FUNDAMENTAL STUDY ON LIQUEFACTION OF KALIMANTAN BROWN COALS

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Abstract

Efek karakateristik batubara terhadap yield produk pencairan batubara diinvetigasi menggunakan sampel batubara dari Berau (Lati dan Kelai), Mulia, Wara dan Kideco. Batubara Banko dari Tanjung Enim, Sumatera Selatan, digunakan sebagai batubara rujukan. Pengujian pencairan dilaksanakanmenggunakan gas-flow type autoclave (5L) dengan umpan batubara 2002 (daf), pelarut batubara turunan (b.p.300-420°C) 400g, katalis Fe 1.0-3.0 wt% (daf), elemen sulfur (rasio atom S/Fe 2.,0) pada tekanan 14.7MPa, suhu 450°C selama 60menit. Sampel limonit dari Soroako Indonesia dan Yandi Yellow dari Australia, yang terutama terdiri dari α-FeOOH, digunakan sebagai bahan katalis pada pengujian pencairan batubara. Studi ini merekomendasikan bertambahnya minyak yield akibat peningkatan kandungan oksigen dalam batubara, rasio atom H/C dan kandungan besi dalam batubara, mengingat besi dalam batubara dapat berfungsi sebagai katalis dalam pencairan batubara. Limonit Soroako medium grade (MGL) menunjukkan aktifitas lebih tinggi dibandingkan Yandi Yellow (YY) karena sifat transformasinya membentuk kristal-kristal pyrrhotite (Fe$_{1-X}$S), fase aktif dalam pencairan batubara. Sejumlah senyawa nitrogen dan sulfur dalam minyak batubara telah berhasil dihilangkan melalui proses hydrotreatment selanjutnya memperbaiki mutu produk
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Keywords: *pencairan, batubara berau, limonit,* pyrrhotite, *soroako,* hydrotreatment

1. INTRODUCTION

BPPT in cooperation with NEDO, Kobe Steel Ltd and Sojitz Corporation, Japan, has conducted feasibility study on coal liquefaction in Indonesia from 1994-2004. Brown coals from South Sumatra and Kalimantan were examined using an autoclave to investigate the influence of coal properties on the product yields (Kaneko, T., 2002). Since catalyst is also a key material on the development of coal liquefaction technology, further study was conducted through the development of highly dispersed limonite catalysts for the Brown Coal Liquefaction (BCL) process. Successful operations using a 0.1t/d Bench Scale Unit (BSU) at TCLC was accomplished (Kaneko, T., and et al., 2001; Kaneko, T., and et al., 2001; Kaneko, T., and et al., 1998; Kaneko, T., and et al., 2000)

Coal contains lots of heavy metal which can poison catalyst used in coal liquefaction therefore after reaction catalyst cannot be reused and should be disposed. Economically, the utilization of natural minerals for coal liquefaction catalyst is very important.

Study conducted in Takasago Laboratory suggests that limonite has an excellent catalytic activity in coal liquefaction (Kaneko, T., and et al., 2002). Because limonite is natural minerals, potentially it can be utilized as an inexpensive catalyst in coal liquefaction commercial plant. It is very fortunate that Indonesia possesses a large amount of limonite deposit in Soroako, Sulawesi. This Soroako limonite is a byproduct of nickel ore mining activity which is buried back to the mine site because of insufficient nickel content in the limonite.

Liquefied coal produced from coal liquefaction process must be treated further to produce stable-high quality synthetic fuel oil, through hydro-treatment process. The hydro-treatment of liquefied oil has been carried out too using a fixed bed reactor with a conventional Ni-Mo/Al2O3 catalyst to improve the storage stability and fuel characteristics of product oils. Almost nitrogen and sulfur compounds in crude liquefied

oil can be successfully removed by the two-step hydro-treatment, resulted significant improvement in the storage stabilities and fuel properties.

Results from an operation of a 40bbl/d PDU (Process Development Unit) in Funakawa, Japan PDU demonstrated that the coal-derived gas oil met all specifications of Japanese Industrial Standards (JIS) by blending with petroleum-derived gas oil at 3:7 of the mixing ratio (Omori, K., and et al., 2001).

In this study the effect of coal properties and liquefaction reactivity of Berau, Mulia, Wara, and Kideko coals on product yields of coal liquefaction are examined as part of the feasibility study activities. The hydro-treatment of liquefied oil from Berau-Lati coal is also carried out using a fixed bed reactor with a conventional Ni-Mo/Al2O3 catalyst.

2. EXPERIMENTAL

2.1. Coal Samples
Coal samples, Berau Lati and Berau Kerai, Mulia, Wara and Kideko, used in the study were received from PT. Berau, PT. Arutmin, PT. Adaro and PT. Kideko in Kalimantan. Banko coal in Tanjung-Enim, South Sumatra, was used as a reference coal. All coal samples were ground to less than 60 meshes and dried in vacuum at 105 prior to the use in the elemental analysis. Proximate and elemental analyses were carried out in accordance with JIS-M8812. Metal contents in coal were measured with an atomic absorption spectroscopy (AAS) method. Petrographic analysis was performed using a microscope to determine the distribution of maserals (Exinite, Vitrinite and Inertinite) and the reflectance (Ro) was measured for Vitrinite.

2.2. Catalyst Preparation
Limonite samples collected from Soroako nickel mine of International Nickel Indonesia (PT. INCO) were used as catalyst raw materials. An Australian limonite Yandi Yellow, which consists of mainly α-FeOOH, was also used in this study. Limonite was dried in vacuum at 105, followed by the finely pulverizing 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.

2.3. Liquefaction Test
Liquefaction tests were carried out mainly using a gas-flow type autoclave (5L) with 200g (daf) of 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. The liquid products were separated into H_2O, distillated oil (C5- b.p. 420°C) and coal liquid bottom (CLB, b.p.> 420°C) by a distillation, and the gaseous products (C1-C4, CO, CO_2) were analyzed using a gas chromatography. The Coal Lliquid Bottom (CLB) recovered after the liquefaction test was used in the bottom recycle operation with 1wt%daf as Fe of Soroako limonite catalyst.

2.4. Hydrotreament of Liquefied Oil
Hydrotreatment of liquefied oil from Berau coal was carried out using a fixed bed reactor (14.7MPa, LHSV 1.0h-1, H2/Oil 1000Nm3/kl-oil) with Ni-Mo/Al2O3 catalyst (TK-565). The first step hydrotreatment was performed at 350°C with crude liquefied oil as a feedstock. Next, the second step hydrotreatment was performed at 380°C. The product oils were separated by distillation into Naphtha (C5-180°C), Kerosene (180-240°C) and Gas Oil (240-350°C) fraction, respectively.

Analyses of product oils were carried out using a combustion quantitative method for sulfur, coulometric titration method for nitrogen. The density at 15 was determined by the floating balance method (JIS-K2249). The type analysis of aromatic carbon was performed using HPLC with n-hexane as a developing solvent.

3. RESULTS AND DISCUSSION

3.1. Properties and Liquefaction Reactivity of Kalimantan Coals
Table 1 shows the properties of Kalimantan coal samples by the comparison with South and Central Banko coals. The moisture contents of Berau and Mulia coals are relatively lower than Banko coal.

All Kalimantan coals have low ash contents less than 3wt%dry. The content of volatile matter (VM) is higher for Mulia coal, following by S-Banko > Wara > Berau > Kideko. It appears that Mulia coal has higher H/C atomic ratio (0.942) than S-Banko coal (0.905). The iron content of coal is 0.28wt% for Berau, 0.22wt% for Mulia, 0.67wt% for Wara, and 0.14wt% for Kideko, respectively. Results of petrographic analysis indicate that Kalimantan coals have high vitrinite content more than 90vol%.

The inertinite content of Wara coal is 5.8vol%, which is slightly higher value among Kalimantan coal samples.

Influences of coal properties on oil yield are shown in from Figure 1 to Figure 3. The liquefaction tests of Kalimantan coals were carried out using a gas flow type 5L-AC with 3wt%daf as Fe of Yandi Yellow (YY) catalyst at 14.7MPa, 450°C, 60min. The oil yields from the

liquefaction of coal samples (Banko, Suban Jeriji, Arahan) in Tanjung-Enim, South Sumatra, were also indicated in the figure for a comparison purpose. It seems that the H/C atomic ratio of coal affects the liquefaction reactivity as shown in Figure 1. The oil yield increased with an increase in the H/C atomic ratio of coal, although Mulia coal showed relatively lower oil yield in spite of its higher H/C atomic ratio. The iron content of coal also influences the oil yield as shown in Figure 2. It suggests that the iron in coal can act as a liquefaction catalyst. The lower oil yield of Wara coal, which has high iron content, may be caused by other factors. Moreover, the oxygen content of coal influences the oil yield more strongly as shown in Figure 3. It appeared that oil yield increased with a decrease in the oxygen content of coal. Berau and Mulia coals showed the higher oil yield than Wara and Kideko coals. As a result of these preliminary examinations, Berau and Mulia coals were selected as a hopeful feedstock for the coastal case coal liquefaction in Kalimantan.

Table 2 shows properties of Berau coal samples with a different sulfur content received in 2002. The sulfur content of coal sample is 1.4 for low sulfur Lati, 2.3 for high sulfur Lati, 1.1 for low sulfur Kerai and 2.2 for high sulfur Kerai, respectively. Lati coal samples show lower oxygen contents and lower H/C atomic ratio compared to Kerai coal samples. It should be noted that Lati coal samples have high sodium content of 0.3-0.4wt%dry coal. It is important to examine the existing chemical form of sodium compound in coal, as sodium in coal may be converted to $NaCl$, Na_2CO_3 or other metal carbonate complexes and deposit in the liquefaction reactor. High sulfur coals are limited to use widely by the environmental regulations. These high sulfur coals can be utilized effectively as a feedstock for coal liquefaction, as sulfur in coal can be recovered as a by-product in the BCL process.

Table 1. Properties of Various Coal Sample

| Coal	Berau	Mulia Arutmin	Wara Adaro	Kideko	S-Banko PTBA	C-Banko PTBA
Proximate Analysis						
Moisture (WtT%)	22,30	33,60	42,60	38,40	40,90	36,10
Ash (Wt%dry)	1,70	2,60	2,80	1,80	5,50	2,20
V.M (Wt%dry)	49,20	55,10	50,50	47,80	51,20	45,40
Elemental Analysis						
C (Wt%daf)	71,10	71,30	70,40	70,10	70,30	70,20
H	5,10	5,60	5,10	5,00	5,30	5,10
N	1,60	0,90	1,30	1,10	1,30	1,10
S	0,70	0,30	0,20	0,20	1,50	0,50
Odiff	21,50	21,90	23,00	23,60	21,60	23,10
H/C atomic ratio	0,86	0,94	0,87	0,86	0,91	0,87
Metal Composition						
Fe (Wt%dry)	0,28	0,22	0,67	0,14	0,76	0,26
Ca	0,09	0,03	0,11	0,06	0,54	0,50
Mg	0,02	<0,01	0,04	0,01	0,07	0,05
Na	0,00	0,00	0,00	0,00	0,01	<0,01
Si	0,02	0,58	0,19	0,92	0,26	0,12
Al	0,31	0,25	0,30	0,47	0,26	0,24
Maseral						
Exinite (Vol%)	2,50	0,80	4,30	4,00		6,60
Vitrinite	94,20	95,80	89,90	94,00		90,70
Inertinite	3,30	3,40	5,80	2,00		2,70
Reflectance (Ro)						
Of Vitrinite	0,43	0,39	0,40	0,39		0,38
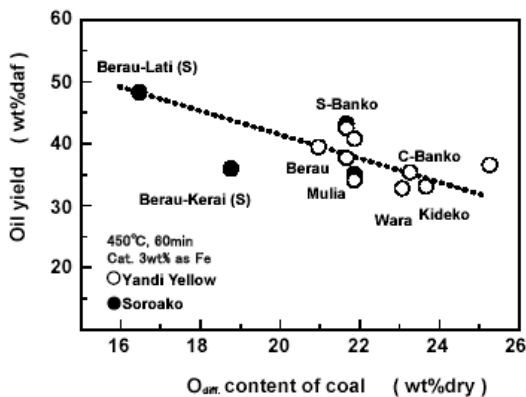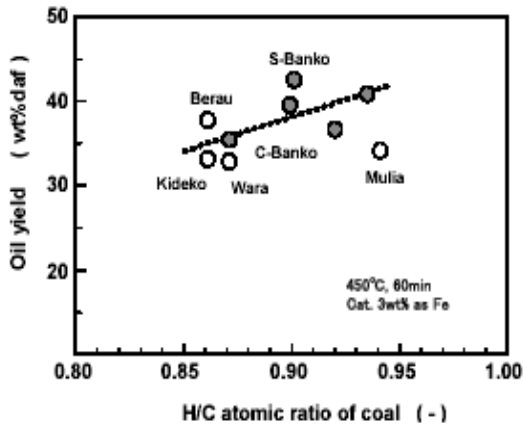
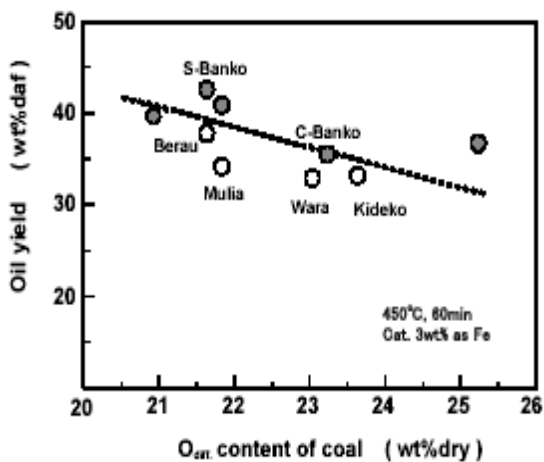Fig.5 Correlation between oil yield and oxygen content of coal

Fig.3 Influence of oxygen content on oil yield

Table 2. Property of Berau Coal with Different Sulfur Content

Berau Coal	Lati		Kerai	
	Low Sulfur	High Sulfur	Low Sulfur	High Sulfur
Proximate Analysis				
Moisture (Wt%)	22.6	22.9	25.5	24.8
Ash (Wt%dry)	5.5	4.0	2.1	4.3
V.M (Wt%dry)	46.1	49.0	50.4	51.4
Elemental Analysis				
C (Wt%daf)	73.8	74.3	72.5	72.2
H	5.3	5.5	5.5	5.6
N	1.7	1.4	1.3	1.3
S	1.4	2.3	1.1	2.2
Odiff	17.8	16.5	19.6	18.7
H/C atomic ratio	0.862	0.888	0.910	0.931
Metal Composition				
Fe (Wt%dry)	0.52	0.46	0.16	0.85
Ca	0.40	0.47	0.24	0.62
Mg	0.10	0.10	0.04	0.08
Na	0.31	0.39	0.01	0.01
Si	0.92	0.08	0.12	0.03
Al	0.17	0.02	0.17	0.07
Cl	0.0	0.0	0.0	0.0

Figure 4 shows the product yields in the liquefaction of high sulfur Berau coals with Soroako limonite catalyst of 3wt%daf as Fe by the comparison to Mulia and South Banko coals. It appeared that high sulfur Lati coal showed higher oil yield with lower CLB yield, compared to Lati, Mulia and S-Banko coals. The oxygen content of coal affects on oil yield as shown in Figure 5. Both H_2O and $CO+CO_2$ yields of Lati coal were considerably lower than those of Kerai and Mulia coals.

Figure 6 shows FT-IR (PAS: Photo Acoustic Spectroscopy) spectrum of Berau and Mulia coal. It appears that the peak intensity of carboxyl group (around 1700cm-1) of Lati coal is smaller than that of other coals, although FT-IR spectrums are similar with each other. Figure 7 shows the result of quantitative analysis for COOH group in coal with a back-titration method. The results from the determination of COOH group are consistent with the peak intensities of around 1700cm-1 in the FT-IR spectrums. The amount of COOH group is lower for Lati coal, followed by Kerai and Mulia coal. In general, the larger amount of COOH group in coal can result in the larger CO_2 yield in coal liquefaction.
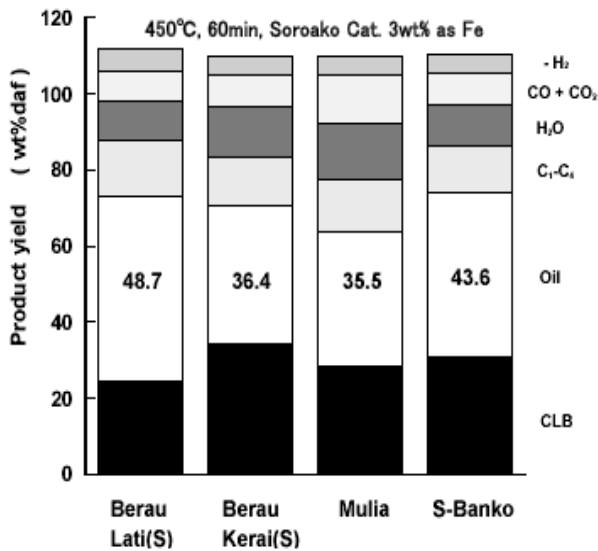
450°C, 60min, Soroako Cat. 3wt% as Fe

- H₂	$- H_2$

Fig.4 Product yields in the liquefaction of high sulfur Berau coals with Soroako catalyst
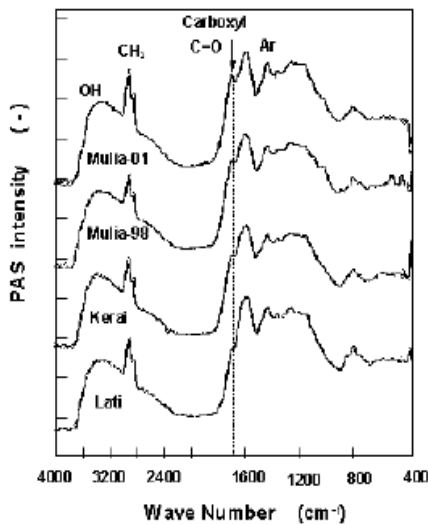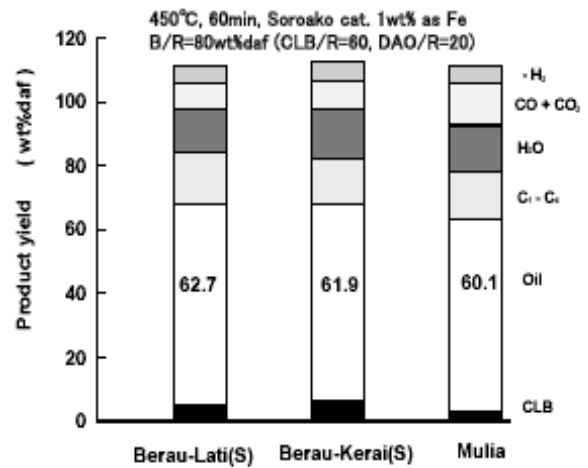

Fig.6 FT-IR (PAS) spectrum of coal

Figure 8. Product Yields In Coal Liquefaction Under The Bottom Recycle Conditions

Figure 8 shows the product yields in the liquefaction of high sulfur Lati, Kerai and Mulia coal under the bottom recycle conditions with 1wt%daf as Fe of Soroako limonite catalyst. The oil yield was 62.7wt%daf for Lati, 61.9wt% for Kerai, and 60.1wt% for Mulia, respectively.

Table 3 shows the properties of liquefied oils and CLB-THFS obtained from the coal liquefaction under the bottom recycle conditions. The results of elemental analysis indicated no drastic changes in the properties of liquefaction products in spite of the different coal properties. The sulfur contents of products from high sulfur Lati coal were 0.2wt% for oil and 0.3wt% for CLB-THFS, comparable to those from low sulfur Mulia coal. It appeared that most of sulfur in coal could be eliminated as H2S, not remaining in liquid products.
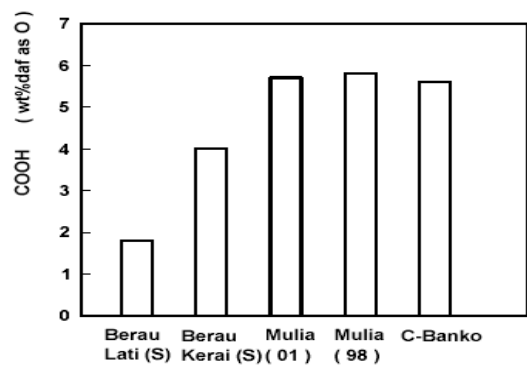

Fig.7 Determination of COOH group in coal

Table 3. Property of Liquefied Oil and CLB-THFSObtained From Coal Liquefaction Under Bottom Recycle Conditions

Coal	Berau- Lati (S)	Berau-Kerai (S)	Mulia
Liquefaction	14.7 MPa, 450 C B/R=80 wt%daf	60 min, Soroako cat (CLB/R=60 and DAO/R=20wt%daf)	1wt% daf as Fe
Oil yield	62.7 wt%	61.9wt%	60.1wt%
CLB yield	4.9 wt%	6.2 wt%	3.1wt%
Oil properties (C5-420 C)			
C	87,2	87,0	87,3
H	9,1	9,2	9,2
N	0,9	0,8	0,8
S	0,2	0,1	0,1
Odiff	2,6	2,8	2,6
H/C atomic ratio	1,252	1,269	1,265
CLB properties (CLB-THFS, 420 C)			
C (wt%)	87,8	87,3	87,6
H	6,3	6,8	6,6
N	2,2	1,9	1,8
S	0,3	0,2	0,1
Odiff	3,4	3,9	3,9
H/C atomic ratio	0,861	0,935	0,904
1H-NMR Index			
fa	0,73	0,68	0,7
O	0,35	0,39	0,38
Ln	1,96	2,09	2,03
Hau/Ca	0,66	0,7	0,69

In contrast to sulfur, nitrogen in coal remained in the liquefied oil and CLB-THFS. The nitrogen contents of the products in the liquefaction of Lati coal were 0.9wt% for liquefied oil and 2.2wt% for CLB-THFS. The oxygen compound also remained in the liquefaction products, such as phenols in oils and furans in CLB-THFS. These hetero compounds in the liquefied oil can be completely removed by the 2-step hydrotreatments with Ni-Mo/Al2O3 catalyst in the BCL process to improve the storage stability and to produce the clean transportation fuels.

These results suggested that Berau-Lati coal could be one of the hopeful feedstock for the coastal case coal liquefaction in Indonesia because of the relatively lower moisture and oxygen contents. High sulfur coal can be effectively utilized as a feedstock, as the sulfur in coal can be recovered as a by-product in the BCL process.

3.2. 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 Kalimantan coal. A large amount of limonite is deposited on the nickel mine in Soroako, Sulawesi Island as shown in Figure 9. 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 4 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 indicated that both Soroako and YY limonite consisted of mainly α-FeOOH. The compositions of Soroako (MGL) and YY limonite were estimated from results of XRD, TGA and TEM-EDX analyses as follows (wt%dry); MGL: α-FeOOH (70.9), Al(OH)3 (20.5), SiO2 (1.3), NiFe2O4 (3.5), FeCr2O4 (3.0) YY: α-FeOOH (91.6), Al2Si2O5(OH)4 (5.5), SiO2 (1.9).

Preliminary pulverization test for the Soroako limonite was carried out using a small type of planetary ball mill to prepare the liquefaction catalyst. Table 5 shows physical properties of finely pulverized limonite catalysts.

Table 4. Metal Composition of Soroako Limonite in Indonesia (wt%dry)

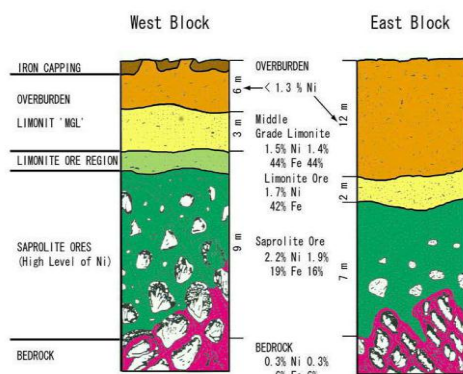| Limonite ore	Total-Fe	Si	Al	Ni	Co	Cr	Ca	Mg	Na	S
Soroako HGL	45,4	0,6	7,08	0,88	0,02	1,38	<0.01	0,33	<0.01	0,16
MGL	45,5	0,78	5,95	1,33	0,09	1,16	0,05	0,15	<0.01	0,25
Over Burden (ref)	41,4	1,68	6,06	1,65	0,08	1,45	<0.01	0,33	<0.01	0,13
Yendi Yellow	55,6	2,32	1,41	<0.01	<0.01	<0.01	0,24	0,12	<0.01	0,01

Figure 9. Limonite Deposit in Soroako Nickel Mine, Sulawesi

Table 5. Physical Properties of Pulverized Catalyst

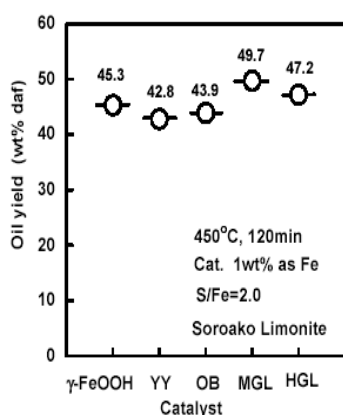| Limonite | Pulverization | | Characterization of pulverized catalyst | |
| | Particle size Abrasion | | Surface area | Crystallite size of α--FeOOH |
	Dp50 (μm)	(wt%)	BET (m²/g) from peak (101)in XRD	(nm)
HGL	0.62	0.06	58,1	12,5
MGL	0.72	0.06	54,7	11,3
OB	0.92	0.03	61,1	8,3
Ref. Yandi Yellow	0.66	0.03	39,2	21,5

Figure 10. Comparison of Liquefaction ActivityAmong Limonite Catalyst

Figure 11 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 100oC/min and 10MPa of cold H2 under near liquefaction conditions in the absence of coal. It appeared that the increase in crystallite size of pyrrhotite depended on the raw material of iron catalyst. The Soroako limonite (MGL) can be transformed into a small crystallite size of pyrrhotite (10-20nm) at low temperatures around 300-350oC, following by the increase in crystallite size to about 35nm at 450oC and 30min of holding time.

It should be noted that the crystallite size of pyrrhotite formed from Soroako limonite was remarkably smaller than that from YY or α-FeOOH catalyst at high temperatures around 450oC.

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 pyrrotite particles. A strong interaction or bond formation such as Fe-O-Al between Al(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 CLB-THFI derived from the Soroako limonite catalyst. Unfortunately, it was 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.
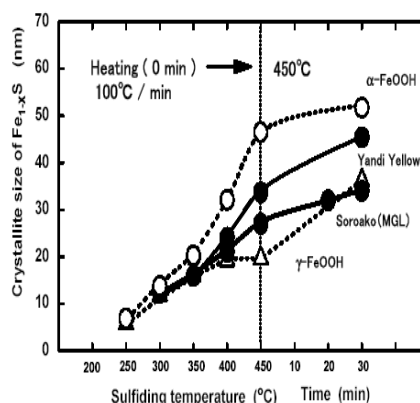

Figure 11. Transformation of Iron Based Catalyst into Pyrrhotite by the Addition of Element Sulfur at 2.0 Atomic Ratio of S/Fe in the Absenceof Coal

These results indicate that Soroako limonite can be effectively used as a catalyst material for a commercial plant of the coastal case coal liquefaction, because of the high liquefaction activity and domestic inexpensive resources near Kalimantan.

3.3. Quality of Liquefied Oil

Table 6 shows properties of crude liquefied oils from Berau and Mulia coals with the comparison to the Banko coal (F/S data). The crude liquefied oil was divided by a distillation into Fr-1 (C5-180), Fr-2 (180-300) and Fr-3 (300-420) fractions. The crude liquefied oils from high sulfur Berau coal indicate the higher sulfur content and the lower H/C atomic ratio, compared to those from the Bank coal. It appeared that the sulfur content was higher for the light fractions of Fr-1, while the nitrogen content was higher for the heavy fractions of Fr-3. The oxygen contents of both Fr-1 and Fr-2 were higher than that of Fr-3 fraction.

These hetero-atom compounds in the crude liquefied oils can be completely removed by the in-line 2-step hydrotreating in the BCL process as shown in Figure12. The efflux from the liquefaction reactor is immediately separated into gas and liquid phases in the separater under the high-temperature and high-pressure conditions. The gas phase of the separater is directly introduced to the in-line hydrotreating section, and whole crude liquefied oil (C5-420) is hydrotreated in the fixed bed reactor with a conventional Ni-Mo/Al2O3 catalyst. The 1st hydrotreated Fr-3 fraction is recycled to the slurry making section as a recycle solvent, while the lighter fractions are further hydrotreated in the 2nd fixed bed reactor to completely remove the nitrogen and sulfur compounds.

Table 6. Physical Properties of Pulverized Catalyst

| Coal	Berau-Lati (S)	Berau-Kerai (S)	Mulia	Banko (F/S)
Fr-1 (C5-180 C)				
C (wt%)	83	83.7	83.6	82.9
H	11.7	12.4	12.3	12.8
N (ppm)	4500	5300	5300	4200
S (ppm)	5100	1900	1900	900
Odiff	4.3	3.5	3.5	3.8
H/C atomic ratio	1.698	1.761	1.761	1.853
Fr-2 (180-300 C)				
C (wt%)	85.6	85.0	85.8	85.7
H	9.5	9.,5	9.5	9,7
N (ppm)	6300	6100	6200	7200
S (ppm)	1800	1600	700	600
Odiff	4.2	4.8	4.1	3.8
H/C atomic ratio	1.325	1.335	1.322	1.358
Fr-3(300-420 C)				
C (wt%)	87.2	88.3	88.9	87.7
H	9.1	8,5	8.5	9.3
N (ppm)	9000	9500	9200	8300
S (ppm)	2000	1200	1100	600
Odiff	2.6	2.2	1.6	2.1
H/C atomic ratio	1.145	1.159	1.145	1.273
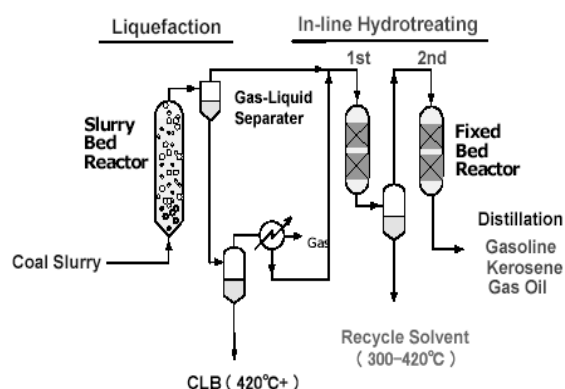
Figure 12. Conceptual Flow of Inline Hydro-Treatment Section in BCL Process

Table 7 shows the properties of the product oil in the 2nd hydrotreatment with comparison to that of Banko coal (F/S data with Criterion 424 catalyst: Ni-Mo-P/Al2O3). The liquefaction of Berau-Lati coal, 2-step hydrotreatment and the distillation of product oil were carried out at LSDE through the collaboration between Japan and Indonesia researchers. The product samples were carried back to Japan, followed by further distillation into naphtha, kerosene and gas oil fractions prior to the analysis in oil properties.

Table 7. Properties of Oil Product in Second Hydro-Treatment

2nd Hydro-treatment Products (b.p.°C)	Berau -Lati			Banko (F/S)
	Naphtha	Kerosene	Gas Oil 240-350	Gas Oil 240-350
	-180	180-240	350	350
Elemental Analysis				
C (Wt%)	85.06	86.52	86.93	88.38
H (Wt%)	14.34	13.25	12.81	11.53
N (ppm)	<10	<10	<10	7
S (ppm)	30	<10	<10	14
H/C atomic ratio	2.009	1,825	1.756	1.554
HPLC Analysis				
Total aromatics (vol%)		11,7	19.4	41.8
1- ring		11,7	17.9	37.0
2-ring		0	1.4	3.4
3-ring		0	0,1	1.4
Olefines		0	0	0.3
Saturates		88.3	80.6	57.9
Denisity (g/ml,15 C)	0.801	0.880	0907	0.915
Centane Index			38.6	29.6
Cetane Number				35.0

Figure 13. Improvement of Cetane Indexby Reduction of Aromatics

It appeared that the gas oil fraction of Berau-Lati coal had the higher H/C atomic ratio, lower amount of aromatics and the lower density, compared to Banko coal. The calculated cetane index was 38.6 for Berau-Lati coal, significantly higher than 29.6 of Banko coal. It suggested that the actual cetane number of the gas oil fraction of Berau-Lati coal could become more than 40. The cetane index of gas oil fraction can be further increased by the reduction of aromatics as shown in Figure 13. The sulfur contents of the gas oil and kerosene fractions were less than 10ppm, advantageously low levels compared to the petroleum gas oil fractions. 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.
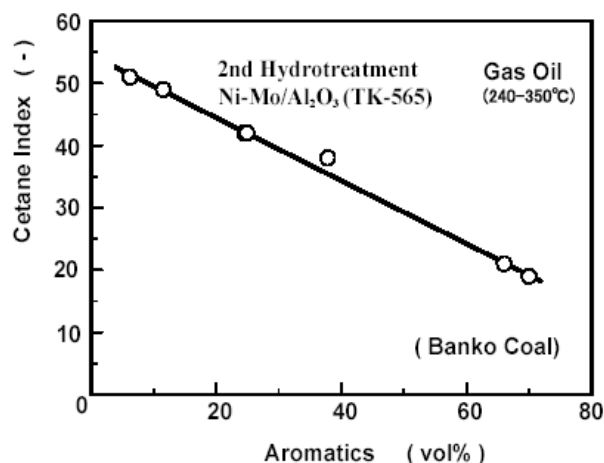
The current results from the Process Development Unit (PDU) for the upgrading of liquefied oils in Funakawa, Japan indicate that the octane number (RON) of gasoline fraction can be increased to 106 by the catalytic reforming of the coal–derived naphtha fraction at 490, 3.5MPa and LHSV of 1.0h-1. It has been demonstrated that the coal-derived gasoline, kerosene and gas oil met all specifications of Japanese Industrial Standards (JIS) by the blending at 3:7 of the mixing ratio with petroleum-derived oils (Omori, K., and et al., 2001).

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 Berau coal. The smoke point and the cetane number can be increased by the hydrogenation of the aromatics in the upgrading section of the BCL process.

These results suggest that low sulfur and low aromatics transportation fuels can obtain from Kalimantan coals by the application of the BCL process. The in-line 2-step hydrotreatment in the BCL process can be a promising way to produce the high quality transportation fuels from the low-rank coals in Indonesia.

4. SUMMARY

Berau coal in Kalimantan was selected as one of the hopeful feedstock for the coastal case coal liquefaction in Indonesia because of the relatively lower moisture and the higher liquefaction reactivity. High sulfur coal can be effectively utilized as a feedstock, as the sulfur in coal can be recovered as a by-product in the BCL process.

Coal properties can affect on oil yield in coal liquefaction. Oil yield increased with a decrease in the oxygen content and with increase in the

H/C atomic ratio and the iron content of coal. Iron in coal can act as a catalyst in coal liquefaction.

The influences of coal properties on oil yield can be effectively minimized by the bottom recycle operation in the BCL process. Under the bottom recycle conditions, Mulia coal showed similar oil yield to the Berau coal. The bottom addition is a promising way to obtain a high oil yield over 60wt%daf in coal liquefaction. The results of elemental analysis indicated no drastic changes in the properties of liquefaction products in spite of the different feed coal.

Soroako limonite in Sulawesi Island can be utilized as an inexpensive catalyst material for the coastal case coal liquefaction in Kalimantan, because of the pulverizing easiness and a high catalytic activity in coal liquefaction. Soroako limonite (MGL) exhibited a higher liquefaction activity than Yandi Yellow (YY) limonite because of the transformation into small crystallite size of pyrrhotite (Fe1-XS), which is recognized as an active phase in coal liquefaction.

Almost of nitrogen and sulfur compounds in crude liquefied oils were successfully removed by the two-step hydrotreatment, resulted in the improvement of product qualities. 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 cetane index for gas oil fraction of Berau coal was 38.6 and the actual cetane number can be expected up to more than 40. The sulfur contents of kerosene and gas oil fraction were less than 10ppm, extremely low levels compared to petroleum kerosene and 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 Berau coal.

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