

Characterization Of Coal Fly Ash As Converter Catalytic Material In Reducing Hc And Co Exhaust Gas From Motor Vehicles

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Abstract: Coal fly ash is an industrial waste from PLTU industrial activity, such as PLTU Asam-Asam lies in Kab. Tala from South Kalimantan Province which use coal as its fuel. Generally, fly ash would be thrown over landfill or being piled up within industrial area. Piling up of this coal fly ash in certain period would be highly accumulated in certain location and causing problem for environment due to its ash waste which is included in dangerous and toxic waste material (B3) thus creating problem for industry itself. Therefore, utilization effort for this fly ash is highly needed to improve its economy value and to reduce its negative effect toward environment. Objective of this study was to found out fly ash characteristic as converter catalytic material in reducing exhaust gas. Method being used in this study was activating fly ash using H₂SO₄ solution with different concentration from 7M, 8M and 9M. From IR experiment, there were wave number shift from wave number 455.17 cm⁻¹ shifted to 459.03 cm⁻¹ with intensity reduction in which it shows weak Si-O bonds. This phenomenon showed that H₂SO₄ concentration could cause weak interaction of Si-O with pollutant substance thus pollutant substance which exist in the pores of fly ash would be gone and creating a cleaner fly ash. This was strengthened by the missing of absorption band from wave number 2906.53 cm⁻¹. From the result of several treatment in concentration variation using H₂SO₄ solution, it is expected that 9 M H₂SO₄ solution is potential to be used as converter catalytic material, in which using this substance it is expected that coal fly ash would be able to become a converter catalytic which could reduce exhaust gas emission.

Keyword: converter catalytic, fly ash, HC and CO exhaust gas emission

I. Introduction

Waste resulted from activity's residue and mostly not being used again, thus this waste should be thrown away or being manage to be useful. This waste could take liquid and solid form. Fly ash is industrial waste from PLTU, coal fly ash mostly thrown to landfill or just piled-up within industrial area. Piling up of this coal fly ash would be highly accumulated in this industrial location and causing problem for environment. Amount of coal fly ash produced every day could reach 500-1000 ton. Production of fly ash was unavoidable by PLTU, including PLTU Asam-Asam which use coal as its fuel. Combustion in this area would produce coal fly ash which is categorized into B3 waste or a dangerous and toxic waste. This was becoming a problem for PLTU Asam Asam from South Kalimantan Province, thus an effort to be able in utilizing this fly ash is highly needed. Several experiment concerning fly ash utilization was on going to improve its economic value and to reduce its negative effect toward environment. Today, most coal fly ash was used in cement factory as one of the compound material for concrete and as adsorbent. This was due to its porous structure and its large surface area, also carrying chemical content such as SiO₂, Al₂O₃, and MnO. Other than as an adsorbent, coal fly ash was also expected to be able in being used as converter catalytic material which using chemical substance such as Ti, Si, Al and it has lots of common chemical compound with zeolite which had catalytic nature to accelerate reaction, thus by using small treatment it would be able to make coal fly ash into converter catalytic material to reduce exhaust gas emission of motor vehicles.

Exhaust gas of motor vehicles that are dangerous for our health were CO, HC and NO_x produced by imperfect combustion in machine chamber. One that being exposed toward CO would suffer serious toxicity due to relatively slow decomposition of HbCO which could impede molecular activity in its function to carry oxygen to all parts of the body. Hydrocarbon exhaust gas in the air would react with other compound and forming new bond called plicyclic aromatic hydrocarbon (PAH) and if this PAH was enter the lungs, it could create wound and stimulate the forming of cancer cells. The same goes for NO_x gas which also create toxicity in human body and even could paralyze human body. These gas was colorless and odorless, but it contain toxic that highly jeopardize our health. If these gas was not controlled, exhaust gas emission from motor vehicles

would become a serious threat for our environment and our health. One effort that could be done to control exhaust gas emission of motor vehicle was by installing converter catalytic in exhaust gas pipe of motor vehicles.

The background of this study was to utilize coal fly ash as converter catalytic material in reducing CO and HC gas emission from motor vehicles. This was based on the porous structure and large surface area of fly ash, also there were chemical content which has oxidizing function such as contained within zeolite as oxidator and can be used as converter catalytic material. One of this compound was Si and TiO₂ which could accelerate reaction in the hope to create fly ash as a potential material for converter catalytic in reducing exhaust gas emission of motor vehicles. Coal fly ash could be used as an alternative for active carbon and zeolite. However, carbon as combustion residue in fly ash had equal quality with active carbon thus investigation concerning separation of carbon residue would potential in improving the economic value of fly ash. Therefore, by using fly ash as converter catalytic material, it is expected that this treatment would be able to overcome industrial pollution of PLTU in village Asam-Asam, Regency. Tala, South Kalimantan Province and also air pollution due to motor vehicles particularly it HC, CO and NO_x gas emission. The most important thing is that study result showed that based on fly ash characteristic, it could be used as converter catalytic material to reduce exhaust gas emission of motor vehicles.

Several overview was done over catalytic material for converter catalytic other than noble metal such as copper, brass, manganese, zeolite, active carbon from coconut shell. Study conducted by Muhandi (2006) showed that active carbon could absorb lead (Pb), while KrisTri Basuki (2008) using catalyst from coconut shell charcoal with TiO₂, showed 90% reduction of gas absorption for CO and NO₂. Eko Dedy SETiawan (2001) using zeolite catalyst to reduce exhaust gas emission in motor vehicles with 4 step 1 cylinder machine using gasoline fuel. His result showed that zeolite catalyst could reduce HC emission 58,23%, NO_x 50,82% while in CO there were increase for 33,93%.

Objective of this study were a) to found out characteristic of coal fly ash by using H₂SO₄ solution with different concentration as variation, b) making converter catalytic from coal fly ash according to treatment design.

II. Study Method

Time and Place of Study

Study was conducted in Analytical Chemistry Laboratory, Chemical Major, Natural Science and Mathematic Faculty, Brawijaya University. Analysis for fly ash was using XRF instrument and conducted in Common Laboratory, State University of Malang and preparation for coal fly ash was done in Basic Laboratory of Lambung Mangkurat University in Banjarmasin.

Material and Tools

Material used in this study was coal fly ash from PLTU Asam-Asam, South Kalimantan Province, H₂SO₄ p.a (98%, bj 1,84), distillate water and Whatman filter paper no. 41. Steps of this study consist of preparation for fly ash, fly ash activation using H₂SO₄, and characterization of fly ash using XRF and FTIR.

Data Collection Method

Methods used within this activity were:

Preparation and activation of coal fly ash

100 g fly ash was washed with distillate water, dried in oven with temperature 110°C for 3 hours and then placed in desiccators. Dried fly ash was weighted until achieving constant weight and then sieve using 150 mesh sifter. Solid material would be sieve using 180 mesh sifter. Solid material residue in second sieving would be used for next stage.

Fly ash from preparation stage was weighted to achieve 10 g, and then placed into chemical glass and added with 30 mL H₂SO₄ solution with concentration variation of 7M, 8M and 9M. This mixture was immersed for 24 hours and then filtered and washed with distillate water until reaching pH 7. Fly ash after being washed would be dried in oven with temperature 110°C for 3 hours.

Characterization of fly ash using infra red spectrophotometer

Characterization with infra red spectrophotometer was done toward fly ash mixed with KBr dust >200 mesh with ratio 1:3 (sample : KBr dust). This mixture was put into rust proof pil and this pil was put into pellet die. Pellet die was vacuum using hydraulic pump for 2 minutes to form KBr pellet. KBr pellet was put into sample compartment for analysis. Measurement was done using FTIR-8000 PC Shimadzu software in computer with wavelength number 400 cm⁻¹ until 4000 cm⁻¹. Spectra formed would be printed and interpreted using identification table for function cluster.

Characterization of fly ash using X-ray Fluorescence (XRF)

Characterization using X-ray Fluorescence (XRF) was done toward dust sample of fly ash in which sample was put into sample compartment and XRF was done for 10 minutes.

Data Analysis Method

From all this test, analysis would be done toward activation using several variation of H₂SO₄. From laboratory test result regarding characterization of fly ash, it would be interpreted into data. This result would become the first step into making converter catalytic material to reduce exhaust gas emission of HC and CO from motor vehicles.

III. Result And Discussion

Activation of Fly Ash using H₂SO₄

In this study before fly ash could be done as adsorption material or as converter catalytic material for exhaust gas from motor vehicle, coal fly ash should be activated chemically using sulphate acid with variation in concentration using 7, 8 and 9M. Activation chemically using H₂SO₄ was meant to purified pollutant substance within fly ash using H⁺ ion. More H⁺ ion entering fly ash structure meant higher pollutant substance in fly ash being replaced by H⁺ (H-fly ash) thus fly ash would carry active cluster which release proton easily that is Bronsted acid cluster (Poerwadi et al., 1995). Phenomenon in replacement of pollutant substance (X) within fly ash with H⁺ ion during fly ash activation using H₂SO₄ could cause Bronsted acid cluster (Figure 1) (Nicolette R, 2005).

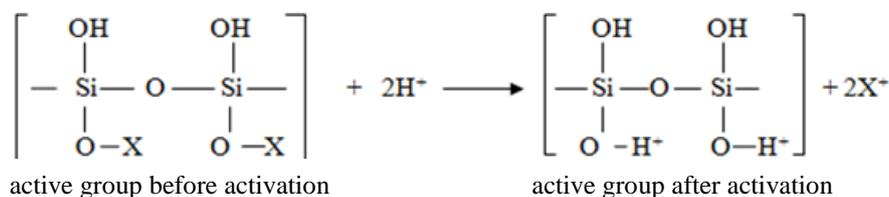


Figure 1. Activation process using acid toward fly ash

We need to found out fly ash element using XRF. For characterization result of fly ash before and after activation using 7, 8 and 9M concentration was shown in Table 1.

Table 1. Composition of fly ash from XRF

No	Element	Percentage			
		Yet Activation	H ₂ SO ₄ 7M	H ₂ SO ₄ 8M	H ₂ SO ₄ 9M
1	Fe	58.84	38.23	36.10	29.40
2	Ca	13.30	7.49	5.98	6.51
3	Si	16.90	34.5	40.20	47.7
4	Al	4.20	5.10	5.50	5.70
5	K	0.88	1.50	1.65	2.11
6	Ti	1.32	1.96	2.13	2.49
7	V	0.05	0.05	0.05	0.08
8	Cr	0.14	0.11	0.11	0.11
9	Mn	0.75	0.44	0.42	0.43
10	Ni	0.89	0.91	0.95	1.01
11	Cu	0.15	0.17	0.18	0.19
12	Zn	0.01	0.05	0.05	0.08
13	Sr	0.71	0.81	0.86	0.75
14	Ba	0.81	1.80	1.80	1.50
15	Eu	0.60	0.30	0.30	0.20
16	Mo	0.00	6.49	3.10	3.10
17	Re	0.50	0.20	0.20	0.20

Based on data in Table 1, Fe and Si had higher percentage compared to other element in fly ash. Fly ash content before activation was quite high with 58.84% while Si content was only 16.90%. After activation using H₂SO₄, with concentration 7M and 8M, Si element start to increase into 34.50% and decreasing into 36.10%, however, Fe element still higher with 38.23% and 36.10%. After activation with 9M H₂SO₄, Si element increasing into 47.70% while Fe element decreasing into 29.40%, this was due to replacement of Fe²⁺ with H⁺ ion as a result from fly ash activation using H₂SO₄ as being shown in reaction of Figure 1. From XRF data it is found out that activation using 9M H₂SO₄ is the best result, and later it is expected to obtain good adsorption/catalyst capacity in reducing HC and CO exhaust gas of motor vehicles.

To found out active cluster in coal fly ash, characterization was done using FTIR. For fly ash characterization using FTIR before and after activation using 9M H2SO4 was showed in Figure 2.

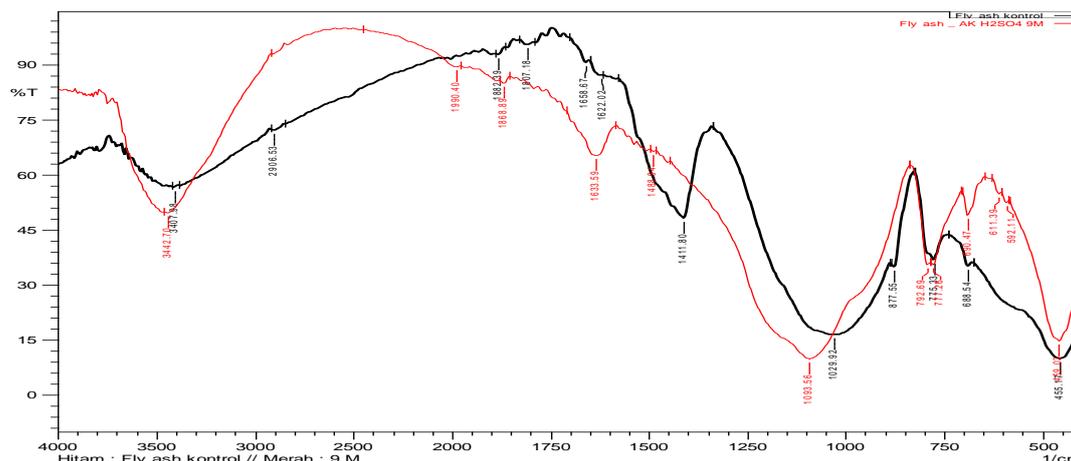


Figure 2. IR spectrum of coal fly ash. Fly ash curve before activation (black), fly ash curve after activation using H2SO4 (red)

Based on spectrum data in Figure 2, fly ash spectra before activation showed absorbance band in wave number 3407.98 cm⁻¹ which is extension vibration of –OH cluster, wave number 1400.80 cm⁻¹ which is bending vibration of –OH cluster from H₂O molecules being absorbed, wave number 1029.92 cm⁻¹ which is wide and with sharp intensity had showed asymmetrical span vibration of Si-O-Si which show the presence of Si-O cluster or Al-O cluster in TO₄ bonding with Si-OH structure. Width of the peak showed that there were lots of Si-OH cluster thus crystallinity in fly ash structure is decreasing. Absorbance in 775.33 cm⁻¹ showed symmetrical span vibration of Si-O-Si followed by bending mode of Si-O in number 455.17 cm⁻¹, showing porous structure of fly ash.

After fly ash was activated using H₂SO₄, it showed that absorbance band in wave number 3407.98 cm⁻¹ was shifted to 3442.70 cm⁻¹ along with intensity decrease. Shift was also occur in wave number 1400.80 cm⁻¹ toward 1633.59 cm⁻¹ showing weak interaction of –OH cluster. This indicated that acid treatment would disrupt structure–OH cluster due to release of water molecule bonded physically in fly ash. The emergence of new absorbance band in wave number 792.69 cm⁻¹, 777.26 cm⁻¹ and 1093.56 cm⁻¹ is asymmetrical span vibration of O-Si-O. Shift was also occurs in wave number 455.17 cm⁻¹ which shifted into 459.03 cm⁻¹ along with decreasing intensity which showed that Si-O bond is weak. This phenomenon proved that concentration of H₂SO₄ could cause weak interaction of Si-O cluster with pollutant substance thus pollutant substance in fly ash pores would be gone and creating cleaner pores of fly ash. This was strengthened by the loss of absorbance band for wave number 2906.53 cm⁻¹.

IR analysis of fly ash before and after activation was given in Table 2.

Table 2. Fly ash analysis before and after activation

No	Type of Vibration	Wavenumber (cm ⁻¹)	
		Fly ash before activation	Fly ash after activation
1	-OH stretch from Si-OH or water	3407.98	3442.70
2	-CH ₃ bend	2906.53	-
3	-OH bend (OH bending vibration of water molecules)	1622.02	1633.59
4	C=O stretch of carboxyl	1411.80	-
5	Si-O stretch asymmetrical from Si-O-Si	-	Serapan baru 1157,21; 1114,78 dan 1095,13
6	Si-O stretch from Si-OH	1029.92	1093.56
7	Si-O bend from Si-O-Si	775,33	792,69 dan 777.26
8	T-O (T = Fe or Al) bend	455.17	459.03

IV. Conclusion

From this study it can be concluded that after activation using H₂SO₄ with concentration 9M, Si and Ti compound had improve to 47.70% and 2.49% while Fe compound was decreasing to 29.40%. With increase of Si and Ti from coal fly ash di PLTU Asam Asam from South Kalimantan, it is expected that this result had the potential to become converter catalytic material in reducing exhaust gas emission of HC and CO from motor vehicles.

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Reference

- [1]. Catalyst for claisen-Schmidt Condensation Reaction, American chemical Science journal
- [2]. Deepti Jain, Ash Rani (2011), MgO Enriched Coal Fly Ash as Highly Avctive heterogenous Base
- [3]. Eko Dedy Setiawan, 2001, Menggunakan katalis Zeolit untuk mengurangi emisi gas buang pada motor bensin 4 langkah 1 silinder berbahan bakar bensin (Using a zeolite catalyst to reduce exhaust emissions in gasoline engine 1 cylinder 4 stroke gasoline)
- [4]. Fuller, M. J and Warwick, M.E, 1976 The Catalytic Reduction of Nitric Oxid By CO Over SnO₂ – CuO Gells". Journal of Catalysis. 42 p.418-424
- [5]. Gulari, E, Guldur, C., Srivananavit, S, and Osuwan, S.1999, The development Oxide based Catalys "CO Oxidation by Silver Cobalt Composite Oxide". Applied Catalys A; General 184.P147-163
- [6]. Kim, D.S and Wachs, I.E, 1993."Suface Chemistry of Suported Chromium Oxide Catalysts". Journal of Catalysis. 142. P.166-171
- [7]. Kris Tri Basuki 2008, penurunan konsentrasi Co dan NO₂ pada emisi gas buang menggunakan arang tempurung kelapa yang disisipi TiO₂ (a decrease in the concentration of CO and NO₂ in the exhaust emissions using coconut shell charcoal is inserted TiO₂)
- [8]. Pelangi, 1999, Upaya Mengurangi Emisi Gas Buang Kendaraan Bermotor (Jurnal). (Efforts to Reduce Emissions Vehicle (Journal)
- [9]. Philip Kristanto, Johan Wahyudi, 2008, Reduksi Emisi Gas Buang CO dan HC Pada Motor Bensin Dengan Katalis Zeolit. jurnal Kimia Lingkungan Vol.10 (Exhaust Emission Reduction of CO and HC On Motor Gasoline With Zeolite Catalysts. journal Environmental Chemistry Vol.10) ISSN 1411-1543
- [10]. Susanto, 2006, Jurnal PMKI ITS Surabaya Jurusan Kimia."Studi Penggunaan katalis CuO/y-Al₂O₃ sebagai katalitik konverter untuk mereduksi emisi CO.p.3-5 (Journal PMKI ITS Department of Chemistry. "Study Use of catalyst CuO / y-Al₂O₃ as catalytic converters to reduce emissions CO.p.3-5)