

LIQUEFACTION OF BANKO COAL WITH LIMONITE CATALYST

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Abstract

Pengaruh karakteristik batubara terhadap yield produk pencairan batubara telah diinvestigasi menggunakan sampel batubara peringkat rendah dari SUamtera Selatan dan Kalimantan. Pengujian pencairan dilaksanakan menggunakan gas-flow type autoclave (5L) dengan umpan sampel batubara 200g (daf), 400g pelarut turunan (b.p.300-420 °C), 1.0-3.0 wt% daf katalis Fe and elemen sulfur (rasio atom S/Fe 2.0) pada tekanan 14.7MPa, 450°C untuk selama 60min dengan aliran gas H₂ konstan (0.5%H₂S). Ditemukan bahwa yield minyak meningkat dengan peningkatan rasio atom H/C dari batubara, tetapi pengaruh karakteristik batubara terhadap pencairan sangat rumit. Studi ini menunjukkan limoni Soroako (MGL) mempunyai aktifitas pencairan lebih tinggi dibandingkan katalis limonit synthetic □-FeOOH dan Yandi Yellow (YY) karena transformasinya menjadi pyrrhotite (Fe_{1-x}S) Kristal berukuran kecil melalui penambahan elemen sulphur. Namun, ukuran kristal pyrrhotite dari katalis Soroako bertambah dengan mudah dibandingkan kristal dari katalis YY denganr esikel, menghasilkan yieldminyak yang sama melalui penambahan CLB. Yield minyak pencairan batubara Banko mencapai 66.7wt%daf pada ondisi operasi suhu 450°C selama 60min dengan katalis Soroako 1wt%daf sebagai Fe dan 100wt%daf penambahan CLB. Pada proses upgrading produk pencairan, kebanyakan senyawa nitrogen dan sulfur di dalam minyak berhasil dihilangkan melalui proses two-stage hydrotreatment, sehingga terjadi perbaikan stabilitas selama penyimpanan dan mutu produk. Bilangan oktan (RON) minyak ringan dari batubara (75,8) sedikit lebih tinggi dari minyak petroleum (65-70) sedangkan smoke point kerosene lebih rendah (14mm) dibandingkan standard petroleum berkaitan erat dengan komposisi minyak yang kaya aromatis.

Keywords: Soroako, Yandi Yellow, pyrrhotite, hydrotreatment

1. INTRODUCTION

BPPT in cooperation with NEDO, Kobe-steel, Nissho Iwai Corporation, and Mitsubishi Chemical Corporation, has conducted feasibility study on Indonesian brown coal liquefaction from 1994-2003. Parts of the feasibility study, some fundamental research on coal liquefaction have been conducted both in Takasago Laboratory, Japan and Puspipstek, Serpong, Indonesia. Among others are:

- Investigation on the effect of coal properties on the product yields, or the liquefaction reactivity of low-rank coals in Indonesia
- Investigation on the effect of using highly dispersed limonite catalysts for BCL process
- hydro-treatment of liquefied oil tests using a fixed bed reactor with a conventional Ni-

Mo/Al₂O₃ catalyst to improve the storage stability and fuel characteristics of product oils

The study shows that Banko coal from Tanjung-Enim, South Sumatra, has the highest oil yield in coal liquefaction among the tested coals in Indonesia. While the use of highly dispersed limonite catalysts for the improved BCL process has been successfully conducted through the operation of a 0.1t/d Bench Scale Unit (BSU) at TCLC [1-3]. From the economic point of view, natural minerals in Indonesia should be utilized as a catalyst material for the liquefaction of Banko coal. It is very fortunate that a large amount of limonite is deposited on the nickel mine in Soroako, Sulawesi, Indonesia. This Soroako limonite is not utilized yet because of the insufficient nickel content in it. Therefore, nickel containing Soroako limonite can be utilized

as an inexpensive catalyst material, and nickel may be expected to promote the liquefaction activity of iron catalyst in coal liquefaction.

This paper presents the investigation results on the effect of coal properties on the product yields of Banko coal liquefaction. It appears that the oil yield increases with an increase in H/C atomic ratio of coal, but the influences of coal properties on the liquefaction are really complicated. Soroako limonite catalyst showed an excellent catalytic activity because of the transformation into a smaller crystallite size of pyrrhotite (Fe_{1-x}S) that is an active phase in coal liquefaction. Almost nitrogen and sulfur compounds in crude liquefied oil are successfully removed by the two-stage hydrotreatment, resulted in the improvement of the storage stabilities and fuel properties. The liquefaction of Banko coal with Soroako limonite catalyst could be a promising way to obtain clean transportation fuels from low-rank coals in Indonesia.

2. EXPERIMENTAL

1) Coal Samples

Core samples of Central and South Banko Coals are used to clarify the influence of coal properties on the product yields. All coal samples are ground to less than 60 mesh and dried in vacuum at 105°C prior to the use in the elemental analysis and liquefaction test. Both proximate and elemental analyses are carried out in accordance with JIS-M8812. Metal contents in coal are measured with an atomic adsorption spectroscopy method. Petrographic analysis is performed using a microscope to determine the distribution of maserals (Exinite, Vitrinite and Inertinite) and the reflectance (Ro) is measured for Vitrinite.

(2) Limonite Samples

Limonite samples used as catalyst raw materials in this study is from Soroako. An Australian limonite Yandi Yellow, which consists of mainly $\alpha\text{-FeOOH}$, is used for a comparison purpose. Limonite is dried in vacuum at 105°C prior to the use in this study.

(3) Catalyst Preparation

Soroako limonite (SO) is finely pulverized to sub-micron particle size using a ball mill or an agitated beads mill with a coal-derived solvent prior to the use in coal liquefaction as a catalyst. A finely pulverized Yandi Yellow (YY), synthetic $\alpha\text{-FeOOH}$ and $\gamma\text{-FeOOH}$ catalysts are used for a comparison of liquefaction activities. The average particle size of catalyst is determined with a laser diffraction method using a particle analyzer.

(4) Liquefaction Test

Liquefaction tests are carried out mainly using a gas-flow type autoclave (5L) with 200g(daf) of Banko coal, 400g of coal-derived solvent (b.p.300-420 °C), 1.0-3.0 wt% daf as Fe of catalyst and elemental sulfur (S/Fe atomic ratio of 2.0) at 14.7MPa, 450°C for 60min under a constant gas flow of $\text{H}_2\text{-0.5\%H}_2\text{S}$. The liquid products are separated into H_2O , distilled oil ($\text{C}_5\text{- b.p.420 }^\circ\text{C}$) and coal liquid bottom (CLB, b.p.> 420 °C) by a distillation, and the gaseous products ($\text{C}_1\text{-C}_4$, CO, CO_2) are analyzed using a gas chromatography. Each product yield is calculated and indicated on daf coal basis.

5) Characterization of Catalyst.

Catalyst samples for analysis are prepared by washing with THF to remove the solvent, followed by vacuum drying to remove the THF. The properties of catalyst are characterized using a quantitative metal analysis, powder X-ray diffraction (XRD), thermal gravimetric analysis (TGA), and transmission electron microphotograph (TEM) with an energy dispersed X-ray (EDX) detector.

6) Hydrotreatment of Liquefied Oil

Hydrotreatment of liquefied oil from Banko coal is carried out using a fixed bed reactor (14.7MPa, LHSV 1.0h⁻¹, H_2/Oil 1000Nm³/kl-oil) with Ni-Mo-P/Al₂O₃ catalyst (Criterion- 424). The first step hydrotreatment is performed at 350°C with crude liquefied oil as a feedstock. Next, the second step hydrotreatment is performed at 380°C using the light fraction ($\text{C}_5\text{-300 }^\circ\text{C}$) recovered from the first hydrotreated product oils. The product oils are separated by distillation into Fr-A ($\text{C}_5\text{-100 }^\circ\text{C}$, Light Gasoline), Fr-B (100-150 °C, Heavy Gasoline), Fr-C (150-240 °C, Kerosene) and Fr-D (240-350 °C, Gas Oil), respectively. Analyses of product oils are carried out using a combustion quantitative method for sulfur, coulometric titration method for nitrogen. The fluorescent indicator adsorption (FIA) analysis is performed in accordance with ASTM-D1319-61T to determine the contents of aromatics, olefins and saturates. The fuel indexes, such as density, octane number, smoke point, cetane index, kinematic viscosity, Saybolt color and the pour point, are measured to examine the fuel characteristics of the product oils by the improved BCL process.

3. Results and Discussion

(1) Liquefaction Reactivity of Banko Coal

In the preliminary study, five low-rank coals (Central Banko, Adaro, Pasir, Berau and Cerenti)

in Indonesia are tested using a batch type autoclave to examine the liquefaction reactivity by a comparison with Yallourn coal in Australia. Figure 1 shows the correlation between H/C atomic ratio of coal and the oil yield in coal liquefaction. The oil yield increases with an increase in H/C atomic ratio of coal, and low-rank coals in Indonesia shows the higher oil yield than Yallourn coal in Australia. It seems that oil yield increases also with a decrease in oxygen content according to the decrease in H₂O and CO + CO₂ yields. Central Banko coal exhibits the highest oil yield among the tested five coals in Indonesia. Results from the petrographic analysis indicates that the vitrinite contents ranged from 86 to 96vol% and inertinite contents are less than 5vol% for low-rank coals in Indonesia. The moisture content of Central Banko coal is about 35wt%, lower than that of Yallourn coal. These results suggest that Central Banko coal in Tanjung-Enim, South Sumatra, is favorable feedstock for coal liquefaction in Indonesia.

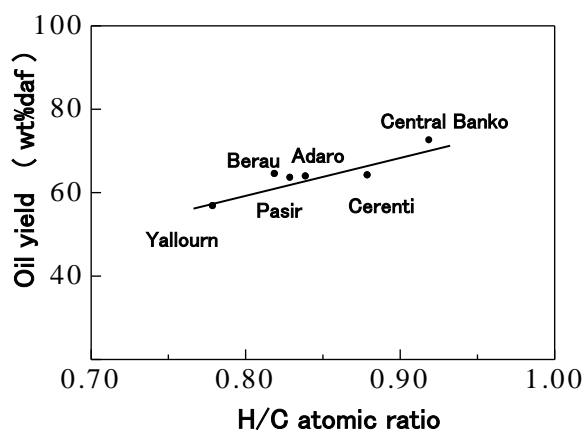


Figure 1 Liquefaction reactivity of low-rank coal in Indonesia

Figure 2 shows the influence of coal properties on oil yield in the liquefaction of Banko coals at 450°C, 120min with 1wt%daf of YY catalyst and the addition of 50wt%daf of CLB-THFS. The oil yield increases slightly with an increase in H/C atomic ratio or with a decrease in ash content of Banko coal. It is found that the oil yield strongly depends on the sulfur or iron contents in coal. The correlation between H/C atomic ratio and oil yield can be weakened by the influences of ash and iron contents in coal. If the catalyst loading is less than 1wt%daf as Fe, oil yield increases linearly with iron content in coal. These results suggest that FeS₂ (pyrite) in coal has a catalytic activity in coal liquefaction.

Figure 3 shows the effect of catalyst loading on oil yield with Central Banko (CB-9510) and South Banko (SB-98A) coal. The iron content (1.77wt%daf) of SB-98A is higher than that (0.27wt%daf) of CB-9510. The oil yield increases significantly with an increase in the loading of YY catalyst. It seems that the oil yield from SB-98A coal is apparently higher than that from CB-9510, but the oil yield from CB-9510 becomes exactly equal to the oil yield from SB-98A when the total iron content (Fe in catalyst and Fe in coal) is the same in the reactor. These results indicate that iron in coal can act as an iron catalyst in coal liquefaction.

In the improved BCL process, a part of CLB is recycled into the reactor to obtain a high oil yield. Since the CLB contains the used iron catalyst, which is transformed into pyrrhotite (Fe_{1-x}S), the iron content in the reactor increases to 3-5wt%daf as Fe by the CLB recycling operation even when the initial catalyst loading is 1wt%daf.

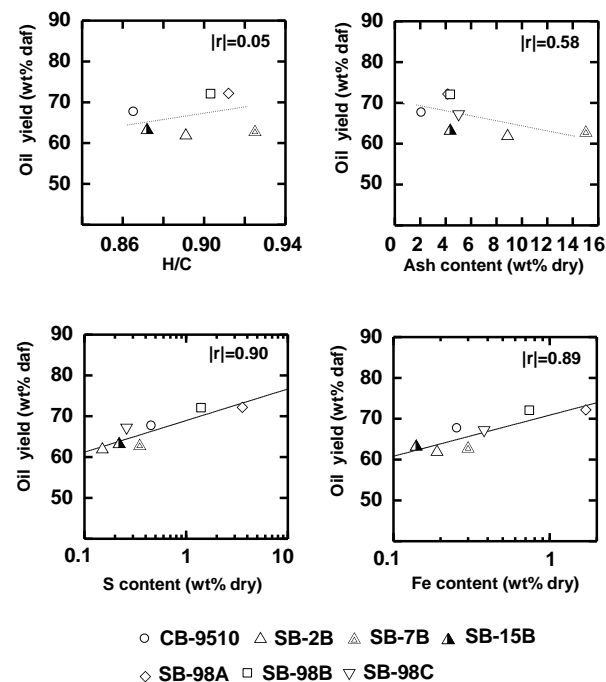


Figure 2. Influence of Coal Properties on Oil Yield in the Liquefaction of Banko Coal

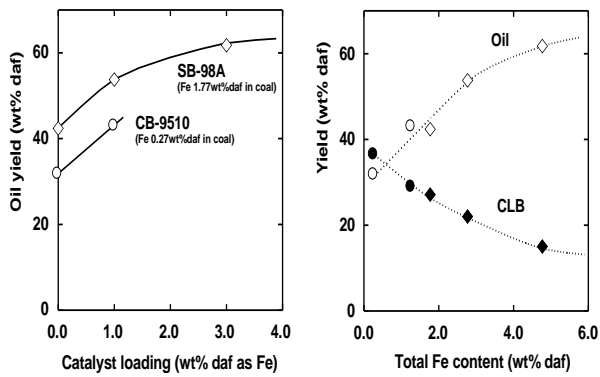


Figure 3. Effect of Catalyst Loading on Oil Yield (450°C, 120min, CLB=0)

Figure 4 shows the comparison of product yields in the liquefaction of Central and South Banko coal with 1wt%daf as Fe of Soroako limonite catalyst and the addition of CLB (60wt%daf). The oil yields from two coals are nearly equal, 65.9wt%daf for Central Banko coal and 65.2wt%daf for South Banko Coal. It appears that product yields of C₁-C₄, CO+CO₂ and H₂O are also the same between two coals. These results suggest that the CLB recycle is a promising way to obtain a high oil yield by the liquefaction of Banko coal. The influences of the coal properties can be effectively minimized by the CLB recycling operation in the improved BCL process.

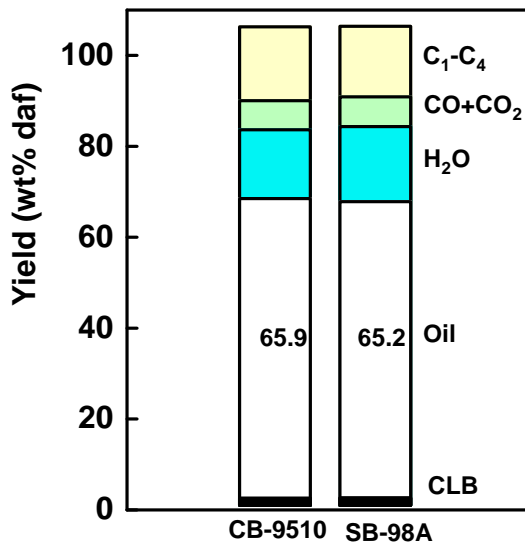


Figure 4. Comparison of product yields between Central and South Banko coal (Solv./CLB/Coal=40/60/100, 450°C, 120min, 1wt%daf as Fe of SO catalyst)

(2) Liquefaction Activity of Soroako Limonite Catalyst

From the economic point of view, the natural minerals in Indonesia should be utilized as a catalyst material for the liquefaction of Banko coal. A large amount of limonite is deposited on the nickel mine in Soroako, Sulawesi Island as shown in Figure 5. The limonite is conveniently classified into HGL (High Grade Limonite), MGL (Medium Grade Limonite) and OB (Overburden). The majority of this Soroako limonite is mined with nickel ore and buried back to the deposit because of the insufficient nickel content to the economical nickel recovery. Table 1 shows metal composition of the Soroako limonite, compared to Yandi Yellow (YY) limonite in Australia. The Soroako limonite contains particularly about 1-2wt% of Ni and Cr. The Fe content of Soroako limonite is 41-45wt%, which is slightly lower than that of YY due to the higher content of mineral matters. Results from XRD and TGA analyses indicate that both Soroako and YY limonite consist of mainly α -FeOOH. The compositions of Soroako (MGL) and YY limonite are estimated from results of XRD, TGA and TEM-EDX analyses as follows (wt% dry):

- 1). MGL: α -FeOOH (70.9), Al(OH)₃ (20.5), SiO₂ (1.3), NiFe₂O₄ (3.5), FeCr₂O₄ (3.0)
- 2). YY: α -FeOOH (91.6), Al₂Si₂O₅(OH)₄ (5.5), SiO₂ (1.9)

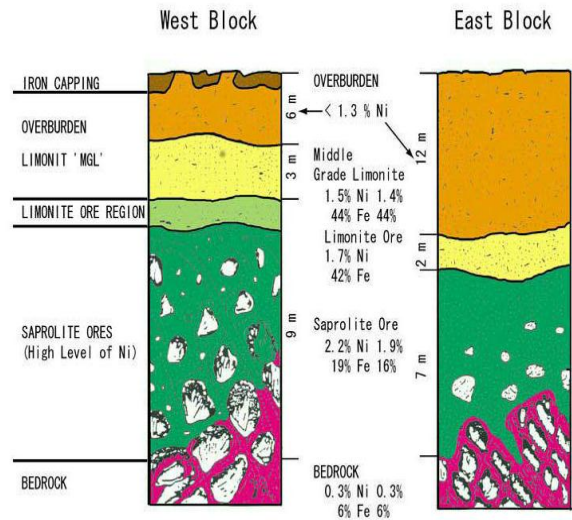


Figure 5. Limonite deposit on the nickelmine in Soroako, Sulawesi Island

Preliminary pulverization test for the Soroako limonite is carried out using a small type of planetary ball mill to prepare the liquefaction

catalyst. Table 2 shows physical properties of finely pulverized limonite catalysts. Soroako limonite can be easily pulverized to sub-micron particle size with a small abrasion of medium ball as well as YY limonite. Characterization results indicate that the finely pulverized Soroako limonite catalyst has higher surface area and smaller crystallite size of α -FeOOH, compared to those of YY catalysts.

Figure 6 shows the comparison of liquefaction activities of the limonite catalysts and a synthetic γ -FeOOH catalyst in the liquefaction of Banko coal at 450°C for 120min with 1wt%daf as Fe of catalyst loading. It appears that the Soroako limonite (MGL and HGL) catalysts exhibit higher oil yield than the γ -FeOOH or the YY catalyst, although the oil yield with OB catalyst is slightly lower than the γ -FeOOH catalyst. The Soroako limonite (MGL) is hopeful as a catalyst raw material for coal liquefaction because of the abundant and easily available resources.

Table 1. Metal composition of the Soroako limonite, compared to Yandi Yellow (YY) limonite

Limonite ore	Metal composition (wt% dry)									
	Total-Fe	Si	Al	Ni	Co	Cr	Ca	Mg	Na	S
Soroako										
HGL	45.4	0.6	7.08	0.88	0.02	1.38	<0.01	0.33	<0.01	0.16
MGL	45.5	0.78	5.95	1.33	0.09	1.16	0.05	0.15	<0.01	0.25
Over Burden (ref.)	41.4	1.68	6.06	1.65	0.08	1.45	<0.01	0.33	<0.01	0.13
Yandi Yellow	55.6	2.32	1.41	<0.01	<0.01	<0.01	0.24	0.12	<0.01	0.01

Table 2. Physical properties of finely pulverized limonite catalysts

Limonite	Pulverization ^{*1}		Characterization of pulverized catalyst	
	Particle size Dp50, (mm)	Abrasion ^{*2} (wt%)	Surface area BET, (m ² /g)	Crystallite size of γ -FeOOH from peak(101) in XRD (nm)
HGL	0.62	0.06	58.1	12.5
MGL	0.72	0.06	54.7	11.3
OB	0.92	0.03	61.1	8.3
ref.				
Yandi Yellow	0.66	0.03	39.2	21.5

*1 By using a planetary mill at 250rpm for 2h with 15g limonite and 45g decaline.

*2 Abrasion was determined by the weight loss of medium ball after the pulverization.

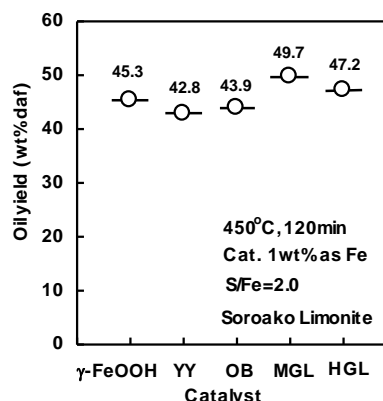


Figure 6. Comparison of the liquefaction activity among limonite catalysts

It has been generally recognized that pyrrhotite (Fe_{1-x}S) is an active phase of iron based catalyst in coal liquefaction. The smaller crystallite size of pyrrhotite can result in the higher liquefaction activity. Figure 7 shows the transformation behavior of iron based catalysts into pyrrhotite with the addition of elemental sulfur at 2.0 atomic ratio of S/Fe using an autoclave equipped with an infrared furnace at the heating rate of 100°C/min and 10MPa of cold H_2 under near liquefaction conditions in the absence of coal. It appears that the increase in crystallite size of pyrrhotite depends on the raw material of iron based catalyst. The Soroako limonite (MGL) can be transformed into a small crystallite size of pyrrhotite (10-20nm) at low temperatures around 300-350°C, following by the increase in crystallite size to about 35nm at 450°C and 30min of holding time. It should be noted that the crystallite size of pyrrhotite formed from Soroako limonite is remarkably smaller than that from YY or α -FeOOH catalyst at high temperatures around 450°C. This result suggests that the mineral matters such as Si and/or Al compounds in the limonite can contribute to the depression of the agglomeration of pyrrhotite particles. A strong interaction or bond formation such as Fe-O-Al between $\text{Al}(\text{OH})_3$ and α -FeOOH in the Soroako limonite may significantly suppress the agglomeration of fine particles of pyrrhotite, leading to the higher catalytic activity in coal liquefaction. On the other hand, nickel can be expected to promote the catalytic activity of iron catalyst in coal liquefaction. The regressive analysis between oil yield and metal contents in the Soroako limonite catalysts shows the positive effect for Fe, Ni, Al, and the negative effect for Si and Cr on the liquefaction activity. The result from TEM-EDX analysis indicated that nickel atoms could exist around the local structure of pyrrhotite in the CLB-THFI derived

from the Soroako limonite catalyst. Unfortunately, it is quite difficult to clarify the effect of nickel on the liquefaction activity due to the insufficient low nickel content for the successful characterization techniques such as XPS.

Figure 8 shows the liquefaction activity and the crystallite size of pyrrhotite for the fresh and the recycle catalyst (CLB-THFI) recovered from the autoclave after the liquefaction of Banko coal.

For the fresh catalyst, Soroako limonite shows a higher oil yield and a smaller crystallite size of pyrrhotite, compared to YY catalyst. However, for the recycle catalyst, the oil yield is almost the same between Soroako and YY catalyst. It appears that the crystallite size of pyrrhotite from the Soroako limonite can be increased more easily than that from YY catalyst during the recycle use.

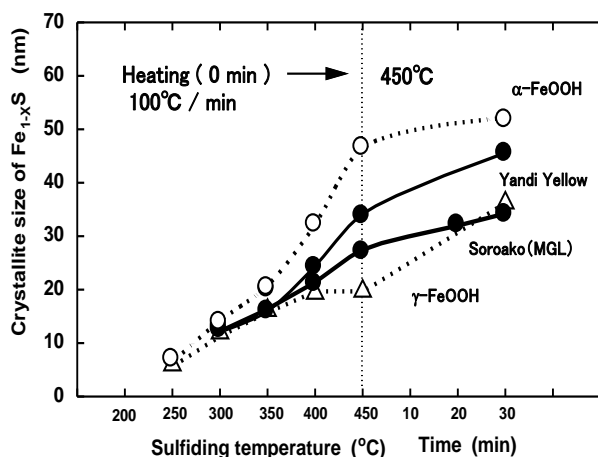


Figure 7. Transformation of iron based catalysts into pyrrhotite ($Fe_{1-x}S$) by the addition of elemental sulfur at 2.0 atomic ratio of S/Fe in the absence of coal

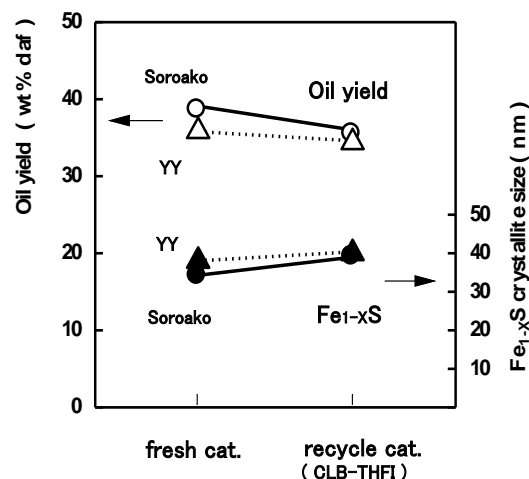


Figure 8. Oil yield and crystallite size of pyrrhotite for fresh and recycle catalysts

The catalyst deactivation can be caused by the crystal growth of pyrrhotite. Consequently, Soroako limonite catalyst showed the same oil yield with YY catalyst under the addition of CLB as shown in Figure 9. The oil yield reaches to 66.7wt%daf in the liquefaction of Banko coal at 450°C for 60min. There is no remarkable difference in product yields between Soroako and YY catalyst.

These results suggest that the Soroako limonite can be used as a catalyst raw material in a commercial plant for the liquefaction of Banko coal in Indonesia.

(3) Quality of liquefied oil

Large amounts of nitrogen, sulfur and oxygen compounds are contained in the liquefied crude oils from coal. In the improved BCL process, these hetero-atoms are removed by the 2-step in-line hydrotreatment with a conventional Ni-Mo/ Al_2O_3 catalyst. Table 3 shows the properties of liquefied oils derived from Banko coal.

It is found that the nitrogen and sulfur contents in both fractions of Fr-1 and Fr-2 are significantly decreased by the 2-step hydrotreatment of liquefied oils. The hydrotreated Fr-3 fraction is mostly used as a recycle solvent because of its high H/C atomic ratio. The storage stabilities of liquefied oils are remarkably improved by the removal of hetero-atoms as shown in Figure 10.

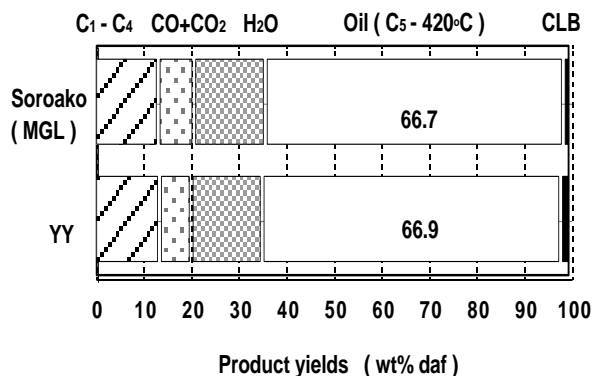


Figure 9. Comparison of product yields between Soroako and YY catalyst in the liquefaction of Banko coal at 450°C, 60min under the addition of CLB

Table 4 shows the fuel properties of second hydrotreated product oils. The product oil is conveniently divided into 4 fractions corresponding to light gasoline (LGS), heavy gasoline (HGS), kerosene, and gas oil. LGS fraction has low contents of aromatics (4.2%) and benzene (0.2%). The octane number (RON) is 75.8, slightly higher compared to 65-70 of petroleum oil. Naphtha fraction will be used as a feedstock for catalytic reforming to produce a blend stock of high octane number gasoline. The octane number can be expected to go up to over 95 (RON) through the application of a conventional reforming technology.

The aromatics content of kerosene fraction is 25.2%, and the results from HPLC analysis shows that most of aromatics consist of 1-rings such as alkyl benzenes. The smoke point is 14mm, slightly lower than 16mm of the petroleum standard in Indonesia. The gas oil fraction has a lot of aromatics of 41.8% consist of 37% of 1-rings, 3.4% of 2-rings and 1.4% of 3-rings. The calculated cetane index is 29.6 and the actual cetane number by the engine test is 35.0. It seems that the disagreement between cetane index and cetane number can be caused by the different compositions from petroleum gas oils. The cetane number of gas oil fraction is lower than 45 of the petroleum standard due to the aromatics rich composition of coal-derived gas oil. However, the sulfur content of gas oil fraction is 14ppm, extremely low level compared to petroleum gas oils.

Table 3. Properties of liquefied oils derived from Banko coal

Liquefied oil from Banko coal	Density (g/ml) at 15°C	Elemental analysis (wt%)				H/C atomic ratio	
		C	H	N (ppm)	S (ppm)		
Fr-1 Crude liquefied oil (C ₅ -180°C)	0.796	82.9	12.8	4224	921	3.8	1.84
1st Hydrotreated	0.8	86.2	13.6	308	5	0.2	1.88
2nd Hydrotreated	0.803	86.2	13.9	1	-	0	1.92
Fr-2 Crude liquefied oil (180-300°C)	0.968	85.7	9.7	7182	564	3.8	1.35
1st Hydrotreated	0.923	88	11.1	1061	5	0.8	1.5
2nd Hydrotreated	0.885	87	13	1	-	0	1.78
Fr-3 Crude liquefied oil (300-420°C)	0.994	87.7	9.3	8259	572	2.1	1.26
1st Hydrotreated	0.963	88.6	10.5	1445	5	0.8	1.41

* 1st hydrotreatment: 350°C, LHSV 1.0h⁻¹, 2nd hydrotreatment: 380°C, LHSV 1.0h⁻¹

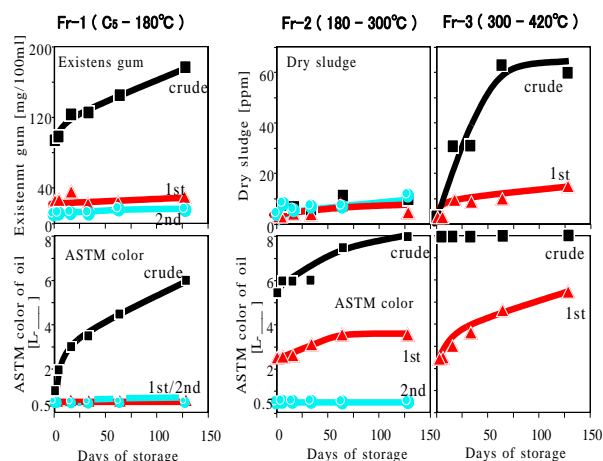


Figure 10. Improvement of storage stability by the hydrotreatment

Table 4. Fuel properties of hydrotreated product oil derived from Banko coal

Hydrotreated Product Oil		LGS	HGS	Kerosene	Gas Oil
Boiling Point (°C)		-100	100-150	150-240	240-350
Density (g/cm ³) @15°C		0.751	0.782	0.866	0.915
FIA (vol%)					
Saturates		95.8	87.4	74.8	57.9
Olefines		0	0	0	0.3
Aromatics		4.2	12.6	25.2	41.8
GC	Benzene	0.2			
	Toluene	1.3			
	Xylenes	2.6			
HPLC	Total Aromatics			27.6	41.8
	1-ring			27.1	37
	2-ring			0.5	3.4
	3-ring			0	1.4
Octane (RON)	Number	75.8			
Smoke (mm)	Point			14	
Cetane Index					29.6
Cetane (Engine Test)	Number				35
Kinematic Viscosity (mm ² /s)					4.5
Saybolt Color				+30	
Pour Point (°C)					-17.5
Sulfur (ppm)		8	16	3	14
Nitrogen (ppm)		1	1	1	7

In the background of recent tightening to the environmental restriction, low sulfur fuels are required for diesel cars. Further decrease in aromatics can be expected to improve the market adaptability of coal liquefied oils. Fig.11 shows the improvement of the smoke point for kerosene fraction and the cetane number of gas oil fraction with the decrease in aromatics of coal liquefied oils. These data (●) are obtained from the up-grading tests using 40bbl/d PDU (Funakawa, Japan) with Ni-W/Al₂O₃ catalyst for the liquefied oils from Tanitoharum coal by the researchers of PEC (Petroleum Energy Center) [6]. It is found that our data (●) from Banko coal in Table 4 are successfully fitted on the same line with data from Tanitoharum coal. These results indicate that the decrease in aromatics can lead to the improvement of the smoke point up to 17mm and the cetane number over 45 for the liquefied oils from Banko coal.

Recently, the decrease in aromatic compounds is considered as an important problem as well as a deep desulfurization of petroleum gas oil because of a severe environmental restriction for diesel fuels. In the

improved BCL process, sulfur and nitrogen compounds in the crude liquefied oils can be completely removed by the 2-stage in-line hydrotreatment. Coal liquefied oils have an advantage that sulfur level is extremely lower than petroleum oils. Further decrease in aromatics can result in the further improvement of the market adaptability for coal liquefied oils in Indonesia.

4. CONCLUSION

Banko coal in Tanjung-Enim area, South Sumatra, is a favorable feedstock for coal liquefaction because of the highest oil yield among five low-rank coals in Indonesia. The oil yield increases slightly with an increase in H/C atomic ratio, with a decrease in the ash content, and with an increase in the iron content of Banko coal. Iron in coal can act as catalyst in coal liquefaction. The CLB addition is a promising way to obtain a high oil yield over 65wt%daf in the liquefaction of Banko coal. The influences of coal properties on oil yield can be effectively minimized by the CLB recycling operation in the improved BCL process.

Soroako limonite can be utilized as an inexpensive catalyst material for the liquefaction of Banko coal because of the pulverizing easiness and a high catalytic activity in coal liquefaction.

Soroako limonite (MGL) exhibits a higher liquefaction activity than a synthetic γ -FeOOH and Yandi Yellow (YY) limonite catalyst because of the transformation into a small crystallite size of pyrrhotite (Fe_{1-x}S) through the addition of elemental sulfur. However, the crystallite size of pyrrhotite from the Soroako catalyst is increased more easily than that from YY catalyst by the recycle use, resulted in the same oil yield under the CLB addition. The oil yield reaches to 66.7wt%daf in the liquefaction of Banko coal at 450°C for 60min with 1wt%daf as Fe of the Soroako catalyst under 100wt%daf of the CLB addition. There is no remarkable difference in product yields between the Soroako and YY catalyst.

Most nitrogen and sulfur compounds in crude liquefied oils are successfully removed by the two-stage hydrotreatment, provides improvement in the storage stabilities and product qualities. The octane number (RON) of light gasoline fraction is 75.8, slightly higher than 65-70 of petroleum oils. Naphtha fraction can be used as a feedstock for the catalytic reforming to produce a blend stock of high octane number gasoline. The octane number can be expected to go up to over 95 (RON) through the application of

a conventional reforming technology in the improved BCL process.

The smoke point of kerosene fraction is 14mm, slightly lower than 16mm of the petroleum standard in Indonesia due to the aromatics rich composition. The cetane index is 29.6 and the actual cetane number by the engine test is 35.0 for gas oil fraction. The sulfur content of gas oil fraction is 14ppm, extremely low levels compared to petroleum gas oils.

Further decrease in aromatics can be expected to improve the smoke point and cetane number, leading to promote the market adaptability of liquefied oils from Banko coal. The smoke point and the cetane number can be increased by the hydrogenation of the aromatics in the improved BCL process.

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