EFFECT OF HYDROTHERMAL DEWATERING ON COKE ADDITIVE MAKING FROM LOW RANK COAL (LRC)

PENGARUH PENURUNAN KADAR AIR TERHADAP PEMBUATAN ADITIF KOKAS DARI BATUBARA PERINGKAT RENDAH

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ABSTRACT

This paper describes a study of the effects of hydrothermal dewatering (HTD) of Jambi, Pendopo and Wahau low rank coals, on additive characteristics. Hydrothermal upgrading and dewatering of the coals were carried out in a batch-type autoclave reactor at temperatures 350°C at a maximum pressure of 30 bar for 30 min. The dried sample resulted from hydrothermal process mixed with liquid fraction tar solvent at 250-350°C with ratio 4:6, was input in the 0,5 l autoclave to conduct hydrogenation process with variation initial hydrogen pressure of 10, 20, 30, 40, 50 bar, and reaction temperature of 400°C for 1 hour. The process of hydrothermal treatment before hydrogenation produced a higher calorific value having an average of >8000 cal/g (air dried basic, adb). Ash content and volatile matter for the coal were increased with the increasing initial hydrogen pressure. Corrected hydrogen content steadily increased after hydrothermal process and hydrogenation while the corrected oxygen decreased drastically after the hydrogenation process. Fuel ratio of Jambi, Pendopo and Wahau coals after hydrothermal process also increase reached 1.58, 1.04 and 1.77 respectively. Overall results indicate the importance of introducing a hydrothermal treatment step for the improvement of the coke additive characteristics.

Keywords: hydrothermal dewatering, low rank coal, corrected hydrogen and oxygen, coke additive

SARI

Makalah ini mempelajari pengaruh proses hydrothermal dewatering *(HTD) batubara peringkat rendah dari Jambi, Pendopo dan Wahau terhadap karakteristik aditif kokas. Proses pengeringan ini dilakukan dalam otoklaf tipe bacth pada suhu 350°C, tekanan 30 bar selama 30 menit. Batubara kering hasil proses HTD dicampur dengan larutan tar yang mempunyai titik didih 250-350°C dengan perbandingan 4:6, dimasukkan ke dalam otoklaf 0,5 l kemudian dihidrogenasi. Tekanan awal hidrogen bervariasi yaitu 10, 20, 30, 40 dan 50 bar, suhu reaksi 400°C selama 1 jam. Hasil penelitian menunjukkan bahwa proses pengeringan batubara sebelum hidrogenasi menghasilkan batubara dengan kalori > 8000 cal/g (adb). Kandungan abu dan zat terbang untuk ketiga batubara tersebut naik seiring dengan naiknya tekanan hidrogen. Hidrogen terkoreksi juga mengalami kenaikan setelah proses HTD dan hidrogenasi. Fuel ratio batubara Jambi, Pendopo dan Wahau mengalami kenaikan mencapai 1,58, 1,04, dan 1,77. Secara keseluruhan dapat dikatakan bahwa proses HTD merupakan tahapan awal sebelum hidrogenasi yang sangat berpengaruh positip terhadap karakteristik aditif.*

Kata kunci: hydrothermal dewatering*, batubara peringkat rendah, hidrogen dan oksigen terkoreksi, aditif kokas*

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INTRODUCTION

Coke-making technology from coking coals has long been known, but the use of Indonesian coal which is generally categorized as low rank coal to produce metallurgical coke remains to be studied. Considering the limited amount of coking coal in Indonesia and has not been inventoried, whereas the amount of non-coking coal is relatively large, it is necessary to carried out a study of the right coke-making technology to address the domestic needs of coke.

One of the technologies being studied is the additive making from low rank coal (LRC) for metallurgy coke. Additives is prepared by hydrogenate coal at a temperature of 400 - 500°C and about 10-50 bar hydrogen pressure with tar as solvent (Jiang et al., 2007 and Schieke, 2010). Tar which used as solvent is a waste product of coal gasification process

Conversion of low rank coal into additive is technologically feasible and commercially attractive. This additive research has been carried out since 2010 in R & D Center for Coal Technology. It conducted trials of additive making from high rank coal (calorific value > 5500 kcal/kg) with pyrrhotite catalysts formed from the reaction between sulfur and lateritic iron ore. The results showed that the additive can be prepared by hydrogenation of coal. Hydrogenation process lowers the O/C ratio and maintains a constant H/C ratio. The reduced amount of oxygen and the presence of adequate hydrogen causes coal become caking and can be used as an additive. The disadvantage of this process is the presence of catalyst which is mixed with additive so this process produces an additive having high ash content and sulfur.

A problem encountered by hydrogenation process of LRC is high hydrogen consumption due to high oxygen content of LRC. Therefore, in this present study, it carried out a pretreatment process of LRC before hydrogenation process. Hydrothermal dewatering (HTD) technology is one of an alternative pre-treatment process because HTD is able to produce dry coal with greater calorific value and lower oxygen content. The reaction temperature and the holding period of HTD were important operating parameter to obtain a usable solid fuel. Higher reaction temperature and longer holding period of HTD will produce more uniform and denser product. In addition, higher reaction temperature and longer holding period will produce products with lower organic chlorine content (Prawisudha, et al., 2012). This paper describes how the HTD technology influences the quality of additive. The purpose of this research is to find the most efficient method of additive making from Indonesian LRC for metallurgy coke making.

METHODOLOGY

Materials

The coals used for the research were Jambi, Pendopo, and Wahau LRC. The analysis of those LRC are shown in Table 1. The coal were pulverized to less than 200 mesh and dried at 150°C for 2 hours before use. The hydrogenation solvent was obtained from Palimanan. It is liquid fraction tar solvent of 250-350°C as a by-product of hydrogenated coal gasification. The catalyst, lateritic iron ore of -325 mesh from South Kalimantan was used in this research.

Coal Hydrogenation

The first study is coal hydrogenation to make good coking additive for coke. The equipments used for hydrogenation tests was a 0.5 l batch autoclave equipped with shaking type (54 times per minute) at variation initial hydrogen pressure of 10, 20, 30, 40, 50 bar, reaction temperature and reaction time of 400°C and 1 hour. The experiments were performed using coal added lateritic iron ore catalyst, liquid fraction tar solvent with weight ratio of coal and solvent of 4:6. After the reaction, liquid products and solid residue were dissolved with 250 ml of hexane, stirred until blended. Then, a screening process was performed, the residue left on filter paper was dried at room temperature. The dried residue were performed a proximate, ultimate and free swelling index (FSI) analyses.

Coal Hydrothermal

Coal and water with a ratio of 1:3 were put in a 5 l autoclave, stirred gently until evenly distributed. The autoclave was sealed until there was no leaks, then streamed N_2 gas up to 30 bar (30 bar PoN_2). The heater and coal sample were heated with the temperature condition in the reactor stabilized in 350°C, pressure of 30 bar, and 30

Table 1. The analysis results of the proximate, ultimate and calorific value of LRC

Parameter	Jambi	Pendopo	Wahau	Method
Moisture (% adb)	10.46	17.37	12.34	ASTM D.3302
Ash (% adb)	2.71	6.19	2.58	ASTM D.3174
Volatile matter (% adb)	45.08	41.54	48.36	ISO 562
Fixed carbon (% adb)	41.75	34.90	36.72	ASTM D.3172
Carbon (% adb)	63.73	53.79	62.32	ASTM D.5373
Hydrogen (% adb)	5.50	6.65	5.80	ASTM D.5373
Nitrogen (% adb)	0.85	0.80	0.75	ASTM D.5373
Total sulfur (% adb)	0.08	0.29	0.15	ASTM D. 3176
Oxygen (% adb)	27.13	32.28	26.98	ASTM D.5865
Calorific value (cal/g. adb)	5.715	4.932	5.819	ASTM D.720
Free Swelling Index (FSI)	0	0	0	

Note: adb = air dried basis

minutes reaction time. Samples that have been dried were analyzed partly for the hydrogenation process. Dried sample resulted from hydrothermal process mixed with liquid fraction tar solvent of 250-350°C, coal and solvent ratio 4:6, was put in the 0,5 l autoclave to conduct hydrogenation process with variation initial hydrogen pressure of 10, 20, 30, 40, 50 bar, the reaction temperature of 400oC for 1 hour. Once the process was complete the autoclave was turned off. Samples that have been dried were analyzed for their proximate, ultimate, calorific value and FSI.

RESULTS AND DISCUSSION

The Catalytic Coal Hydrogenation Products

The aim of catalytic hydrogenation was to determine the characteristics of coal after the adding of hydrogen. In the hydrogenation process, the coals which have long clusters of hydroxyl were divided into small groups (Maity et al., 2003). The results for the catalytic hydrogenation process are listed in Table 2, 3 and 4.

Ash content in Jambi coal after the catalytic hydrogenation was visibly increased in accordance with the increasing hydrogen pressure due to the effect of adding catalyst. Catalyst will partially become ashes if hydrogenated, as a result of the increasing ash content of hydrogenation results. Except for ash content at 30 bar hydrogen pressure which is decreased, due to volatile matter and fixed carbon of the coal at 30 bar seemingly changed more gaseous. Total carbon content as seen in Table 2 increased significantly from 63.73 to >80%. Through hydrogenation, the coal moisture content decrease in accordance with the decrease of water content, and the calorific value of the coal will increase. Total hydrogen content was relatively constant, but the corrected hydrogen content was slightly increased, due to the decreased of water content; the content of moisture was very influential to the corrected hydrogen calculation. Whereas, corrected oxygen content decreased sharply or release due to the increase of hydrogen pressure.

Similar to Jambi coal, the ash content of Pendopo coal (Table 3) also increased with the increase of hydrogen pressure. Whereas, moisture content decreased from 17.37 to 3.29% after the hydrogenation with a pressure of 40 bar. Volatile matter of Pendopo coal are fluctuative, volatile matter at 30 bar hydrogen pressure sharply increased and then decreased at 40 bar hydrogen pressure because of the moisture content at 30 hydrogen pressure is decrease. Total hydrogen content decreased, this condition was not in accordance with the purpose, most likely this was due to the hydrogen atoms seamlessly put into the coal in the hydrogenation. The corrected hydrogen content increased on the hydrogen pressure of 40 and 50 bars. Corrected oxygen drastically decreased after hydrogenation process was perfectly proceed. The calorific value drastically increased from 4,932 into the highest level of 6,648 cal/g.

Table 2. The experimental results of Jambi coal hydrogenation with lateritic iron ore catalyst (tempature = 400 ºC)

Parameter	Jambi Coal					
	raw coal	10 bar	20 bar	30 bar	40 bar	50 bar
Moisture (% adb)	10.46	3.86	6.42	5.12	4.20	5.20
Ash (% adb)	2.71	7.56	7.62	7.16	8.22	8.61
Volatile matter (% adb)	45.08	42.26	39.82	43.58	39.57	43.70
Fixed carbon (% adb)	41.75	46.32	46.14	44.14	48.01	42.49
Carbon (% adb)	63.73	82.70	82.00	81.87	80.97	80.88
Hydrogen (% adb)	5.50	5.29	5.29	5.48	5.34	5.35
Hydrogen *	4.34	4.86	4.58	4.91	4.87	4.77
Nitrogen (% adb)	0.85	1.22	1.28	1.20	1.24	1.31
Total sulfur (% adb)	0.08	0.37	0.38	0.36	0.42	0.45
Oxygen (% adb)	27.13	2.86	3.43	3.93	3.81	3.40
Oxygen *	17.83	0.00	0.00	0.00	0.08	0.00
H/C *	0.73	0.68	0.62	0.68	0.69	0.67
O/C *	0.19	0.00	0.00	0.00	0.00	0.00
Calorific value (cal/g. adb)	5.715	6.461	6.351	6.262	6.556	6.088
Free Swelling Index (FSI)	0	1	1	1	1	1.5

Note: adb = air dried basis; * = corrected

Table 3. The experimental results of Pendopo coal hydrogenation with lateritic iron ore catalyst (temperature = 400 ºC)

Parameter	Pendopo Coal					
	raw coal	10 bar	20 bar	30 bar	40 bar	50 bar
Moisture (% adb)	17.37	4.13	4.99	3.91	3.29	4.03
Ash (% adb)	6.19	8.74	8.90	9.25	9.50	10.02
Volatile matter (% adb)	41.54	37.73	37.93	49.12	41.71	41.89
Fixed carbon (% adb)	34.90	49.40	48.19	46.71	45.50	44.06
Carbon (% adb)	53.79	81.34	81.20	80.21	79.83	80.13
Hydrogen (% adb)	6.65	4.98	5.17	5.02	5.14	5.24
Hydrogen *	4.72	4.52	4.62	4.59	4.77	4.79
Nitrogen (% adb)	0.80	1.27	1.29	1.32	1.28	1.25
Total sulfur (% adb)	0.29	0.48	0.46	0.43	0.45	0.46
Oxygen (% adb)	32.28	3.19	2.98	3.77	3.80	2.90
Oxygen *	16.84	0.00	0.00	0.29	0.88	0.00
H/C *	0.87	0.64	0.65	0.66	0.69	0.69
O/C *	0.19	0.00	0.00	0.005	0.01	0.00
Calorific value (cal/g. adb)	4,932	6,648	6,519	6,436	6,359	6,206
Free Swelling Index (FSI)	0	1	1	1	1	1.5

Note: adb = air dried basis; * = corrected

Table 4. Experiment results of Wahau coal hydrogenation with lateritic iron ore catalyst (temperature = 400 ºC)

Parameter	Wahau Coal					
	raw coal	10 bar	20 bar	30 bar	40 bar	50 bar
Moisture (% adb)	12.34	4.01	3.57	2.74	3.47	3.12
Ash (% adb)	2.58	4.06	4.22	4.20	4.81	4.83
Volatile matter (% adb)	48.36	36.15	35.50	37.29	34.37	37.72
Solid carbon (% adb)	36.72	55.78	56.71	55.77	57.35	54.28
Carbon (% adb)	62.32	85.70	81.24	80.98	78.98	81.88
Hydrogen (% adb)	5.80	5.06	5.06	5.10	5.07	5.17
Hydrogen *	4.43	4.61	4.66	4.79	4.68	4.82
Nitrogen (% adb)	0.75	1.23	1.36	1.38	1.40	1.38
Total sulfur (% adb)	0.15	0.32	0.33	0.32	0.36	0.37
Oxygen (% adb)	26.98	3.63	7.79	8.02	9.38	6.37
Oxygen *	16.01	0.07	4.62	5.58	6.30	3.60
H/C *	0.75	0.62	0.66	0.69	0.69	0.68
O/C *	0.17	0.00	0.04	0.05	0.06	0.03
Calorific value (cal/g. adb)	5,819	7,285	7,367	7,327	7,401	7,179
Free Swelling Index (FSI)	0	1	1	1	1	1

Note: adb = air dried basis; *=corrected

The hydrogenation influence by the addition of catalyst on Wahau coal as seen in Table 4 tend to be equal with the influence on Jambi coal. The content of moisture, volatile matter, total oxygen content and corrected oxygen decreased with the increase of hydrogen pressure. The correction of oxygen content decreased sharply at 10 bar of hydrogen pressure, indicated that it is possible to maintain hydrogenation condition at around 10 bar hydrogen pressure. On the contrary total carbon content, corrected hydrogen, total nitrogen and calorific value increased with the increase of hydrogen pressure.

The Hydrogenation Products after Hydrothermal Process (Pre-treatment)

The aim of this part of study was to determine the best pre-treatment process with the lowest oxygen consumption parameters (0%) and the most excellent additives quality with H/C ratio <0.70. The aim could be achieved by examining: the influence of coal pre-treatment method on the oxygen content in the coal, the influence of several ways of pre-treatment on the process characteristics (temperature and pressure) of additive making and additive quality.

One of the pre-treatment methods was hydrothermal. Sakaguci et al. (2008) suggested that the hydrothermal treatment of brown coal that performed at temperatures above 150°C will change the composition of the oxygen functional group and will produce coal with high carbon content and low oxygen contents. Coal products of hydrothermal process were further hydrogenated by lateritic iron ore catalyst. The results were shown in Tables 5, 6 and 7.

Table 5 presents the analysis results after pre-treatment with hydrothermal on Jambi coal at varying hydrogen pressure. It shows that with the adding of water then heated at a temperature of 350°C can reduce the water content from 10.46 to 5.66% and increase the calorific value from 5,715 to 6,730 cal/g. Further, total carbon increased, however hydrogen, nitrogen and oxygen decreased after the hydrothermal process. On the contrary, hydrogen and corrected oxygen content increased with the hydrothermal process due to the influence of the water addition. Similar to the research conducted by Mursito et al. (2010) on peat derived from Pontianak, showed that the hydrothermal processes carried out at temperature of 150 to 380°C, the carbon content of peat

Table 5. The analysis results of Jambi coal after hydrothermal and the catalytic hydrogenation (temperature = 400 ºC)

Parameter	Jambi Coal						
	Raw coal	Hydro thermal	Hydrothermal and Hydrogenation				
			10 bar	20 bar	30 bar	40 bar	50 bar
Moisture (% adb)	10.46	5.66	5.17	5.99	5.66	4.28	6.15
Ash (% adb)	2.71	3.28	6.69	6.90	7.52	8.44	8.08
Volatile matter (% adb)	45.08	42.11	41.03	43.88	45.18	46.19	52.56
Fixed carbon (% adb)	41.75	48.95	47.11	43.23	41.61	41.09	33.21
Carbon (% adb)	63.73	66.60	80.10	80.30	79.31	78.27	77.67
Hydrogen (% adb)	5.50	5.21	5.42	5.76	5.81	5.64	6.30
Hydrogen *	4.34	4.58	4.85	5.09	5.18	5.16	5.62
Nitrogen (% adb)	0.85	0.67	1.81	1.30	1.88	1.34	1.30
Total sulfur (% adb)	0.08	0.16	2.00	2.28	2.41	2.56	2.21
Oxygen (% adb)	27.13	24.08	3.98	3.46	3.07	3.75	4.44
Oxygen *	17.83	19.05	0.00	0.00	0.00	0.00	0.00
H/C *	0.73	0.78	0.69	0.72	0.74	0.76	0.81
O/C *	0.19	0.20	0.00	0.00	0.00	0.00	0.00
Calorific value (cal/g, adb)	5,715	6,730	8,345	8,279	8,371	8,246	8,250
Free Swelling Index (FSI)	0	0	1	1	1	1	1

Note: adb = air dried basis; * = corrected

Table 6. The results of Pendopo coal after hydrothermal process and hydrogenation with catalyst (temperature=400º C)

Parameter	Variable process						
	Raw coal	Hydrothermal bar	Hydrothermal and Hydrogenation				
			10 bar	20 bar	30 bar	40 bar	50 bar
Moisture (% adb)	17.37	3.75	3.69	3.74	6.27	4.58	5.25
Ash (% adb)	6.19	2.46	4.70	5.35	6.01	5.30	3.93
Volatile matter (% adb)	41.54	36.95	38.37	40.32	42.14	45.54	46.38
Fixed carbon (% adb)	34.90	56.84	53.24	50.59	45.58	44.58	44.44
Carbon (% adb)	53.79	71.08	82.24	81.76	80.09	80.42	83.04
Hydrogen (% adb)	6.65	4.71	5.37	5.52	5.77	5.98	6.16
Hydrogen *	4.71	4.29	4.96	5.10	5.07	5.47	5.58
Nitrogen (% adb)	0.80	0.69	1.25	1.27	1.29	1.25	1.77
Total sulfur (% adb)	0.29	0.25	1.58	1.77	1.74	1.75	1.45
Oxygen (% adb)	32.28	20.81	4.86	4.33	5.10	5.30	3.65
Oxygen *	16.84	17.48	1.58	1.01	0.00	1.23	0.00
H/C *	0.87	0.70	0.70	0.72	0.71	0.78	0.76
O/C *	0.19	0.18	0.01	0.01	0.00	0.01	0.00
Calorific value (cal/g. adb)	4,932	7,426	8,759	8,843	8,912	8,993	8,879
Free Swelling Index (FSI)	0	1	1	1	1	1	1

Note: adb = air dried basis; * = corrected

increases due to the increasing temperature. Nakagawa et al. (2004) said that the equilibrium moisture contents and oxygen content decreased after hydrothermal. The equilibrium moisture content was directly related to carboxyl group content

After the hydrothermal process the coal was hydrogenated with variable pressure. The results showed that the calorific value was higher than the calorific value of hydrothermal result, having an average of >8000 cal/g. Ash content and volatile matter increased with the increasing of initial hydrogen pressure. The experiment study of alkaline hydrothermal treatment of high-sulfur, high-ash coal by incrementally adding increasing quantities of NaOH (Mursito et al., 2011) has led us to conclusion, alkaline addition during the hydrothermal treatment process contributes to an increase in coal quality by de-ashing and desulfurization. Hydrogen content steadily increased after the hydrothermal process and hydrogenation while oxygen decreased drastically after the hydrogenation process.

The effect of hydrothermal before hydrogenation in variable hydrogen pressure to the Pendopo coal characteristic seems to be very positive. The calorific value of Pendopo coal, e increase was almost 4,000 cal/g (200%) from the original calorific value of 4,932 cal/g to the highest calorific value of 8,993 cal /g (adb) with hydrogen pressure of 40 bar. Moisture content decreased significantly from 17.37 to 3.69% at 10 bar. Total hydrogen content and corrected hydrogen after hydrothermal seems decreased, then increased after the hydrogenation process. Total oxygen content decreased after hydrothermal process and hydrogenation, corrected oxygen increased after hydrothermal due to the effect of water adding and then decreased after the hydrogenation process in line with the increase in hydrogen pressure due to the effect of hydrogen addition.

Table 7. The results of Wahau coal after the hydrothermal and hydrogenation with catalyst (temperature = 400ºC)

Parameter	Varible process						
	Raw coal	Hydrothermal bar	Hydrothermal and Hydrogenation				
			10 bar	20 bar	30 bar	40 bar	50 bar
Moisture (% adb)	12.34	4.81	2.71	3.78	2.45	6.08	5.54
Ash (% adb)	2.58	9.69	9.99	9.31	10.84	11.13	10.82
Volatile matter (% adb)	48.36	41.72	43.40	43.65	47.94	45.19	53.44
Fixed carbon (% adb)	36.72	43.78	43.90	43.26	38.77	37.60	30.20
Carbon (% adb)	62.32	63.58	75.39	75.90	74.98	74.36	72.98
Hydrogen (% adb)	5.80	4.95	5.53	5.68	5.62	5.74	6.09
Hydrogen *	4.43	4.42	5.23	5.26	5.35	5.06	5.47
Nitrogen (% adb)	0.75	0.61	0.81	0.77	1.24	1.24	1.68
Total sulfur (% adb)	0.15	0.26	2.38	2.12	2.43	2.49	2.41
Oxygen (% adb)	26.98	20.91	5.90	6.22	4.89	5.04	6.02
Oxygen *	14.64	16.10	3.19	2.44	2.44	0.00	0.48
H/C *	0.75	0.79	0.81	0.80	0.83	0.77	0.85
O/C *	0.15	0.18	0.03	0.02	0.02	0.00	0.00
Calorific value (cal/g. adb)	5,819	6,166	8,136	8,056	8,145	8,243	8,156
Free Swelling Index (FSI)	0	0	1	0	1	1	1

Note: adb = air dried basis; * = corrected

Table 7 shows that the condition is similar with Jambi and Pendopo coal in which moisture and oxygen decreased in line with the increase of hydrogen pressure, whereas the content of ash, solid carbon, total carbon, hydrogen, nitrogen and sulfur increased. Hydrothermal treatment above 250°C can increase the hydrogen content of treated coal due to the donation hydrogen reaction of active H and HO radicals coming from water and/or the hydration (Wang et al., 2008).

The Effect of VM/FC Ratio (Fuel Ratio) on Initial Hydrogen Pressure after Hydrothermal and Hydrogenation

The effect of hydrothermal before hydrogenation was very positive, as can be seen from the increasing VM/FC ratio (Figure 1). A steady increase in the VM/FC ratio of Jambi coal reached 1.58. VM/FC ratio of Wahau coal decreased at 40 bar hydrogen pressure then increased sharply to 1.77. VM/FC ratio of Pendopo coal despite the increase only reached 1.04.

Figure 1 shows that the higher the hydrogen pressure, the VM/FC ratio tends to increase. It means the hydrothermal process before hydrogenation successfully input more hydrogen to fixed carbon becoming gas compared to non hydrothermal.

The Influence of Hydrogen Pressure on the Calorific Value in Hydrogenation Process with and without Hydrothermal

The influence of hydrothermal before hydrogenation can also be seen from the data of calorific values before and after the hydrothermal. The result can be seen in Figure 2 and 3.

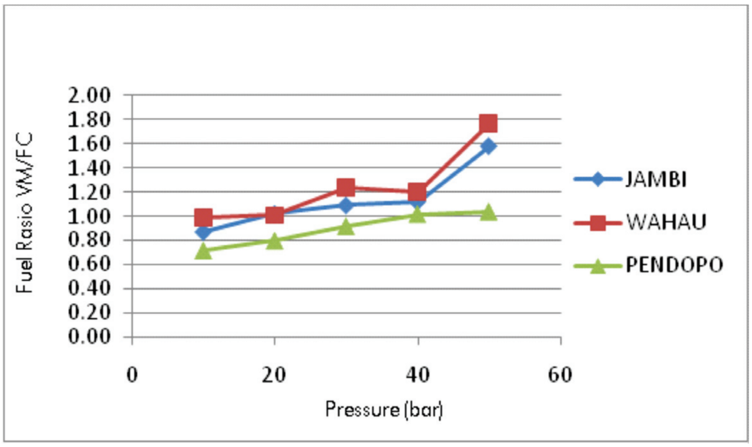

Figure 1. Relationship of VM/FC towards the initial hydrogen pressure after hydrothermal and catalytic hydrogenatio

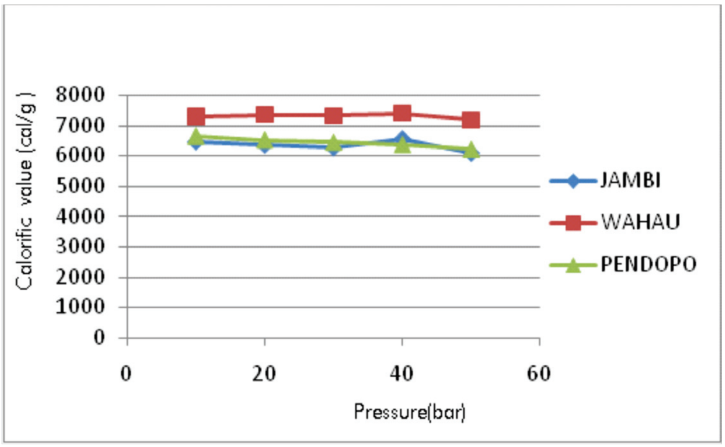

Figure 2. The effect of hydrogen pressure on the calorific value after the hydrogenation process without hydrothermal

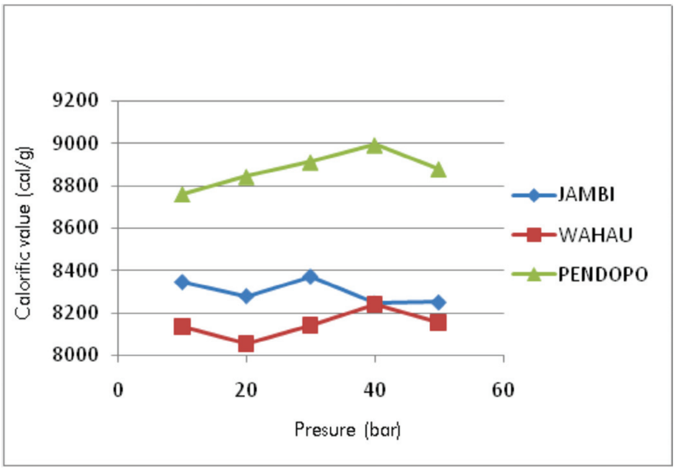

Figure 3. The effect of hydrogen pressure on the calorific value through hydrothermal before hydrogenation process

Figure 2 shows the relationship between hydrogen pressure and calorific value after hydrogenation without undergo the hydrothermal process. The calorific value of Jambi, Pendopo, and Wahau coals increased, but the increase was not significant. With hydrogen pressures >50 bar, their calorific values decreased. Their highest calorific value after hydrogenation process were achieved at 40 bar.

Figure 3 shows the effect of hydrothermal before hydrogenation. The calorific value of Pendopo coal increased significantly at 40 bar hydrogen pressure to 8,993 cal/g (adb), then decreased to 8,879 cal/g (adb). The calorific value of Jambi coal achieved the highest increase at 30 bar hydrogen pressure that was 8,371 cal/g (adb). The calorific value of Wahau coal was relatively small compared to the increase in Jambi and Pendopo coals. The highest calorific value of Wahau coal was obtained at 40 bar hydrogen pressure.

Based on Figure 2 and 3 it can be concluded that the hydrothermal process before hydrogenation can increase the calorific value compared to the hydrogenation without hydrothermal due to the decrease of ash content and the increase of fixed carbon after hydrothermal process.

The Relationship Between the H/C Atomic Ratio and O/C Ratio of the Hydrogenation Product

Based on the chemical structure, coal is a hydrocarbon consisting of a variety of atoms. The number of carbon atoms (C) in coal generally consists of 65-75% of C aromatic type 15-25% of C hydro aromatic type and 5-10% of C aliphatic type. The ratio of hydrogen and carbon (H/C) mole in coal ranges between 0.3 and 0.8. The higher the coal rank, the higher the carbon content while the content of oxygen, hydrogen, and aromatic properties decreased. Therefore, the ratio of hydrogen to carbon (H/C) decreased with the increasing of coal rank. From the observation result, the relationship between H/C with O/C in the hydrogenation products as shown in Figure 4 indicates that the hydrogenation products of Jambi, Pendopo and Wahau coal have H/C ratio of approximately 0.6, so it decreased from the H/C ratio of the original coal (Jambi, Pendopo and Wahau) which ranged between 0.7 and 0.9. This shows that the coal rank of Jambi, Pendopo and Wahau coal increased after hydrogenation.

Figure 5 presents the relationship between H/C with O/C of hydrothermal and hydrogenation products. The result shows that the hydrothermal products and coal hydrogenation had H/C ratio around 0.7, therefore the H/C ratio of hydrothermal coal should be lower than that of raw coal. However, the H/C of hydrothermal coal are almost the same as of the raw coal, suggesting the hydrothermal treatment migh thave certain extent of the hydrogenation to the coal (Shui, et al., 2011). This is likely because after hydrothermal the oxygen content was reduced, so the oxygen consumed hydrogen slightly. Most of the hydrogen coal was used for carbon hydrogenation.

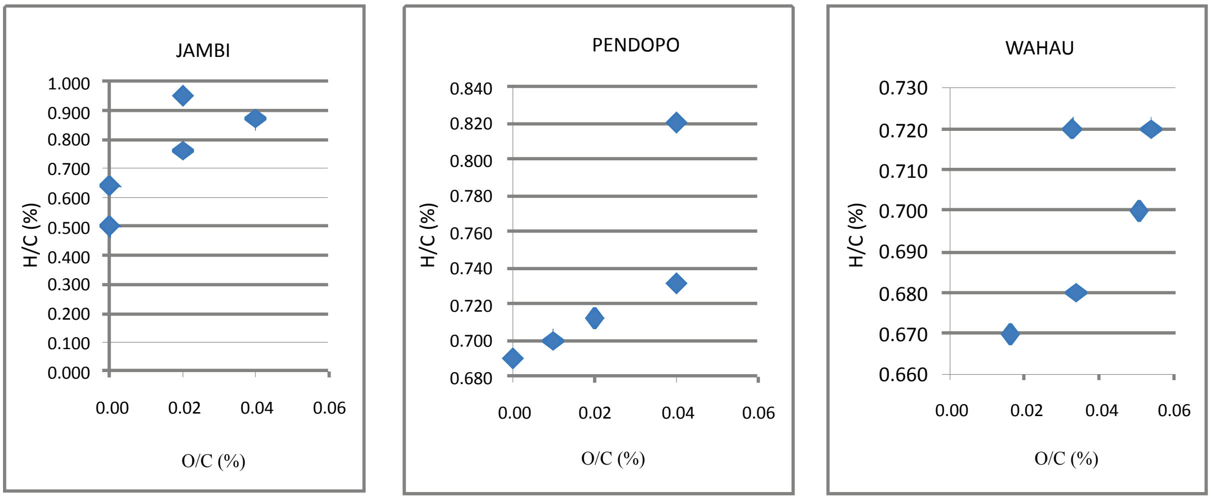

Figure 4. The relationship between H/C with O/C hydrogenation product

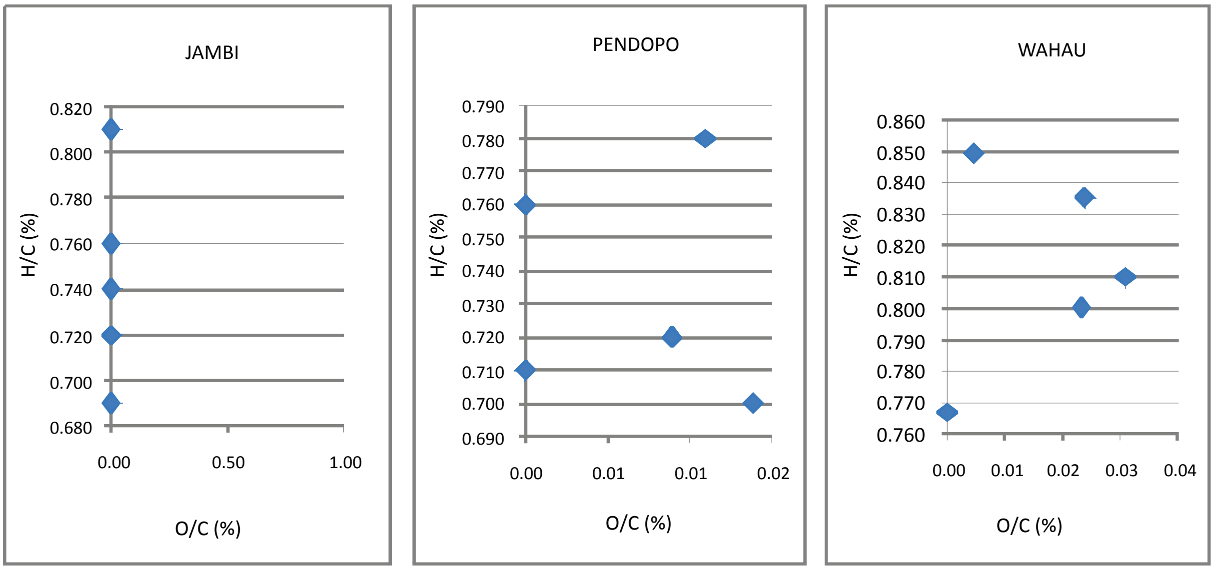

Figure 5. The relationship between H/C with O/C of hydrogenated and hydrothermal products

Characteristics of Coke Product

Hydrogenation products of Jambi, Pendopo and Wahau coal at 30 bar pressure was attempted to make coke. Carbonization results were tested its density load strength and FSI. The success of the research can be determined by comparing the results of coke testing from Jamb, Pendopo and Wahau coals with experimental results of coke in Japan. The results are presented in Table 8.

Table 8 shows that coking of experimental results from Jambi, Pendopo and Wahau coal have load strength of 3.9, 1.2 and 1.5 (kN) and density of 1.46, 104 and 1.32 (g/cm^3) respectively. The lowest density and load strength was Pendopo coke because of the highest moisture content. Compared to the coke from LRC of hydrogenation results in Japan (Ningsih, 2013) , the value of density and load strength were higher. Those results show that coke products tend to have an increasing a better quality.

Table 8. The analysis of FSI, density and pressure strength of coking coal of hydrogenation products at 400ºC and 30 bar

Analysis	Jambi Coke	Pendopo Coke	Wahau Coke	Coke made in Japan
FSI	1	1	1	-
Density g/cm^3	1.46	1.04	1.32	0.96
Load strength kilogram force (kN)	3.9	1.2	1.5	0.9

CONCLUSIONS

From the research results of additive making from low rank coals (Jambi, Pendopo and Wahau) it can be concluded:

– After the hydrogenation process with the adding of catalyst, the total carbon content significantly increased, however, the oxygen content decreased. Ash content increased from 2.7-6.2 to 4.8-10.0 (% adb)

– The process of hydrothermal treatment before hydrogenated produced a higher calorific value compared to the calorific value of the sole hydrogenation process, corrected hydrogen content increased from 4.0 (% adb) to 5.0-6.0 (% adb) while the corrected oxygen decreased sharply from 17.8 to 0.00 (% adb).

– An increase in fuel ratio of (Jambi, Pendopo and Wahau) coal after hydrothermal process reached 1.58, 1.04 and 1.77.

– H/C ratio of hydrogenated products and H/C ratio of hydrothermal and hydrogenated products were between 0.6 and 0.7; decreased from H/C ratio of the original coal (Jambi, Pendopo and Wahau), which ranged between 0.7 and 0.9. This means that the three coals have an increase in their coal ranks, either after hydrogenation or after the hydrothermal then hydrogenation.

– Cokes of experimental results derived from Jambi and Wahau coal have a higher a density and a load strength than the coke resulted from coal hydrogenation in Japan.

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