil indicated a proper consistency with those having the commercial diesel fuels. As a result, it was revealed that the low-cost material could be successively used in the heavy oil upgrading to obtain the light diesel fuels.

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1. Introduction

Even today, the crude oil and its derivatives have still attracted great interest in the world due to its potential of global energy supply at a significant ratio of 32% [1,2]. Furthermore, it was envisaged that the interest trend would increase in the near future due to the high demand for primary energy and various petroleum products as the result of improvements in industrialization and in the global population growth as well [3]. The great amount of the primary energy demand is fulfilled using petroleum fuels in different physical forms. Today, the light oil can be successively refined into various liquid and gaseous fuels. However, after a period of time stated as the year 2035, the existence reserves of the light oils may not be sufficient to supply the required demand unless these reserves are replaced with various alternative energy resources [2]. In this regard, the exploitation of heavy crude oil and its derivatives has become prominent because of the total reserve quantity about 9–13 trillion barrels [4]. However, they cannot be refined into synthetic light crude oil, or light and medium crude oil with-

out being exposed to any kind of suitable upgrading processes in their actual forms [5]. The reason is that they have undesired inherent characteristics such as high density and viscosity, low API gravity, high contents of resins and asphaltenes, and high metal contents of Ni and V as well [6]. Furthermore, these properties lead to major challenges such as clogging of pipes, high-pressure drop, break in production [7] during the processing stages conducted in terms of exploitation [8]. Therefore, these key parameters such as viscosity, density, and chemical composition belong to a liquid energy source are assessed in terms of determining its quality and its various processes including transportation, upgrading and refining [9].

Following the heavy crude oils being upgraded by means of proper technologies such as coking, cracking, visbreaking, and hydro conversion [10], they can be refined into a marketable product such as fuels and petrochemicals [7]. Recently, the cracking processes (thermal and/or catalytic) among these are widely performed for the conversion of heavy oils into light oils and various value-added products (gasoline, diesel, etc.). The products such as gasoline and diesel are mainly composed of small hydrocarbon molecule groups [11], therefore, the upgraded oils (lower viscosity) obtained from the thermal cracking of heavy crude oil can be converted into various products consisting the hydrocarbon molecules with smaller chain length by applying catalytic cracking.

Diesel fuels are the mixtures of various hydrocarbons (alkanes, naphthene, olefins, and aromatics) having carbon numbers of C₉–C₂₇ [12]. Moreover, some of the important physicochemical

Abbreviations: API, American Petroleum Institute; TULD, thermal upgrading light diesel; CULD, catalytic upgrading light diesel; ASTM, American Society for Testing and Materials; GC–MS, gas chromatography–mass spectrometry; EN 590, European Committee's standard related to diesel fuels; C, carbon; H, hydrogen; cSt, centistokes; CaO, calcium oxide; V, vanadium; Ni, nickel.

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properties representing the quality of diesel fuels are sulfur and aromatic contents, distillation curve, density, viscosity, cetane number (cetane index), etc. [12,13].

The study was initially aimed at enabling the light diesel (distillate) fuel from the thermal upgraded oil of Bati Raman heavy crude oil by means of catalytic cracking using low-cost material. Then, these fuels were characterized according to the essential fuel parameters such as hydrocarbon group distributions, API gravity, density, and viscosity in terms of evaluating the catalyst effect over the light diesel fuels (thermal and catalytic).

2. Materials and methods

The light diesel fuel-boiling range 240–290 °C was obtained by cracking the pre-upgraded oil of Bati Raman heavy crude oil by means of experimental apparatus stated in the previous study [14]. In catalytic cracking process, the thermally activated molasses soil (at 900 °C for 6 h) was used as a low-cost catalyst. The catalytic cracking process was carried out on a nearly 110 g of pre-upgraded oil and at optimal catalyst ratio of 10.0 wt%. The loaded reactor was placed into the electrical heating mantle and it was heated. The temperature of organic vapor rising from the reactor was monitored by using a thermometer. During each cracking run, the light diesel fuel boiling point range of 240–290 °C was collected and dehydrated over anhydrous sodium sulfate. Finally, the dehydrated oil was kept in a tightly sealed sample container.

The obtained light diesel fuel was labeled as TULD (thermal upgrading light diesel). In addition, the other diesel fuel sample labeled as CULD (catalytic upgrading light diesel) obtained from catalytic upgrading of the first upgraded oil with catalyst ratio of 10 wt%. Lastly, the physicochemical properties of diesel samples (TULD and CULD) were characterized according to the relevant ASTM standard test methods (ASTM D 446 and ASTM D 1217). On the other hand, the GC–MS technique was performed on both diesel fuels using the analysis method detailed in the previous study [14].

3. Results and discussion

3.1. Density and viscosity

The yield of the CULD fuel was 26.66 wt% at the catalyst ratio of 10 wt%. It was obvious that the yield was found to be slightly higher than the TULD yield of 26.36 wt%. In addition, the diesel yields obtained in the study were found to be higher than the diesel yield of 22.88 wt% obtained from the distillation of Yemeni petroleum of 31° API gravity [15] and those of others expressed in the study [16]. Furthermore, some of the significant physical properties of both diesel samples were determined and the results were presented in Table 1. As can be seen in Table 1, it was revealed that the values of viscosity and density for the CULD fraction were found to be slightly higher than those of the TULD fraction due

Table 1
Physical properties of the light diesel fuel fractions obtained from thermal and catalytically upgraded oils.

Property	Upgraded oil light diesel fraction (240–290 °C)	
	TULD	CULD
API gravity (°)	28.74	27.78
Density (15.6 °C, g/cm ³)	0.8822	0.8875
Viscosity (40 °C, cSt)	3.7838	4.4043

TULD: thermal light diesel fraction.

CULD: catalytic upgrading light diesel fraction.

to the catalyst on the hydrocarbon distributions of the CULD fraction [17]. On the other hand, the viscosity values of 3.7838 and 4.4043 cSt at 40 °C and the density values of 0.8822 and 0.8875 g/cm³ at 15.6 °C were found for the TULD and CULD fuels, respectively. According to the data, the obtained diesel fuels were considerably in compliance with the requirements of EN 590 Standard in terms of the viscosity of 2.0–4.5 mm²/s and the density of 820–845 kg/m³ [18].

3.2. Distribution of hydrocarbon group types

The fuels were exposed to GC–MS analysis in accordance with the defined method in the previous study [14] order to evaluate

Table 2
Hydrocarbon distribution of the light diesel (TULD) fraction.

Hydrocarbon name and its group	Formula	Retention time (min)	Area %
Paraffins (n- and iso- alkanes)			
Nonane	C ₉ H ₂₀	4.33	0.30
Octane, 2,6-dimethyl-	C ₁₀ H ₂₂	4.42	0.16
Decane	C ₁₀ H ₂₂	4.87	0.48
Octane, 3,5-dimethyl-	C ₁₀ H ₂₂	4.94	0.35
Decane, 2-methyl-	C ₁₁ H ₂₄	5.37	0.32
Decane, 3-methyl-	C ₁₁ H ₂₄	5.49	0.31
Undecane	C ₁₁ H ₂₄	5.88	1.56
Undecane, 3-methyl-	C ₁₂ H ₂₆	6.89	0.16
Dodecane	C ₁₂ H ₂₆	7.53	1.18
Dodecane, 2,6,10-trimethyl-	C ₁₅ H ₃₂	8.66	0.66
Tridecane	C ₁₃ H ₂₈	9.80	1.69
Tetradecane	C ₁₄ H ₃₀	12.44	1.73
Pentadecane	C ₁₅ H ₃₂	15.06	3.39
Hexadecane	C ₁₆ H ₃₄	17.81	4.17
Heptadecane	C ₁₇ H ₃₆	20.30	3.44
Hexadecane, 2,6,10,14-tetramethyl-	C ₂₀ H ₄₂	22.29	1.27
Octadecane	C ₁₈ H ₃₈	22.69	3.00
Nonadecane	C ₁₉ H ₄₀	25.38	1.36
Tricosane	C ₂₃ H ₄₈	37.98	0.93
Tetracosane	C ₂₄ H ₅₀	40.79	0.77
Pentacosane	C ₂₅ H ₅₂	44.34	0.38
	Σ yield		27.61
Naphthenes (cycloalkanes or cycloparaffins)			
Cyclooctane, 1,2-dimethyl-	C ₁₀ H ₂₀	5.07	0.13
Cyclopropane, 1-ethyl-2-pentyl-	C ₁₀ H ₂₀	5.23	0.16
Cyclopropane, nonyl-	C ₁₂ H ₂₄	6.47	0.22
Cyclooctane, 1,2-diethyl-	C ₁₂ H ₂₄	7.22	0.20
Cyclododecane	C ₁₂ H ₂₄	8.35	0.33
Cyclotetradecane	C ₁₄ H ₂₈	10.20	0.16
Cyclopentadecane	C ₁₅ H ₃₀	16.03	0.59
Cyclotetradecane, 1,7,11-trimethyl-4-(1-methylethyl)-	C ₂₀ H ₄₀	24.06	0.53
	Σ yield		2.32
Aromatics			
Benzene, 1,2,3-trimethyl-	C ₉ H ₁₂	9.47	0.24
Benzene, 1,2,4,5-tetramethyl-	C ₁₀ H ₁₄	14.70	0.92
Naphthalene, 1,2,3,4-tetrahydro-1,1,6-trimethyl-	C ₁₃ H ₁₈	19.40	0.93
Naphthalene, 2,7-dimethyl-	C ₁₂ H ₁₂	27.38	0.36
Naphthalene, 2,6-dimethyl-	C ₁₂ H ₁₂	28.35	0.65
Naphthalene, 2-(1-methylethyl)-	C ₁₃ H ₁₄	30.79	0.25
Naphthalene, 1,4,5-trimethyl-	C ₁₃ H ₁₄	32.89	1.57
Naphthalene, 1,4,6-trimethyl-	C ₁₃ H ₁₄	35.98	0.60
Chamazulene (Azulene, 7-ethyl-1,4-dimethyl-)	C ₁₄ H ₁₆	38.35	0.34
Benzene, 1,3,5-tris(1-methylethyl)-	C ₁₅ H ₂₄	42.59	0.18
	Σ yield		6.04
Olefins			
1-Nonadecene	C ₁₉ H ₃₈	10.76	0.34
1-Hexadecene (or Cetene)	C ₁₆ H ₃₂	18.51	0.43
1-Heptadecene	C ₁₇ H ₃₄	21.10	0.25
1-Octadecene	C ₁₈ H ₃₆	23.57	0.56
	Σ yield		1.58

the role of the catalyst on the hydrocarbon composition of both TULD and CULD. The fact that whether a typical fuel is diesel is directly associated with its number of carbon range and its content of hydrocarbon class [19]. The identified hydrocarbon compounds for the TULD and CULD fuels were presented in Table 2 and Table 3 respectively.

The GC–MS results revealed that the paraffinic hydrocarbons were dominant in both the TULD fuel and the CULD fuel when compared with the other hydrocarbons. In addition, the *n*-paraffinic groups were also found to be more supreme than the other paraffinic compounds (see Table 5) identified in these fuels. Besides, the hydrocarbons with the highest concentration (>1%) were determined for the TULD fuel as hexadecane (C₁₆H₃₄, 4.17%), heptadecane (C₁₇H₃₆, 3.44%), pentadecane (C₁₅H₃₂, 3.39%), octadecane (C₁₈H₃₈, 3.00%), tetradecane (C₁₄H₃₀, 1.73%), tridecane (C₁₃H₂₈, 1.69%), undecane (C₁₁H₂₄, 1.56%), nonadecane (C₁₉H₄₀, 1.36%), dodecane (C₁₂H₂₆, 1.18%), hexadecane, 2,6,10,14-tetramethyl- (C₂₀H₄₂, 1.27%), and naphthalene, 1,4,5-trimethyl-

(C₁₃H₁₄, 1.57%) respectively. The hydrocarbons with the highest concentration (>1%) were determined for the CULD fuel were heptadecane (C₁₇H₃₆, 4.04%), pentadecane (C₁₅H₃₂, 3.26%), hexadecane (C₁₆H₃₄, 3.09%), tetradecane (C₁₄H₃₀, 1.74%), tridecane (C₁₃H₂₈, 1.70%), dodecane (C₁₂H₂₆, 1.19%), benzene, 1,2,4,5-tetramethyl- (C₁₀H₁₄, 1.15%), and naphthalene, 1,4,5-trimethyl- (C₁₃H₁₄, 1.11%) respectively. As the result, it was observed that the TULD fuel was found to be rich with the content of *n*-alkane compound having carbon number of C₁₆ while the CULD fuel was found to be richer in terms of *n*-alkane of C₁₇. Moreover, each fuel was found

Table 3
Hydrocarbon distribution of the light diesel (CULD) fraction.

Hydrocarbon name and its group	Formula	Retention time (min)	Area %
Paraffins (<i>n</i>- and iso- alkanes)			
Nonane	C ₉ H ₂₀	4.33	0.26
Decane	C ₁₀ H ₂₂	4.87	0.43
Undecane	C ₁₁ H ₂₄	5.87	0.99
Undecane, 3-methyl-	C ₁₂ H ₂₆	6.88	0.17
Dodecane	C ₁₂ H ₂₆	7.53	1.19
Tridecane	C ₁₃ H ₂₈	9.79	1.70
Tetradecane	C ₁₄ H ₃₀	12.39	1.74
Pentadecane	C ₁₅ H ₃₂	15.24	3.26
Hexadecane	C ₁₆ H ₃₄	17.80	3.09
Heptadecane	C ₁₇ H ₃₆	20.13	4.04
Hexadecane, 2,6,10,14-tetramethyl-	C ₂₀ H ₄₂	22.30	0.93
Octadecane	C ₁₈ H ₃₈	22.68	2.52
Nonadecane	C ₁₉ H ₄₀	25.36	1.57
Eicosane	C ₂₀ H ₄₂	29.03	0.34
Heneicosane	C ₂₁ H ₄₄	33.03	0.44
Tricosane	C ₂₃ H ₄₈	37.97	0.82
Pentadecane	C ₁₅ H ₃₂	40.78	0.73
Hexadecane	C ₁₆ H ₃₄	44.27	0.39
		Σ yield	24.61
Naphthenes (cycloalkanes or cycloparaffins)			
Cyclooctane, 1,2-dimethyl-	C ₁₀ H ₂₀	5.07	0.13
Cyclopropane, 1-ethyl-2-pentyl-	C ₁₀ H ₂₀	5.22	0.16
Cyclododecane	C ₁₂ H ₂₄	8.34	0.25
Cyclopentadecane	C ₁₅ H ₃₀	12.04	0.17
Cyclopentadecane	C ₁₅ H ₃₀	16.01	0.55
Cyclotetradecane	C ₁₄ H ₂₈	17.38	0.44
Cyclotetradecane, 1,7,11-trimethyl-4-(1-methylethyl)-	C ₂₀ H ₄₀	24.06	0.62
		Σ yield	2.32
Aromatics			
Benzene, 1,2,3-trimethyl-	C ₉ H ₁₂	9.45	0.25
Benzene, 1,2,4,5-tetramethyl-	C ₁₀ H ₁₄	14.68	1.15
Benzene, hexamethyl-	C ₁₂ H ₁₈	27.02	0.23
Naphthalene, 2,7-dimethyl-	C ₁₂ H ₁₂	27.35	0.55
Naphthalene, 2,6-dimethyl-	C ₁₂ H ₁₂	28.34	0.58
Naphthalene, 2-(1-methylethyl)-	C ₁₃ H ₁₄	30.77	0.24
Naphthalene, 1,4,5-trimethyl-	C ₁₃ H ₁₄	32.88	1.11
Naphthalene, 1,4,6-trimethyl-	C ₁₃ H ₁₄	35.96	0.62
Benzene, 1,3,5-tris(1-methylethyl)-	C ₁₅ H ₂₄	42.57	0.14
		Σ yield	4.87
Olefins			
5-Octadecene, (E)-	C ₁₈ H ₃₆	6.46	0.21
1-Heptadecene	C ₁₇ H ₃₄	21.08	0.26
1-Octadecene	C ₁₈ H ₃₆	23.57	0.45
1-Nonadecene	C ₁₉ H ₃₈	26.59	0.51
		Σ yield	1.43

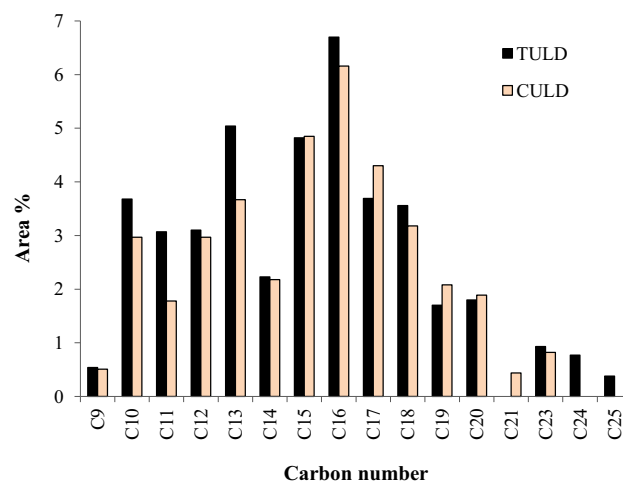


Fig. 1. Carbon number distribution of the TULD and CULD fuel.

Table 4
Carbon range distributions of the light diesel fuel fractions (TULD and CULD).

Carbon number range distributions	Area %	
	TULD	CULD
C ₆ –C ₁₂	10.39	8.23
C ₁₃ –C ₁₆	18.79	16.86
C ₁₇ –C ₂₀	10.75	11.45
C ₂₁ –C ₃₀	2.08	1.26
C ₈ –C ₂₅	42.01	37.80

TULD: thermal upgrading light diesel fraction.
CULD: catalytic upgrading light diesel fraction.

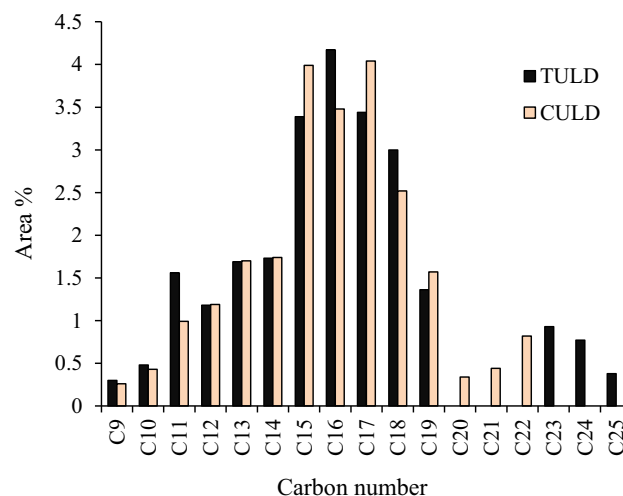


Fig. 2. Carbon number distribution of *n*-paraffins in the TULD and CULD fuel.

Table 5
Hydrocarbon groups of the light diesel fuel fractions (TULD and CULD).

Hydrocarbon group types	Area %	
	TULD	CULD
n-paraffins	24.38	23.51
iso-Paraffins	3.23	1.10
Cycloparaffins	2.32	2.32
Monoaromatics	1.34	1.77
Polyaromatics	4.70	3.10
Olefins	1.58	1.43

TULD: thermal upgrading light diesel fraction.

CULD: catalytic upgrading light diesel fraction.

to be rich in content in terms of the *n*-paraffinic compounds with carbon number in range of C₁₅–C₁₈ (see Fig. 2). The result of the study was in consistency with the relevant literature regarding stated diesel fuel. [20]. The presence of the *n*-alkanes of C₁₅–C₁₈ at maximum concentration can also be associated with the catalytic effect of CaO in the catalyst [17].

As can be seen in Fig. 1, the carbon number distributions of the hydrocarbons in both diesel fuels in the study were in between C₉ and C₂₅. However, the TULD fuel did not include the hydrocarbons of C₂₁ and C₂₂ and the reason could be the cracking effect of thermal cracking process on longer chain hydrocarbons. Furthermore, the highest concentration of both diesel fuels was found to be at the carbon number of C₁₅–C₁₆. These findings were compatible with other results of the study [21,22]. In addition, the percentage concentration of hydrocarbons with certain carbon number range were given in Table 4. For the CULD fuel, the concentration of 8.23% for the hydrocarbon of C₆–C₁₂, namely as light fraction was found to be quite lower than that of 10.39% for the TULD fuel. This decrease can be attributed to the severe cracking effect of the catalyst over the light hydrocarbon molecules. Moreover, as seen in Fig. 1, the absence of hydrocarbons with carbon number (>C₂₃) in the CULD fuel showed the occurrence of strong catalytic cracking effect over hydrocarbon molecules with long chains and it resulted in a higher yield of C₁₇–C₂₀ (see Table 4). In addition, as shown in Table 5, it was clearly seen that the CULD fuel has lower yield of 4.70% for polyaromatics but higher yield of 3.10% for monoaromatics compared to those of the TULD fuel due to the effect of CaO on the condensed aromatics [17]. According to these results, it was stated that the catalyst with rich content in terms of CaO was found to be quite effective on the hydrocarbon distributions of thermal upgrading light diesel fuel.

4. Conclusions

The main conclusions of the study were shown below:

- The yield of light diesel of the catalytic upgrading heavy oil was found to be higher than that of thermal upgrading diesel fuel and those of others originated with light crude oils.
- Both light diesel fuels of the Bati Raman heavy crude oil were in harmony with the commercial diesel fuels in terms of the carbon number distributions of C₉–C₂₅, the viscosity of 3.8–4.4 cSt at 40 °C, the density of ~0.9 g/cm³ at 15.6 °C, and the rich paraffinic hydrocarbon contents.
- The low-cost catalyst showed a strong cracking effect over the light fraction hydrocarbon molecules (C₆–C₁₂) and heavy fraction compounds with long carbon chain (>C₂₃).

- For both diesel fuels, the *n*-alkanes having the highest concentration occurred in carbon number range of C₁₅–C₁₈. Among these hydrocarbons, hexadecane (C₁₆H₃₄, 4.17%) was stated for the TULD fuel and heptadecane (C₁₇H₃₆, 4.04%) was stated for the CULD fuel.
- The catalyst could be used as a low-cost alternative after further improvements of the current catalyst material by means of chemical activation techniques for upgrading of heavy crude oil and its derivatives to obtain the lighter fuels.

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