THE EFFECT OF DISPERSING AND STABILIZING ADDITIVE ON THE STABILITY OF UPGRADED COAL WATER MIXTURE

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ABSTRACT

The upgraded brown coal water mixture (UBCWM) stability with several dispersing and stabilizing additives was studied based on coal sedimentation time history. UBCWM was filled up in a settling column; which has four holes in the upper, upper middle, bottom middle and middle side to take samples. Results indicate that the addition of Naphthalene sulfonate formaldehyde condensate (NSF) as dispersant together with S-194 some kinds of bio polysacharide with long branches produced by Dainippon Pharmaceutical Co. Ltd. as stabilizer, produces UBCWM with the best stability compare with that by using polystyrene sulfonate (PSS) and polymethacrylate (PMA) together with carboxyl methyl cellulose (CMC) and S-60 also some kinds of bio polysacharide, but without branches produced by Dainippon Pharmaceutical Co. Ltd . Even the addition of S-194 results UBCWM with good stability, from the economical point of view, the addition of S-194 is a problem. The price of S-194 is expensive, whereas CMC is abundant and cheap. Therefore, the use of CMC as stabilizing additive was also effective in preparing stable UBCWM according to the stability of the UBCWM by using S-194 and CMC is not significantly different.

Keywords: Additive, upgraded coal, coal water mixture, coal sedimentation

1. INTRODUCTION

Coal is the largest energy resource in Indonesia followed by natural gas, crude oil, hydropower and geothermal. However, oil and natural gas are currently still the main primary energy sources in the country and this pattern has to be changed due to decreasing reserves of oil. The Government's energy diversification policy encourages using coal as a substitute for oil and gas for major domestic industries. Currently, Indonesia's coal resources are estimated at 61.8 billion tons, mostly located in Kalimantan and Sumatera islands (Hadiyanto, 2006). These coals mostly are classified as lignite (58.7%) and the rest are subbituminous (26.7%), bituminous (14.3%) and anthracite (0.3%).

In general, the strategic planning of coal utilization is derived from the quality proportion of the resources in order to meet the market demand and domestic consumption for energy generation. Most of the high rank coal (HRC), which can be used directly, will be allocated for export and domestic use, while the largest portion of the low rank coal (LRC) will be used for mine mouth power plant and undergone through various upgrading and conversion processes or in the form of coal water mixture (CWM) which could be pumped by pipelines and utilized without dewatering.

CWM is coal water slurry in which particles of coal with a certain particle size distribution are suspended in water. Since a mixture of coal and water, CWM is free from some of the major problems

Content

of solid coal, such as powder dust and spontaneous combustion during storage and transportation. Unlike solid coal, moreover, CWM does not require large handling facilities (Hashimoto, 1999).

As mined LRC when mixed with water generally produce CWM with low solid content and low heating value. This CWM is usually unstable and form hard pack sediments quickly. The low heating value and poor storage and flow characteristics of this CWM discourage the use of LRC for preparation of CWM for fuel purposes. To enhance the characteristics of the CWM, the coal is firstly dewatered and upgraded.

Many studies have been conducted on the upgrading of LRC for CWM preparation. Willson, *et al.* (1987), have put forward the hot water drying method (HWD), in comparison with alternate methods. They evaluated the upgraded coal both chemically and physically and measured the rheology of CWM prepared using their upgraded coals. Usui *et al.* (1997) developed an upgrading technique by using the combination of vacuum drying and tar coating processes to study the suitable upgrading process which can produce a highly loaded CWM with reduced viscosity.

In this study, an upgrading method which has been developed and applied, is an upgraded brown coal (UBC) process. The condition of the process at a temperature of about 140°C and pressure of 0.35 MPa, is much milder compared with other upgrading methods (Deguchi, *et al.,* 1999). The addition of low sulfur wax residue (LSWR) of about 1% is very important to prevent the re-absorption of moisture. A pilot plant of 5 tons/day in capacity has been built and operated since 2003 in Palimanan, Cirebon (Daulay *et al.,* 2003). In the future, UBC pilot plant product is available in the form of UBC water mixture (UBCWM).

CWM is required to have as high as possible coal concentration and a moderate viscosity in order to make handling easy. In order to increase the coal concentration and improve the fluidity, the optimization of coal pulverizing method and the use of a suitable additive is also unavoidable. Because the CWM contain solid coal particles in water, unsuitably prepared CWM shows sedimentation of coal particles during long term storage in a tank or during long distance transportation of CWM. Thus it is very important to find a suitable stabilizing additive which can prevent the sedimentation of coal particles which should be used together with a dispersing additive.

The static stability of UBCWM by using an Indonesian upgraded coal produced by UBC process in pilot scale obtains the guiding principal for the preparation of a stable UBCWM. The static stability test was conducted based on sedimentation of the coal in UBCWM by measuring the upper and lower concentration differences. UBCWM was filled up in a settling column with the inner diameter and the height of the glass container are 36 mm and 180 mm, respectively. The settling column has four side holes for sampling purposes to take UBCWM samples (Figure 1).

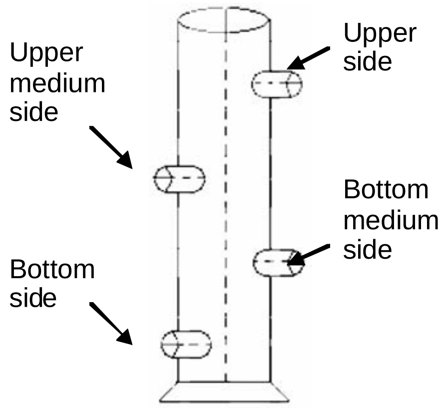

Figure 1. Settling column for UBCWM stability test

2. METHODS

The main purpose of this study is to find the most suitable dispersing and stabilizing additives. An upgraded coal by UBC process from Berau, East Kalimantan was used to prepare UBCWM. The schematic diagram and operational procedure of UBC process has been discussed elsewhere (Deguchi *et al,* 1999, Umar *et al,* 2005). The characteristics of the upgraded coal can be seen in Table 1.

Three kinds of dispersing additives, the naphthalene sulfonate formaldehyde condensate (NSF), polymethacrylate (PMA) and polystyrene sulfonate (PSS) were used in this study. NSF, PMA and PSS have molecular weights of more than ten thousand and exhibit significant dispersing effects on

Content

Table 1. Characteristics of coal used in UBCWM preparation

Moisture in air dried [%]	Ash [%]	Volatile matter [%]	Fixed Carbon [%]	Sulfur [%]	Calorific value [cal/g]
4.81	3.12	46.69	45.38	0.48	6274

the preparation of highly loaded CWM with bituminous coal (Usui *et al.,* 1997).

NSF is a condensation product of formalin and naphthalene sulfonic acid. Sodium and ammonium salts of NSF are cheap and abundant in anionic additives with relatively lower molecular weights produced by SAN NOPCO Ltd. PMA with a molecular weight of 40,000-60,000 produced by NIPPON SHOKUBAI Co., Ltd., and PSS with molecular weight of 10,000-30,000 produced by TOSOH Co. Ltd. These anionic surfactant are adsorbed on the coal surface, and they increase the electrostatic repulsion force between the coal particles (Saeki *et al.,* 1999).

To prevent the sedimentation of coal particle in the mixture, three kinds of stabilizing additives were used as the UBCWM stabilizer. The first was carboxyl methyl cellulose (CMC) an organic material some kinds of clays. This stabilizer reported is very effective in improving the stability of CWM (Saeki *et al.,* 1994). The former additive acts as a dispersing additive and the latter seems to act both as a dispersant and stabilizer i.e. viscosity increasing agent (Usui, *et al.,* 1984). The second and third were S-194 and S-60 produced by Dainippon Pharmaceutical Co. Ltd. These two additives have long molecular chains with a molecular weight of se- veral millions. S-194 has long branches, while S-60 has no branches. The present authors have reported (Saeki *et al.,* 1994) that long molecular chains may produce a network structure in CWM and affect the rheology of CWM and might show good stability.

Suspensions generally show complex flow behaviour. These complexities may be caused by interaction between solid particles and suspension medium. Particle size distribution and surface area characteristics of suspended particles are important factors. According to the previous study, the coal feed for UBCWM preparation was passed through 60 mesh screens (Umar *et al.,* 2006). The solid particle size distribution was measured by using an LA-920 (HORIBA, Ltd.), a laser refraction type particle size measuring instrument. For a homogeneous suspension of the sample, 98% ethanol was used. The weight percent of the solid was about 10 to 30%. From this study, some parameters such as the frequency of particle size distribution, median diameter and surface area can be derived.

UBCWM was prepared by mixing a pulverized coal with distillated water. The variables of this experiment were the kinds of dispersing additive, NSF, PMA and PSS and three kinds of stabilizing additives, CMC, S-194 and S-40 with a fixed amount of 0.3 and 0.01 wt%, respectively. The coal concentration was 60 wt % for additive NSF, and was 57.5% for UBCWM with the addition of PSS and PMA as dispersing additives (Umar *et al.,* 2006). Beside that, UBCWM without the addition of stabilizing additive was also prepared as a reference.

To study the static stability of UBCWM during storage, the laboratory stability test was carried out based on the coal concentration distribution time history. UBCWM samples were taken from the four side holes of the column at the upper, upper medium, bottom medium and bottom side. A sample of about 1 g was placed in a glass container with inner diameter and height are 30 mm and 25 mm, respectively. The glass container including the sample was heated at temperature of 120ºC for 2 hours, was weighed and calculated to the coal concentration. The sample was taken at given time intervals of 0, 10, 20, 30 and 60 days in order to evaluate the sedimentation time history.

For each UBCWM, the apparent viscosity at zero time was measured at temperature of 25°C by using a Brookfield rheometer DV III (spindle RV # 27 was used), at shear rate of 100 s^{-1}.

3. RESULTS AND DISCUSSION

The measurement of particle size distribution is show in Figure 2. It can be seen that there were 2 peaks at 25.9 and 23.1 wt% at the particle size of

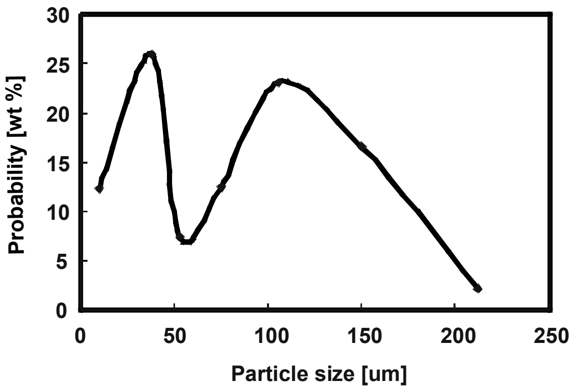

Figure 2. Coal particle size distribution was used for UBCWM preparation

38 and 106 µm, respectively with an average particle size of 61.8 µm and surface area of 3347 cm^2/cm^3. This particle size distribution can be accepted for UBCWM preparation based on the closest packing theory by which interspaces between particles are packed with fine particles having various diameters to obtain densified solid filling factor. However, CWM is necessary to restrict the amount of coarse particles for preventing sedimentation as well as to control the amount of fine particles for reducing a necessary of additive and to maintain the rheological properties at a suitable level. The results of apparent viscosity measurement can be seen in Table 2 and the results of static stability test of the UBCWM at given time interval of 0, 10, 20, 30 and 60 days (Table 3).

UBCWM was generally prepared so that the apparent viscosity gives 1.00 Pa.s at the shearing rate of 100 s^{-1} as a target. As can be seen in Table 2, the apparent viscosity of UBCWBM with the addition of NSF the coal concentration reached 60wt%. Whereas UBCWM using PSS and PMA as dispersing additive, the coal concentration was 57.5 wt%. At the higher concentration level, the apparent viscosity was more than 1.00 Pa.s. This condition will be difficult in handling during transportation and combustion. Therefore, it was difficult to conclude which was the best dispersant to reduce the slurry viscosity.

UBCWM without the addition of a stabilizing additive was lower than that of UBCWM with the addition of stabilizing additives. The effect of each stabilizing additive on the apparent viscosity of UBCWM was not significant at the same concentration level.

From Table 3, it can be seen that all of UBCWM with the addition of stabilizing additive relatively stable after rest for 10 days. The coal concentration is nearly uniform through the depth of the UBCWM layer, although at the day of 20th, 30th and 60th the sedimentation of the coal at the bottom side of the settling column occurred.

3.1 Effect of Dispersing Additive

In order to investigate the influence of dispersing additive on the stability of UBCWM, the coal sedi-

Table 2. The apparent viscosity of UBCWM at shear rate of 100 s^{-1}

UBC concentration	Dispersing additive in 0.3 wt%	Stabilizing additive in 0.01 wt%	Apparent Viscosity [Pa.s]
60 wt%	NSF	*	0.58
		CMC	0.93
		S-194	0.99
		S-60	0.91
57.5 wt%	PSS	*	0.48
		CMC	0.82
		S-194	0.98
		S-60	0.74
57.5 wt%	PMA	*	0.85
		CMC	0.92
		S-194	0.98
		S-60	0.89

Note: (*) without any stabilizing additive

Content

Table 3. The coal concentration in the UBCWM

UBCWM			Coal concentration wt% adb					
		Day		0	10	20	30	60
UBC 60 wt%	NSF 0.3 wt%	-	U		57.1	52.2	46.3	39.5
			UM		59.2	61.3	60.2	61.4
			BM	60.6	62.5	62.8	63.4	65.2
			B		63.7	66.3	72.6	76.3
			B-U		6.6	14.1	26.3	36.8
		CMC 0.01 wt%	U	60.6	60.1	56.8	51.2	47.8
			UM		60.3	60.9	62.1	62.6
			BM		60.7	61.5	62.4	62.9
			B		61.5	63.2	66.8	69.2
			B-U		1.4	6.4	15.6	21.4
		S-194 0.01 wt %	U	60.2	59.9	58.6	54.2	50.2
			UM		60.2	60.4	60.9	61.8
			BM		60.3	60.5	61.1	62.2
			B		60.5	61.5	64.7	66.7
			B-U		0.6	2.9	10.5	16.5
		S-60 0.01 wt%	U	60.7	59.9	56.1	50.9	46.2
			UM		60.2	61.3	62.0	62.9
			BM		61.1	61.6	62.4	63.6
			B		61.7	63.8	67.6	70.2
			B-U		1.8	7.7	16.7	24.0
UBC 57.5 wt%	PSS 0.3 wt%	-	U		54.2	48.7	42.8	38.2
			UM		54.7	56.2	58.7	60.1
			BM	57.5	60.0	60.4	61.1	61.4
			B		61.3	64.8	67.5	70.4
			B-U		7.1	16.1	24.7	32.2
		CMC 0.01 wt%	U		57.3	52.1	47.8	42.8
			UM		57.8	58.1	59.1	60.6
			BM	57.9	57.9	59.2	59.9	60.9
			B		58.8	62.2	64.9	67.4
			B-U		1.5	10.1	17.1	24.6
		S-194 0.01 wt %	U		57.2	54.1	50.2	46.2
			UM		57.6	58.7	59.2	60.0
			BM	57.8	57.9	58.9	59.6	61.2
			B		58.6	59.6	62.3	63.9
			B-U		1.4	5.5	12.1	17.7
		S-60 0.01 wt%	U		57.2	52.4	48.2	43.4
			UM		57.8	58.4	58.9	59.2
			BM	58.1	58.1	58.6	59.2	60.1
			B		59.4	63.1	66.2	69.8
			B-U		2.2	10.7	18.0	26.4
UBC 57.5 wt%	PMA 0.3 wt%	-	U	58.0	54.7	47.4	41.6	36.9
			UM		57.4	58.1	59.2	60.3
			BM		57.8	58.4	60.1	60.8
			B		62.2	68.2	71.2	74.1
			B-U		7.5	20.8	29.6	37.2

Content

Table 3. The coal concentration in the UBCWM

UBCWM			Coal concentration wt% adb					
		Day		0	10	20	30	60
		CMC 0.01 wt%	U		56.8	51.2	46.5	41.5
			UM		57.6	58.2	58.9	59.6
			BM	57.6	57.7	58.8	59.6	60.1
			B		58.5	62.3	65.5	69.3
			B-U		1.7	11.1	19.0	27.8
		S-194 0.01 wt %	U	57.7	56.7	52.7	49.9	45.6
			UM		57.8	58.1	58.7	59.6
			BM		58.1	58.7	59.2	60.1
			B		58.3	61.4	63.1	65.6
			B-U		1.6	8.7	13.2	20.0
		S-60 0.01 wt%	U	57.5	56.0	50.9	47.1	41.9
			UM		57.6	58.1	58.4	59.1
			BM		57.9	58.5	59.0	59.8
			B		58.6	62.6	65.6	69.3
			B-U		2.6	11.7	18.5	27.4

Note: U: Upper side; UM: Upper Middle side; B-U: Bottom side minus upper side; BM: Bottom Middle side; B: Bottom side

mentation history by the addition of NSF, PSS and PMA as dispersing additive was conducted. Figure 4 shows the stability of UBCWM by using the same stabilizer of CMC after 60 days. The differences of coal concentration of the UBCWM between upper side and bottom side of the settling columns were 21.4, 24.6 and 27.8wt% by using NSF, PSS and PMA, respectively.

From Figure 3, it can be seen that the stability of UBCWM by using NSF as dispersing additive is better compared with that of UBCWM using PSS and PMA followed by PSS and PMA, although all of them not significantly show the differences. So, it is difficult to judge which one is the best. However the dispersing additive of NSF could produce UBCWM with higher coal concentration at the same level of viscosity. The higher coal concentration could be cited as easier to burn during UBCWM combustion. Beside that, the price of NSF is the cheapest compare with that of PSS and PMA (Usui *et al.,* 1997).

3.2 Effect of Stabilizing Additive

The effect of stabilizing additive for each dispersing based on coal concentration of the UBCWM after storage in the settling columns after 60 days can be seen in Figure 4 (a, b and c).

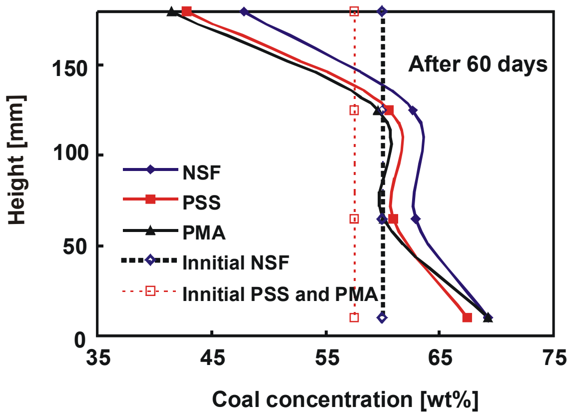

Figure 3. Effect of dispersing additive on UBCWM stability after 60 days

From Figure 4 (a,b and c), it can be seen that the effect of stabilizing additive on UBCWM stability by using NSF, PSS and PMA as dispersing show that the S-194 is the best compared with that of CMC and S-60. The coal concentration shows differences between upper and bottom side of the columns by using NSF without the addition of stabilizer, CMC, S-194 and S-60 were 36.8, 21.4, 16.5 and 24.0 wt% respectively. By using PSS, the differences were 32.2, 24.6, 17.7 and 26.4 wt% and by using PMA the differences were 37.2, 27.8,

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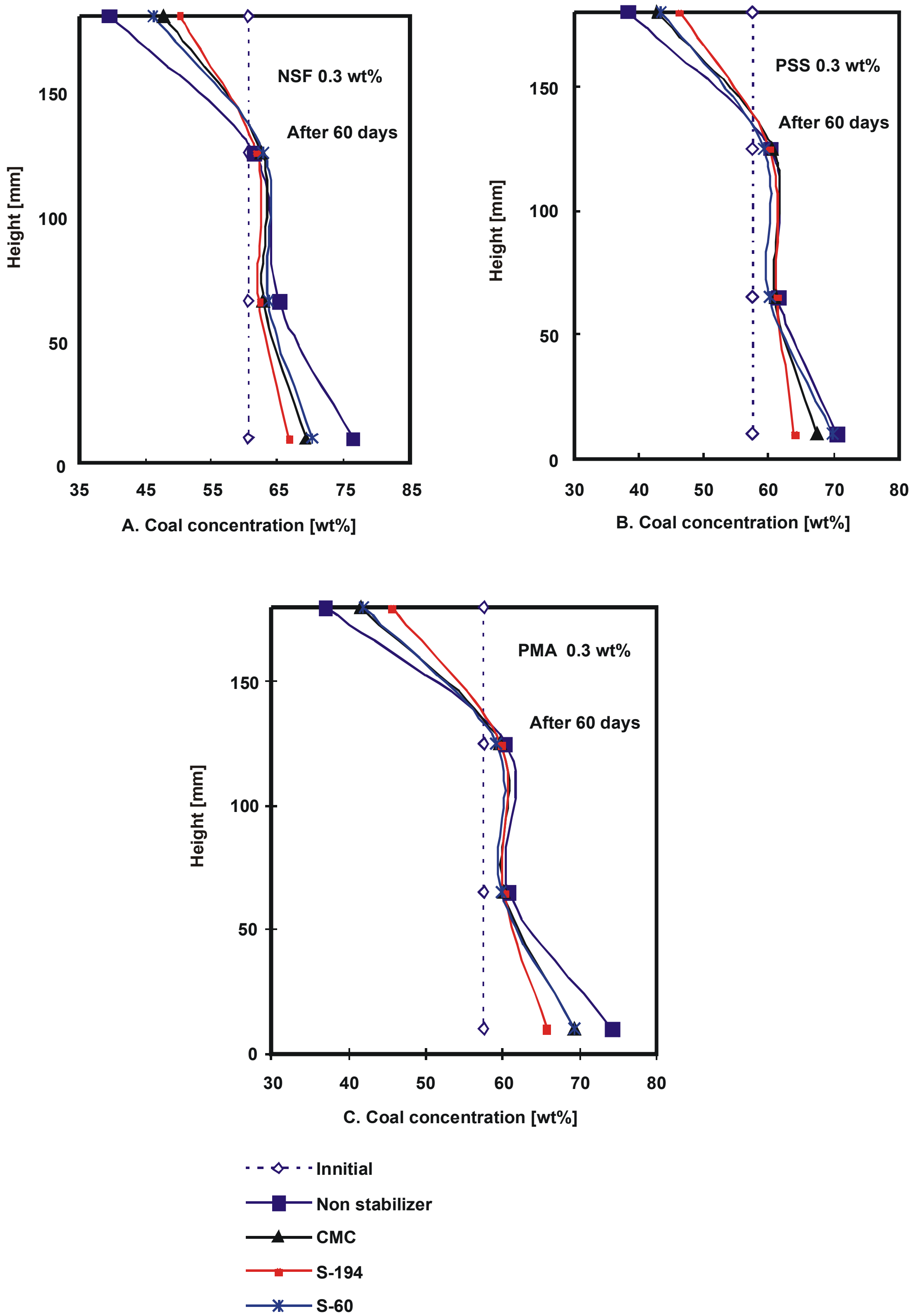

Figure 4. Effect of stabilizing additive on UBCWM stability after 60 days

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20.0 and 27.4 wt% respectively. It could be understood that UBCWM without the addition of stabilizing additive has the poorest stability and shows a significant sedimentation.

So, UBCWM which was prepared only by the addition of dispersing additive without the addition of stabilizing additive is not satisfactory because of unstable condition. It can be explained that the addition of dispersing additive only performs a UBCWM with good fluidity, but not as a stabilizing agent.

4. CONCLUSIONS

- The addition of NSF 0.3 wt% as a dispersing additive, produces UBCWM with the best stability compared with that by the addition of PSS and PMA for each 0.3 wt% after storage for 60 days in the settling columns.

- The use of dispersing additive together with the stabilizing additive is effective in preparing stable UBCWM.

- The addition of S-194 0.01 wt% as stabilizing additive together with a dispersing additive results UBCWM with the best stability followed by CMC and S-60 at the same concentration.

- Since the price of S-194 is expensive, the use of CMC is also effective due to the coal sedimentation history of UBCWM is not significantly difference and the price of CMC is the cheapest compare with that of S-194 and S-60.

- UBCWM produced by the addition of dispersing additive of NSF 0.3 wt% together with stabilizing additive of S-194 or CMC 0.01 wt% has a good fluidity with the apparent viscosity of less than 1.00 Pa.s at the shear rate of 100 s^{-1}.

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REFERENCES

Daulay, B., Salim, A.D., Otaka, Y., Deguchi, T., Makino, E., Rijwan, I. and Umar, D.F., 2003. Pilot plant of upgraded brown coal (UBC) at Palimanan, Cirebon: a challenge of utilizing lignite in Indoneisa. *Proceedings of the 4th International Conference and Exhibition on Coal for Power and Economic Development*, Jakarta.

Deguchi, T., Shigehisa T., and Shimasaki K., 1999. Study on upgraded brown coal process for Indonesian low rank coals, *Proc. International Conference on Clean and Efficient Coal Technology in Power Generation*, Indonesia, pp.176-180.

Hadiyanto, 2006. Anatomi sumber daya batubara serta asumsi pemanfaatan untuk PLTU di Indonesia. *Seminar Nasional Batubara Indonesia.* Pusat Sumber Daya Geologi, Bandung.

Hashimoto, N., 1999. CWM: Its past, present and future, *Coal Preparation* Vol. 21, pp 3-22.

Saeki, T., Usui H., and Ogawa M., 1994. Effect of molecular structure of polysaccharide on the stability of coal water mixtures, *Journal of Chemical Engineering of Japan*, Vol. 27, No. 6, pp. 773-778.

Saeki T., Tatsukawa T. and Usui H., 1999. Preparation techniques of coal water mixtures with upgrading low rank coals, *Coal Preparation*, Vol. 21, pp. 161-176.

Tudor, P. R., Atkinson D., Crawford R. J., and Mainwaring D. E., 1996. The effect of adsorbed and non-adsorbed additives on the stability of coal water suspensions, *Fuel,* Vol. 75 No. 4 pp. 443-452.

Umar, D.F., Usui, H. and Daulay, B., 2005. Characterization of upgraded brown coal, *Coal Preparation,* Vol. 25, No. 1, pp. 313-322.

Umar D. F., Usui H., Komoda Y. and Daulay B., 2006. Preparation of carbonized biomass water mixture and upgraded coal water mixture, *Journal of Chemical Engineering of Japan,* Vol. 37 No. 11, pp. 1206-1212.

Usui, H. and Sano Y., 1984. Thixotropy of highly loaded coal water slurries, *Journal of Chemical Engineering of Japan*, Vol. 17, No 6, pp. 583-588.

Usui, H. and Sano Y., 1986. Thixotropy and stability of highly loaded coal water mixtures *Proc. of World Congress III of Chemical Engineering*, Tokyo, pp 465-468.

Usui, H., Machibara K. and Sano Y., 1987. Effect of stabilizing additives on the stability of coal water mixtures, *Journal of Chemical Engineering of Japan*, Vol. 20 No. 2, pp 192-195.

Usui, H., Saeki T. and Sano Y., 1988. Stability evaluation of coal water mixtures by internal structure stress, *Journal of Chemical Engineering of Japan*, Vol. 21, No. 6, pp 602-607.

Usui H., Tatsukawa T., Saeki T. And Katagiri K., 1997. Rheology of low rank coal slurries prepared by an upgrading process, *Coal Preparation,* Vol. 18, pp119-128.

Willson, W. G., Farnum S. A., Baker G. G. and. Quentin G. H, 1987. Low rank coal slurries for gasification, *Fuel Processing Technology*, 15, pp. 157-172.