Engineering Journal issn: 0125-8281

# Improvement of Pulp and Paper Mill Industries Effluent Quality Using Bagasse Fly Ash

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**Abstract.** Color removal from pulp and paper mill wastewater by using fly ash was one of the effective and low-cost methodologies to improve the quality of the wastewater. The fly ash had the capability to remove various organic pollutants, such as Lignin, Tannins, Humic, and Fulvic, from wastewater. These organic pollutants contribute to the color intensity of industrial effluent. The research examined the removal capability of fly ash including Bagasse fly ash, Rice husk ash and Lignite fly ash. The samplings were analyzed in the laboratory for color removal capability. The test was divided into 3 steps as follows: Step 1 the comparison of color removal efficiency by using 3 types of fly ashes; Bagasse fly ash, Lignite fly ash and Rice Husk ash. Step 2: Analyzing the functional group of fly ashes before and after color removal by using the Fourier Transform Infrared Spectroscopy (FT-IR) to determine the adsorption mechanism and other influencing factors. Step 3: Color removal efficiency by sand filter experiment. The results showed that; Step 1: Bagasse fly ash, Rice husk ash and Lignite fly ash were effective in color removal of 94.2, 74.8 and 71.8 percent at 30 minutes, 24 hours and 4 days, respectively. The pH of the effluent using Bagasse fly ash was constant to 7.5 - 8 and tend to be constant although the ash amount increased, while the pH of Lignite fly ash was in the range of 8 - 9 and tend to increase when amount increased. Step 2: Analyzing the functional group of Bagasse fly ash using the FT-IR found that after using Bagasse fly ash for color removal in the effluent, the graph tends to decrease. Step 3: The result showed that the efficiency of color removal of the sand filter by soil mixed with Bagasse fly ash at ratio 1: 0 - 1:10 (Soil : Bagasse fly ash), which the efficiency of color removal tends to increase with the maximum of 93 % and the minimum of 16% at ratio 1:7 and 1:0, respectively.

**Keywords:** Wastewater, color removal, bagasse fly ash, FT-IR, pulp and paper mill.

**ENGINEERING JOURNAL** Volume 22 Issue 4
Received 30 November 2017
Accepted 2 May 2018
Published 31 July 2018
Online at http://www.engj.org/
DOI:10.4186/ej.2018.22.4.13

DOI:10.4186/ej.2018.22.4.13

## 1. Introduction

It is a fact that water is essential for human, agriculture, and animal. Furthermore, water contributes to the abundance of other natural resources. The water consumption has been increasing dramatically due to the rapidly changes of economic and social development together with the expansion of agriculture and industry, as well as the climate change. It has been estimated that in the next 30 years, the world population will increase to about 9.2 billion people [1]. This will inevitably increase water consumption dramatically. Water consumption can be divided into two categories, consumptive use and non-consumptive use. Consumptive use means water that cannot reused immediately such as infiltration and evaporation, including water which is attached to agricultural products or food, whereas non-consumptive use means wastewater which is treated and released into the natural water to be reused again.

Nowadays, economic and social development together with the expansion of agriculture and industry contribute to higher water consumption, comparing with the beginning of the century. In fact, the water use in agriculture is 5 times more than that of the past, while the water use in industry is 26 times more than that of the past [2]. As a result, the effluent has been increasing significantly. However, only 60-80% of the industrial wastewater is treated in accordance with the standard procedure, while the other passes through no treatment before leaving into the natural river [3]. When the waste increased, there is a need for more disposal area. This corresponds with the pollution statistics which the amount of industrial waste increase rapidly every year. Therefore, the effluent management system of the industrial sector, where pollution is created, has been being received much attention. In addition, because of the driven by domestic and international laws, the competition in global trade as well as the pressure from the society, there is a requirement to reduce emissions for sustainable communities [4].

Wastewater management for the collection, treatment and disposal of wastewater from the plants which are water-intensive industries is becoming more and more importance. Therefore, the Thai government has issued the new environmental law, called "Zero Discharge" which means no drainage from the factories. The Thai government has been enforcing the law to the factories which release effluent into the Chao Phraya River. As a consequence, this raises awareness of many industry factories in the area. The concept of "Zero Discharge" is water treatment and reuse or wastewater as a resource for specific use in the factories. For example, some industries that use large amounts of water for production, such as paper industry and tanning industry, use wastewater as a resource for cropping. This method will not only reduce the pollutants of the effluent but also help the plants grow well. In fact, the plants will grow better than the ones using irrigation because of organic and inorganic in the effluent [5]. However, when the factory has expanded its production capacity, it will use more water, resulting in more effluent. Therefore, the soil will be filled with more wastewater. As a result, there is not enough time for the soil to return to normal condition. This result in wastewater is almost impossible to infiltrate into the subsurface soil which affected the growth of plants. The explanation is that an increase in soil water content often causes a reduction in soil aeration. In addition, some of the effluent will return to the surface soil due to the soil permeability decrease or the gap between the soils is blocked by sewage sludge. The amount of sewage sludge depends on the color intensity of industrial effluent discharged each day. A preliminary study showed that there are a lot of effective methodologies to remove color from effluent. However, these methodologies are so complicate. Consider these methodologies for instance, ozone oxidation for wastewater treatment can reduce the color intensity from effluent by 66.98% but requiring factories to maintain the pH of wastewater to 12 [6]. Burning sewage sludge from paper mill effluents at the temperature of 300 ° C can reduce the color intensity by 95% [7]. Fungal treatment can reduce the color intensity from wastewater by 50-60%. Moreover, it can reduce COD by 15-45% within 2 days [8]. Coagulation treatment of wastewater using Polyaluminium Chloride can reduce the color intensity from effluent by 92% and the optimum pH value for this methodology is 5.0 [9]. The use of agricultural residues for wastewater treatment can not only reduce the color intensity from effluent by 98% but it is also a cost-effective method. These agricultural residues which can be used for wastewater treatment are activated carbon prepared from rubber seed, activated carbon derived from Palm Oil Shell, husk and bagasse fly ash [10], [11], [12], [13].

As a result, wastewater treatment by using wastewater as a resource for specific use or soil permeation should be used together with improving the effluent quality by reducing the color intensity or removing sludge, such as Lignin, Tannins, Humic, and Fulvic. Consequently, the effective method is to improve the effluent quality by color removal first then release it into the plantation field [14]. In particular, the color in the effluent is called "the true color" because it is caused by the degradation of organisms and the yield of the metamorphic process of life which is high stable and difficult to decompose and separate by filtration

[15]. In addition, traditional biological and chemical treatment processes was not effective to remove organic substance which degrades in water [16], [17]. This research uses 3 types of fly ash which are the remaining of industrial heating process such as Bagasse fly ash, Rice husk ash and Lignite fly ash. The fly ash is used in sand filter system for removal color. Then, the effluent is released into the plantation field.

## 2. Materials and Methods

### 2.1. Materials

The materials using in the experiment including:

1. **Pulp and paper mill industries effluent**: The characteristics of pulp and paper mill industries effluent as shown in Table 1.

Table 1. Effluent quality indexes of pulp and paper mill industries.

| Parameter | Month | | | Effluent standard |
|---|---|---|---|---|
| | April 2015 | May 2015 | June2015 | |
| pH | 7.865 | 7.935 | 7.745 | 6.0 - 8.0(1) |
| Temperature (°C) | 38.45 | 31.75 | 32.5 | ≤ 40(2) |
| Conductivity (μS/cm) | 3070 | 3865 | 3125 | Not specify(2) |
| Total dissolved solids (mg/l) | 1886 | 2235 | 1976 | ≤ 3000(2) |
| Biochemical oxygen demand (mg/l) | 6.55 | 5 | 6.3 | ≤ 10(1) |
| Chemical oxygen demand (mg/l) | 150.5 | 146 | 181 | ≤ 360(1) |
| Colors (Pt-Co) | 400 | 500 | 500 | ≤300 ADMI(3) |

(1) Water quality standard using in the Environmental Impact Assessments report (2 modifier edition) of Phoenix Pulp and Paper Public Company Limited (PPPC) at No. TS. 1009.3/11066 at 8 October 2014 [18].
(2) Water quality standard under the Ministry of Industry, the Notification on the Standard on Discharging of Wastewaters., B.E. 2539. [18]
(3) Water quality standard under the Ministry of Industry, the Notification on the Standard on Discharging of Wastewaters., B.E. 2560, effective from 7 June 2017.

2. **Quality improvement materials**: Using 3 types of ash; Lignite fly ash (Fig. 1(a)), Rice Husk ash (Fig. 1(b)) and Bagasse fly ash (Fig. 1(c)) which the physical properties as shown in Table 2.

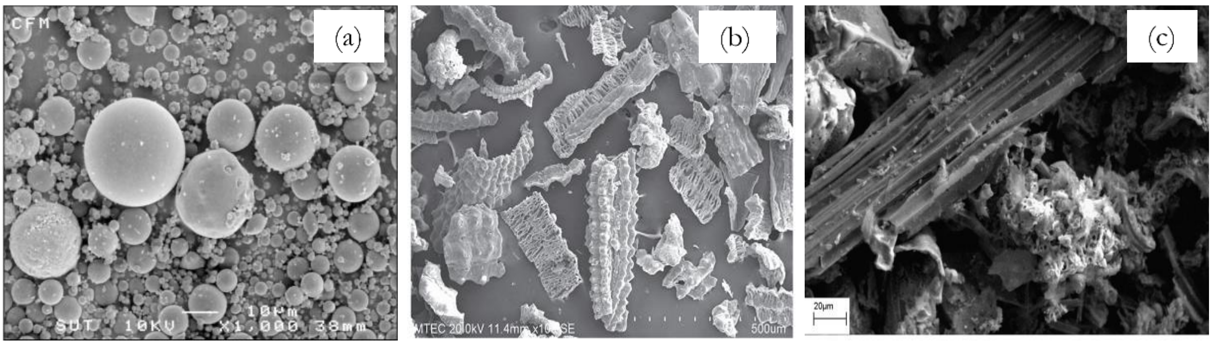



Fig. 1. Scanning electron microscopies (SEM) of (a) Particle size of Lignite fly ash was 28.5 μm [19], (b) Particle size of Rice Husk ash was 4 – 75 μm [20] and, (c) Particle size of Bagasse fly ash was 23(d50) μm [21].

Table 2. Physical properties of bagasse fly ash and rice husk ash.

| Physical Properties | Specific surface area ($cm^2/g$) ( Blaine air permeability test) | Specific gravity |
|---|---|---|
| Bagasse fly ash | 2871.75 | 2.11 |
| Rice Husk ash | 1134.9 | 2.1 |
| Lignite fly ash from Mea Moh, Lampang, Thailand [19] | 2370 | 2.02 |

From Table 2 it indicated that the specific gravity of 3 types ash were similar but Bagasse fly ash presents a higher specific surface area compared with Lignite fly ash and Rice Husk ash, attributed to its high porous and fineness structure.

3. **Sand filter**: The efficiency of color removal by sand filter was determined by using acrylic column which diameter was 0.15 meter, high 0.1 meter as shown in Fig. 2, which each column from bottom to top contained rock, gravel, coarse sand and soil samples in the experiment field which mixed fly ash at the ratio 1:0 – 1:10 (Soil : Fly ash).

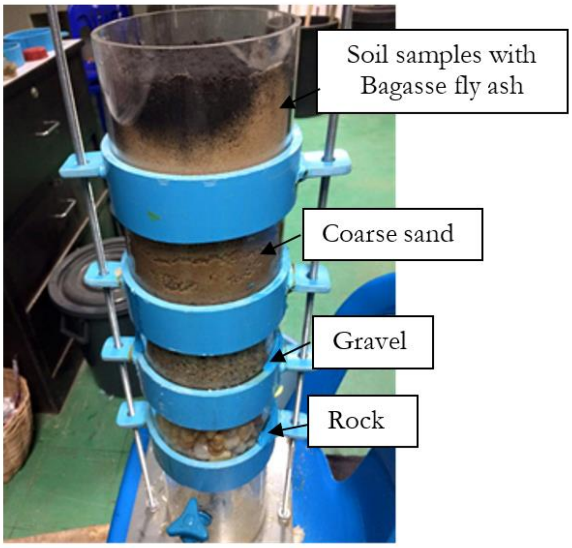



Fig. 2. Sand filter in acrylic column.

### 2.2. Methods

The experiment was divided into 3 steps as follows: Step 1 the comparison of color removal efficiency by using 3 types of ash; Bagasse fly ash, Lignite fly ash and Rice Husk ash. The comparison was calculated by using UV - Visible Spectrophotometer to select material which had the highest efficiency for color removal in the effluent and without the pH changes. Step 2: Analyze the functional group of fly ash before and after color removal by using the FT-IR to determine the color-removal behavior of the fly ash. Step 3: Color removal efficiency by sand filter experiment. The test steps were as follows:

**Step 1. The appropriate amount of fly ash**: The appropriate amount of fly ash for color removal was determined by using 150 ml of effluent per to 1-10 grams of ash, with a total of 60 minutes of adsorption.

**Step 1.1 The equilibrium time**: The equilibrium time of color removal was determined by using 150 ml of effluent and applying to the appropriate amount of ash according to the step 1 for 0.5,1,2,3, 4 and 5 hours.

**Step 1.2 pH test**: pH is considered as an importance variable for using effluent for plantation. Therefore, the pH of effluent from mixed fly ash must be investigated before applying to plantation field. This experiment measures the pH of the effluent when mixing with fly ash in a ratio of 0 to 10 grams, using the equilibrium time from step 1.1.

**Step 2. The analysis of functional groups of fly ash before and after color removal**: Analysis of fly ash behavior before and after of color removal by the Infrared absorption using the FT-IR. The analysis of dried 3 samples were; fly ash, fly ash absorbing effluent for the period of 24 hours and sludge of effluent.

**Step 3. Color removal efficiency by filter experiment**: The experiment performed by fulfill water into the column layers and allowing the effluent pass through the mixed layers until exhausted in order to determine the efficiency of color removal in effluent.

## 3. Results and Discussion

**Step 1. The appropriate amount of fly ash**: The results of this study showed that the color removal efficiency of the various 3 types of ash with increasing amount of ash, tend to increase until 7 grams' amount of ash then it got into equilibrium state, as shown in Fig. 3 which the efficiency of color removal for three types of fly ash, Lignite fly ash, Rice husk ash and Bagasse fly ash were 46.3, 17.4 and 85.8 percent, respectively.

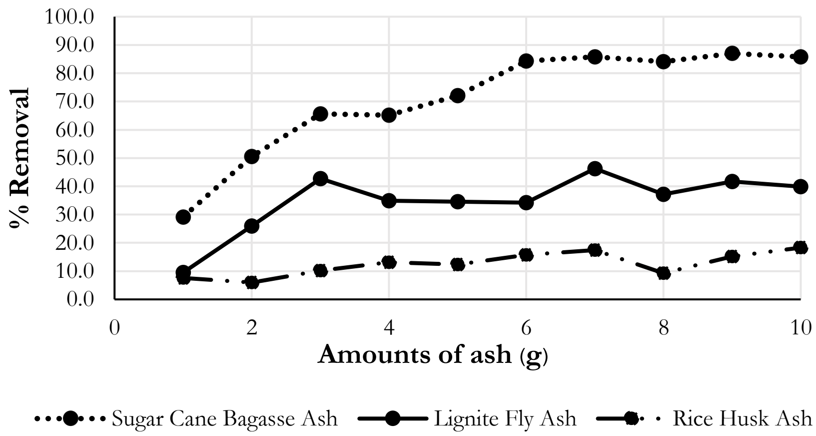



Fig. 3. The relationship between percent of color removal and dosage of three types of ash.

**Step 1.1 The equilibrium time**: The results of this study showed that the color removal efficiency of Lignite fly ash and Bagasse fly ash varied with time, the efficiency of color removal for Bagasse fly ash was 94.2 percent at 30 minutes, then after 30 minutes it got into equilibrium state. The efficiency of color removal for Lignite fly ash was 78.9 percent at 1 day, then after 1 day it got into equilibrium state. The adsorption time had increased, in the beginning, adsorption increased rapidly due to free surface of the adsorbent high capacity then after a longer period of time the adsorption will get into the equilibrium state or absorb more slightly [22], as shown in Fig. 4.

DOI:10.4186/ej.2018.22.4.13

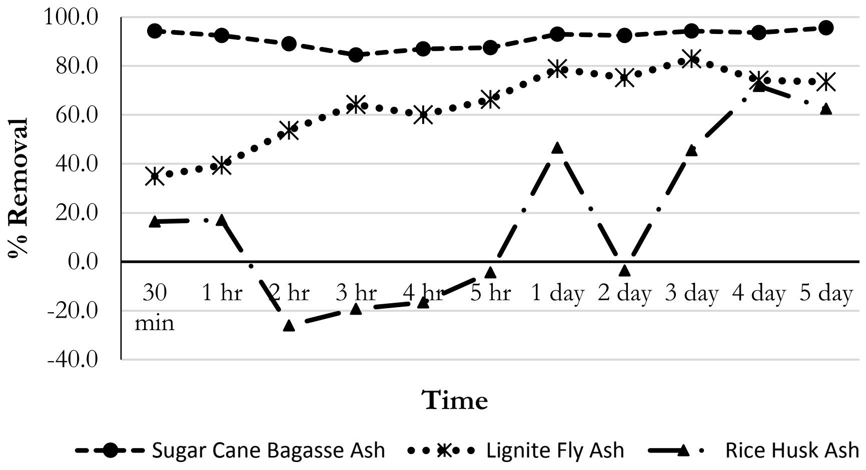
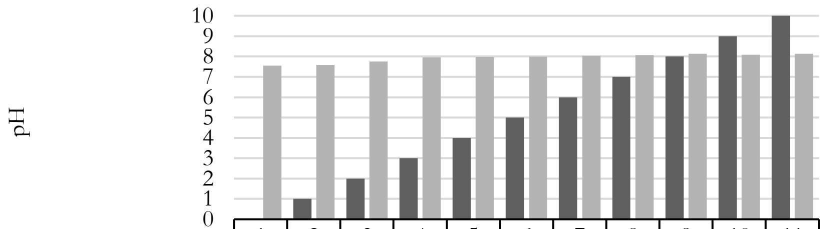



Fig. 4. The relationship between percent of color removal and the adsorption time of three types of ash.

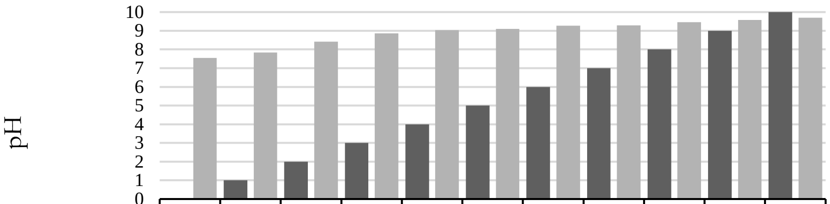
From Fig. 4, the relationship between percent of color removal and the adsorption time of Rice husk ash showed that the color removal efficiency tended to unstable, some samples were higher concentration of color in the effluent such as 2 hr., 3 hr., 4 hr. and 5 hr., with the highest color removal efficiency at 71.8 percent at 4 days. Therefore, using only Lignite fly ash and Bagasse fly ash were further tested in the next steps.

**Step 1.2 pH Test**: The pH test for Bagasse fly ash and Lignite fly ash were tested with 1 to 10 grams of ash at 30 minutes of adsorption time (results of previous step). The results showed that the pH of both effluent before mixing the ash was 7.54, when the bagasse fly ash was mixed from 1-10 grams into the effluent, the pH of the effluent was increasing and constant in the pH range of 8. In comparison, tested by mixing the effluent with Lignite fly ash at 1-10 grams, the pH of the effluent tended to increase. When using more than 3 grams of Lignite fly ash to make the pH higher than 9 which exceeded the standard of effluent quality to be defined, as shown in Figs. 5 and 6. Therefore, after the improvement of water quality with Lignite fly ash, the pH should be adjusted down to the standard [23].




| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Sugarcane Bagasse Ash (g) | 0 | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| pH | 7.54 | 7.59 | 7.76 | 7.95 | 7.97 | 8.00 | 8.04 | 8.07 | 8.13 | 8.09 | 8.14 |

Fig. 5. The relationship between pH and amount of bagasse fly ash.




| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Lignite fly ash (g) | 0 | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| pH | 7.54 | 7.83 | 8.41 | 8.86 | 9.04 | 9.10 | 9.26 | 9.30 | 9.46 | 9.57 | 9.70 |

Fig. 6. The relationship between pH and amount of Lignite fly ash.

From Figs. 5 and 6 Bagasse fly ash was the best materials for improving effluent, the color removal efficiency can be as high as 94.2% compare to Biological Treatment which can only reduce the color intensity by 30% [6]. Apart from this, the ratio of the Bagasse fly ash when mixing with effluent resulting in pH changes was in the range of 7-8. Therefore, using only Bagasse fly ash was further tested in the next steps.

**Step 2. The analysis of functional groups of fly ash before and after color removal**: The FT-IR was used for analysis of functional groups of Bagasse fly ash absorbent before and after color removal. The result was shown in Figs. 7 and 8.

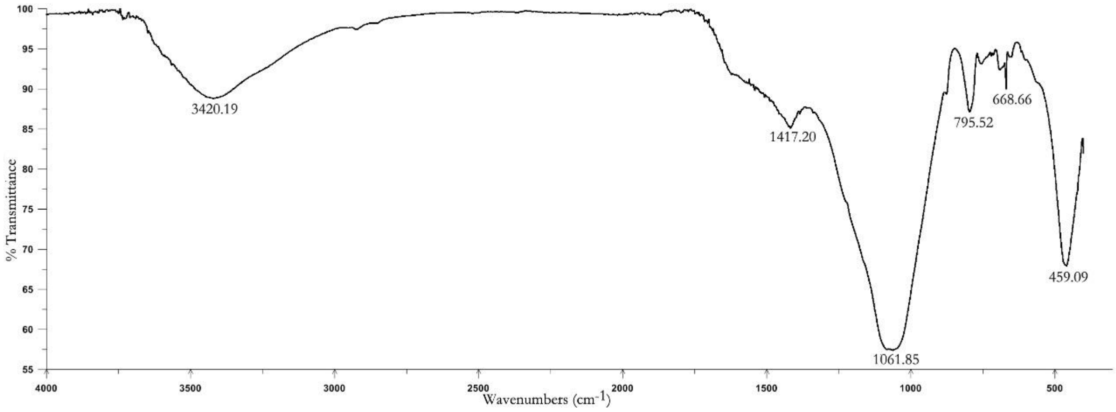



Fig. 7. Graph of functional analysis of Bagasse fly ash by FT-IR technique.

From Fig. 7, the functional groups of Bagasse fly ash were silicon dioxide bonds (Si-O-Si) at 1061cm$^{-1}$, 668 cm$^{-1}$ and 459 cm$^{-1}$, respectively, and the silanol bond (Si-OH) at 795 cm$^{-1}$. From the study of P. Woratanakul (2010) [24], the result showed that Bagasse fly ash has a high of silica content more than 90% which suitable for use as a source of silica in the synthesis of silica composition materials such as Zeolite. S. Chuntawitchaprapha (2011) [25] using the Zeolite synthesis from Bagasse fly ash which tested for heavy metal removal from electroplant wastewater the result shown that the heavy metal removal efficiencies of zinc, copper, chromium, cadmium and nickel were 85.25, 82.63, 79.71, 74.21 and 71.51 %, respectively at pH 5 by using time of 120 minutes for 1 grams of Zeolite.

DOI:10.4186/ej.2018.22.4.13

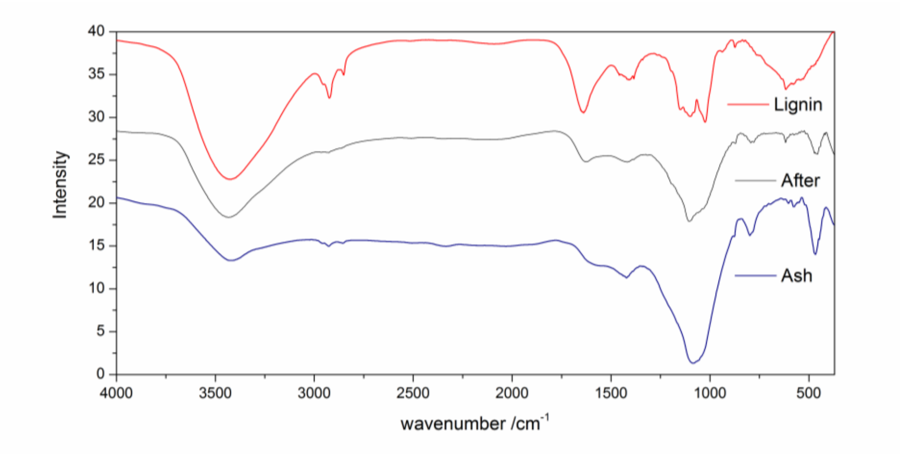



Fig. 8. The comparison of the functions of Bagasse fly ash, sludge from effluent (Lignin) and Bagasse fly ash after effluent adsorption using FT-IR.

From Fig. 8, the sludge from effluent reduces light intensity at typical wave lengths such as 1000-1500 $cm^{-1}$, 2750 - 3000 $cm^{-1}$ and 3250 - 3750 $cm^{-1}$. When adsorbed with Bagasse fly ash, the peaks range of the effluent tends to decrease, such as in the range 1500 - 1750 $cm^{-1}$. From the study of S. Loiha (2013) [26], the structure of Zeolite has any porous and large cavity which enough to the cation exchange and to permit the adsorption and desorption of organic molecules varying size up to 1 nanometer. The study results of W. Wirojanagud et al. (2008) [27] and N. Tantemsapya et al. (2004) [14] also showed that the Bagasse fly ash used in the synthesis of zeolite which has cation on the surface will attract the anion of lignin in effluent.

**Step 3. Color removal efficiency by filter**: The efficiency of color removal by sand filter when mixing Bagasse fly ash with the soil from the experiment field. The result showed that, the efficiency of color removal tended to increase with the maximum of 93 % and the minimum of 16% at ratio 1:7 and 1:0, respectively, as shown in Fig. 9.

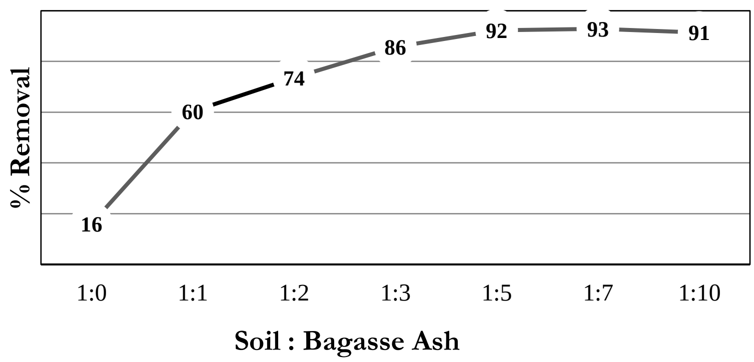



Fig. 9. The efficiency of color removal using the sand filter column.

From the study of I.A.W. Tan et al. (2007) [11] and R. Han et al. (2008) [12], it showed that the contact time increased as the adsorbent area increases, and consequently cause higher efficiency of color removal.

## 4. Conclusions

The pulp and paper mill effluent with high concentration of color (including colors from Lignin, Tannins, Humic, and Fulvic) caused the result of wastewater treatment process was still brown water color. In this study, the researchers used solid waste materials from the factories such as Bagasse fly ash, Lignite fly ash and Rice husk ash to determine the color removal efficiency of pulp and paper mill effluent. The results showed that Bagasse fly ash was the most effective to color removal in effluent with 94.2% removal efficiency which pH also constant to 7.5 - 8, although the ash content increased as much as 10 times. The analysis of functional groups using FT-IR found that the peaks range of the effluent which absorbed by Bagasse fly ash tended to decrease, wherewith Bagasse fly ash has cation on the surface that result in attraction the anion of lignin in the effluent. When mixing Bagasse fly ash with the soil samples from the experiment field and tested color removal efficiency by sand filter, it showed that the maximum color removal efficiency was 93% at the ratio 1:7 (Soil : Bagasse fly ash). As a consequence, not only the Bagasse fly ash was the best material for improving effluent, the color removal efficiency more than 90 % but also the pH of the effluent after using Bagasse fly ash was constant, which in the standard of effluent quality to be defined. Therefore the Bagasse fly ash can adapted to treat the effluent by mixing with soil for plantation in the treatment field of pulp and paper mill industry.

## Acknowledgements

The authors wish to thank SCG Phoenix Pulp and Paper Public Company Limited and Agricultural Machinery and Postharvest Technology Research Center KKU for funding support. Acknowledgements also for valuable data from the Institute of Water Resources and Environment, Thailand. Thanks to Department of Agricultural Engineering, Khon Kaen University for facilitating the study. Finally, special thanks of gratitude to Assistance Professor Dr. Vichian Premkamol for supports encouragement, and all the help.

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