A NOVEL TREATMENT OF DYE WASTEWATER OF BATIK INDUSTRY BY A COMBINATION OF ALUMINIUM SULFATE AND NATURAL IRON COATED-SAND

PENGOLAHAN LIMBAH CAIR DARI INDUSTRI BATIK DENGAN KOMBINASI ALUMINIUM SULFAT DAN PASIR BESI ALAMI

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Abstract
This paper presents the results of dye wastewater treatment of a batik industry in a batch experiment. The experiment was carried out by adding a mixture of aluminium sulfate as coagulant and natural iron oxide-coated sand (NICS) into the wastewater. Sedimentation rate of the formed flock was enhanced by a magnet. Initial COD concentration of the dye wastewater of 1060 mg/L was used in the experiment. Results of this research showed that the efficiency of the dye removal and settling time were affected by iron sand entrapped in the formed flocks. The highest efficiency of COD removal was 95%, or similar to the final COD concentration of 45 mg/L at settling time of 10 minutes. It could be concluded that the combination of aluminium sulfate and NICS could effectively be used in the treatment of the dye wastewater from batik industry.

Keywords: dye wastewater, coagulation, iron oxide-coated sand, batik industry

1. INTRODUCTION
Batik is an ancient method of textile decoration, which has been practiced in many places all over Asia since prehistoric times. The names of the tools and even the name "batik" were adopted from the Indonesian language. Batik means "drawing with wax" (Haake, 1989). In other reference, it is thought to be derived from the word "ambatik" which translated means "a cloth with little dots" (Living in Indonesia, 2010). According to the Ministry of Industry, the number of batik industry in Indonesia was almost 50,000 units and employed more than 800,000 people (Kementerian Perindustrian, 2010).
Batik manufacturing consumed a considerable amount of water in its manufacturing process. The water is primarily utilized in the dyeing and finishing operations. Dye-containing effluent is toxic to the environment since dyes are stable compounds, with low biodegradability and can be carcinogenic (Gregory, 1986). Dyes are normally very large aromatic molecules consisting of many linked rings. Three common dyes, classified according to the fibers to which they can be applied and their chemical nature are: (1) acid dyes; (2) reactive dyes; and (3) disperse dyes (Yang and McGarrah, 2005). Biological treatment processes are frequently used to treat dye wastewater effluents. These processes are generally efficient for biochemical oxygen demand and suspended solids removal, but they are largely ineffective for removing color which is visible even at low concentrations (Slokar and Majcen, 1997; Banat, 1996). Therefore, other techniques or their combination must be applied.

Including coagulation, flocculation, filtration and membrane adsorption, electro coagulation, are used to remove color from dye effluents (Tores et al., 2010; El-Gohary and Tawfik, 2009). Removal of colour from textile wastewater has been reviewed (Pearce et al., 2003). Coagulation/flocculation process is one of the most efficient methods that are widely employed for dye removal from industrial wastewater as it is efficient and simple to operate (Gao et al., 1997; Tan et al., 2000; and Jiang et al., 1996). However, for high loading of dye wastewater, these systems need a relative high sedimentation tank. This paper describes application of aluminium sulfate as coagulant and NICS for pre-treatment of a dye containing effluent from a batik industry. The main objective of using the NICS is to increase a sedimentation time of formed flocks. The iron sand entrapped in the flock can be quickly isolated by a magnet. Mechanism of dye removal by combination of coagulant and iron coated-sand is presented in Figure 1.

**Figure. 1.** Mechanism Dye Removal Using Combination of Coagulant and NICS

### 2. METHODS

**Dye Wastewater and Natural Iron-Coated Sand**

The experiment was performed at laboratory scale and batch test. The dye wastewater samples were collected from a batik industry at Banda Aceh City, Aceh Province. The key compositions of the wastewater are as follows: neutral pH and COD value of 1060 mg/L. A NICS (ferruginous sand) extracted from an iron ore located in District
of Aceh Besar, Aceh Province. The NICS sample and fraction of the iron grain are presented in Figure 2 and Table 1 respectively. Size of the iron sand is reduced about 250 µm prior to application.

**Dye Removal Tests**

Jar tests were carried out to evaluate the efficiency of aluminum sulfate and NICS to reduce dye contaminant in the wastewater from a batik industry. Three hundreds mL dye wastewater was filled into each of six beakers of 1 L volume. Aluminium sulfate solution of 3 mM was added to each beaker. This optimum concentration of coagulant dose was based on a previous test with the jar test on similar dye wastewater. The dye wastewater samples were first agitated vigorously at 200 rpm for 3 min and subsequently mixed at a reduced speed of 90 rpm for 15 min as well as adding the NICS. After coagulation, the samples were settled or magnated and then 50 mL aliquot was sampled from the clean top zone of the beaker for the measurement of residual COD. The treatment was replicated three times with similar condition.

**Analytical Methods**

COD was determined with a HACH DR-2000 model spectrophotometer. The NICS was characterized using SEM-EDAX (JEOL JSM-6390A). The scanning energy for EDAX analysis was performed from 0 to 21 keV with an elapsed time of 80 s. For the NICS investigation, two samples were prepared with pre-treatment by heat at 250°C for one day and without pre-treatment.

<table>
<thead>
<tr>
<th>Size (mm)</th>
<th>(%)</th>
</tr>
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<tbody>
<tr>
<td>≥ 1</td>
<td>0.87 ± 0.02</td>
</tr>
<tr>
<td>0.7 - 0.5</td>
<td>0.05 ± 0.01</td>
</tr>
<tr>
<td>0.5 - 0.4</td>
<td>2.82 ± 0.15</td>
</tr>
<tr>
<td>0.4 - 0.25</td>
<td>19.04 ± 0.22</td>
</tr>
<tr>
<td>&lt; 0.25</td>
<td>76.70 ± 2.30</td>
</tr>
</tbody>
</table>

**3. RESULTS AND DISCUSSION**

**Characterizations of the Natural Iron Oxide-Coated**

SEM photographs of the NICS were taken at 150x, 500x magnifications to observe its surface morphology. Representatives of the SEM photographs are presented in Figure 3a and Figure 3b for sand with and without heat pre-treatment, respectively. The NICS had a very rough and crack it could be found in the surface. The sand surfaces were also apparently occupied by mixture of cations.

Elemental compositions of the NICS were determined from EDAX spectra (Figure 4). The EDAX spectrum showed the presence of some mineral oxides which could be attribute of this NICS. As determined by EDAX, the predominant chemical compositions of the NICS are given in Table 2. These results showed that the predominant chemical compositions of this material included iron oxide, silica, magnesium, sodium, aluminium, titan, and carbon. The EDAX compositional analysis revealed that the surface morphology of the NICS was developed on O and Fe as a principal phase as evident the high intensity peaks of EDAX spectrum (Figure 4).
Process Performances
Table 3 demonstrates the sedimentation effect on the COD removal of the dye wastewater without treated by aluminium coagulant. As the contact time was increased until 24 h, the final COD concentration achieved 89 mg/L (±17 mg/L), which was equivalent to COD removal of 91.6%.

Figure 2. NICS Used in this Study

Figure 3. SEM Photographs of the NICS
In order to achieve Indonesian effluent standards of wastewater from textile industry (i.e. 150 mgCOD/L according to KepMen LH No. KEP-51/MENLH/10/1995), it needs about 2 h for sedimentation. Although it is a relatively very short settling time, this method is complicated to be applied in real situation. It is because the settled dye stuff is easy to re-dissolved in liquid media.

During treatment of the dye wastewater with aluminium sulfate,
the dye will accumulate in flock formation. Adding the particles of NICS into the inner part of the flock, it resulted in complete precipitation and settlement. From visual observation, the sand particle contained flock formation was not easy to redissolved. Table 4 shows that COD final concentration of dye wastewater at treatment with different NICS dosages. Similarly, the aluminium sulfate dosage was fixed at 3 mM. When the dosage of the NICS increased from 15 g/L to 90 g/L, the COD value in initial concentration of 1060 mg/L decreased in a range of 68 ± 10 mg/L to 45 ± 5 mg/L, at precipitation time of 10 min.

Table 3. Effect of Precipitation in COD Removal of the Dye Wastewater

<table>
<thead>
<tr>
<th>Duration of precipitation (h)</th>
<th>COD (mg/L)</th>
</tr>
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<tbody>
<tr>
<td>Start</td>
<td>1060 (initial concentration)</td>
</tr>
<tr>
<td>2</td>
<td>118 ± 9</td>
</tr>
<tr>
<td>8</td>
<td>108 ± 12</td>
</tr>
<tr>
<td>16</td>
<td>97 ± 5</td>
</tr>
<tr>
<td>24</td>
<td>89 ± 17</td>
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</tbody>
</table>

Table 4. COD Removal by Combination of Aluminium Sulfate and NICS

<table>
<thead>
<tr>
<th>Time of precipitation (min)</th>
<th>10</th>
<th>20</th>
<th>30</th>
<th>60</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. 0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>72 ± 12</td>
</tr>
<tr>
<td>2. 15</td>
<td>68 ± 10</td>
<td>62 ± 5</td>
<td>59 ± 10</td>
<td>50 ± 7</td>
</tr>
<tr>
<td>3. 30</td>
<td>60 ± 5</td>
<td>56 ± 9</td>
<td>54 ± 11</td>
<td>42 ± 5</td>
</tr>
<tr>
<td>4. 45</td>
<td>57 ± 7</td>
<td>50 ± 5</td>
<td>45 ± 9</td>
<td>39 ± 10</td>
</tr>
<tr>
<td>5. 60</td>
<td>53 ± 3</td>
<td>44 ± 9</td>
<td>40 ± 8</td>
<td>32 ± 5</td>
</tr>
<tr>
<td>6. 75</td>
<td>49 ± 9</td>
<td>39 ± 7</td>
<td>36 ± 11</td>
<td>27 ± 9</td>
</tr>
<tr>
<td>7. 90</td>
<td>45 ± 5</td>
<td>36 ± 7</td>
<td>31 ± 6</td>
<td>18 ± 6</td>
</tr>
</tbody>
</table>

These results were comparable to COD removal of 94 to 95%. For the control, the sample was only added aluminium sulfate without NICS, the COD was reduced to 72 ± 12 mg/L at settling time of 60 min. The lowest concentration of COD was found at the NICS of 90 g/L at 60 min of settling time. It could be concluded that combination of aluminium sulfate and NICS has significantly reduced the COD value of the dye wastewater. Figure 5 shows representative of dye removal by using combination of NICS and aluminium sulfate.

4. CONCLUSION

The optimum operational dose of the coagulant was 3 mM. In combination of aluminium sulfate and NICS, COD concentration could be effectively reduced to 45 mg/L within 10 minutes of settling time. This result was 20 times shorter than that of experiment without any adding coagulant and NICS. The COD removal achieved 95%, indicated that the combination of aluminium sulfate and NICS was effective for dye
wastewater treatment of batik industry. It might offer prospective application of reusing the treated water and will significantly contribute to reduce fresh-water consumption.

Figure 5. Batik's Wastewater Before (a) and After (b) Treatment by Combination of NICS and Aluminium Sulfate.

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REFERENCES


