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Pre Treatment of River Water By Using Bentonite and Modified Zeolite

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Abstract

River pollution has been a matter of public concern in the big cities. Therefore, various methods are needed in water treatment system to achieve a good quality of water supply. Coagulation-flocculation treatment method was essential of the applicable for water supply treatment. Coagulants were playing the main factors in this process. Environment friendly coagulants are important for replacing the chemical based coagulants that have been used in the past decades. This study focused the removal efficiency of synthesis zeolite and its combination as coagulants in river water treatment through coagulation-flocculation process. Experiment research was conducted to determine removal efficiency of turbidity, suspended solids, colour and iron concentration. Optimization of pH, dose ratio, dose and settling time were investigated for synthesis zeolites and its combinations. It was found that zeolites are able to remove 90% of turbidity, 81% of suspended solids, 80% of colour and 84% of iron (Fe^{2+}) of removal. This result indicates that zeolite may be a good adsorbent based on efficiency of turbidity removal which exceeds more than 80%. Meanwhile, the combination of zeolite-ferric chloride gave better result with 95% of turbidity, 88% of suspended solids, 75% of colour and 88% of iron removal efficiency compared to combination of zeolite-bentonite which the results showed slightly decreased. The combination of modified zeolite-ferric chloride and modified zeolite-bentonite was also investigated. However, the combination of modified zeolite-bentonite was proved that the removal efficiency obtained more than 80% of removal for all parameters. (94% of turdibity, 83% of suspended solids and 84% of iron removal). An experiment result indicates that zeolite synthesized can be a good environment friendly coagulant in river water treatment. Finally, the result of X-Ray Diffraction showed the mineralogy components in zeolite synthesized are higher than natural zeolite which will improve the adsorption capacity.

Keywords:river water treatment, coagulation-flocculation, bentonite, modified kaolin, zeolite, ferric chloride, X-Ray Difrraction.

Introduction

Many water treatment plants use a combination of coagulation, sedimentation, filtration and disinfection to provide clean, safe drinking water to the public. Worldwide, a combination of coagulation, sedimentation and filtration is the most widely applied water treatment technology and has been used since the early 20th century [1].

Coagulation is chemical technique directed towards destabilization of colloidal particles whereas flocculation, in engineering usage, is a slow mixing technique which promotes the agglomeration of destabilized particles [2].

Removal of specific contaminants in coagulation process may be affected by several factors such as temperature, pH, alkalinity and the choice of coagulant [2]. Coagulants that commercialized in the market are mostly chemical based which may create adverse impact to the environment. Thus, natural source coagulants for flocculation process in waster or water treatment such as bentonite and zeolite (from modified kaolin clay) are believed could be the alternatives for pre-treated river water compared the use of commercially coagulants which are not environmental friendly.

The use of bentonite as natural sources will give lots of benefits to improve the water treatment system in Malaysia. Bentonite is cheaper compared to the commercially prepared coagulants thatcurrently used. Further, the composition of bentonite and zeolite will affect of its usage. The effectiveness of zeolites and bentonite are the most importance factor for this study.

In 2008, the number of river basins monitored remained at 143 and the number of monitoring stations were 1,063. Out of these 1,063 monitoring stations, 612 (58%) were found to be clean, 412 (38%) slightly polluted and 39 (4%) polluted. There was a significantly reduction in the number of clear river basins in 2008 compared with 2007. There were 76 (53%) clean river as compared with 91 in 2007. However, the number of polluted river basins remained at 7 [3].

Materials & Methods

The materials used in this investigation are ordinary kaolin, bentonite, rice husk ash (RHA) with a maximum size of 63µm. Kaolinite is one of the most versatile industrial clay minerals by virtue of its chemical inertness between pH 4 and 9 [4]. Bentonite sample was brought from Ipoh Ceramic SdnBhd in Ipoh, Perak in powder form. Then, the bentonite sample was sieved into 63µm. [5]. Thebentonite sample was kept in plastic box and ready to be used. Among the various biomasses with abundant and renewable energy sources, rice husk (RH) is not only a potential source of energy, but also a value added by product. [6]

The main elemental components of RH were C 37.05wt%, H 8.80 wt %, N 11.06 wt %, Si 9.01 wt % and O 35.03 wt %. [7]. The contents of RH are xylan (main components of hemicellulose). The employment of these fractions as resources for producing large scale commodities such as xylose, activated carbon and silicon dioxide can be a good choice.

River Water Sampling

The river water for this study was taken from the Sg. Muda,Penang. The river water sampling procedures done by grab sampling method. Grab sampling method is the process of all the test material is collected at one time. As such, a grab sample reflects performance only at the point in time that the sample was collected.

River Water Characterization

pH is the measures of the alkalinity or acidity concentration in water. Throughout this experiment, the pH adjustment was tested by using pH meter or by means of titration. The acid hydrochloric and sodium hydroxide were used for the adjustment.

The turbidity of SgMuda River water was measured using portable Turbiditymeter (HACH 2100N). The measurement of turbidity is a key test of water quality. The turbiditymeter measures the light transmittance of a sample in NTU's. A 20 ml of sample was analyzed by using turbiditymeter and the result appeared on the digital screen after few seconds.

In this study, the TSS is being measured by using HACH DR 2800 Spectrophotometer. A 10 ml sample was analyzed. The blank sample cell assembly was wiped and then placed it into holder cell of the spectrophotometer for zeroing the instruments. The result will appear on the screen in a few seconds in the unit of mg/L. There are two methods to determine metals which are wet chemistry method and equipment method. For this research, the equipment method chose. The DR 2500 Odyssey Spectrophotometer with the HACH wavelength of 255 (Fe) Ferro Ver (Method 8146) is used to analyze metals like Fe in a sample.

A 25ml of sample was placed into clean sample cell. Then, the Ferro Ver Iron Reagent Powder Pillow was added and mixed into the sample cell. An orange colour is present. Next, the sample reaction will occur within three minutes time. The results were measured in mg/L of Fe.

Synthesis of Zeolite From Kaolin

A method for preparing a modified kaolin clay (zeolites), includes the steps of providing a kaolin clay material having a starting ratio of silica to alumina, mixing the kaolin clay material with acid so as to form a substantially homogeneuos paste. Calcining the paste as to provide a calcined acid treated clay material. Mixing the calcined acid treated clay material in water to form suspension, precipitating aluminium from the suspension to produce a reduced aluminium suspension.

Furthermore, in obtaining a modified clay materials (zeolites) from the reduced aluminium suspension, where in the modified clay material has a final ratio of silica to alumina which is higher than the starting ratio.

Iron content of kaolin was removed by digestion with 6 M HCl, washed with distilled water and dried overnight in order to remove Fe and decolourise the raw material. Thermally activated sample was prepared by calcining clays for 1 hour at 950 ºC in the tube furnace to change kaolin to metakaolin (amorphous solid). Then, it was crushed and sieved to 1.18 mm – 2.00 mm in size. The gel was prepared by dissolving the metakaolin in NaOH solutions. 50 g of metakaolin was stirred at 250 rpm with 1000 mL of NaOH at the temperature of 100 °C for a period of time. The concentrations of NaOH are 2 mol L-1 and the reaction time was 4 hours [8].

The synthesis zeolites are done by followed this procedure. Kaolinite reactivity can be increased by altering its properties with thermal, mechanical or chemical treatment. Chemical (acid or alkaline) reactions are usually of little use, even under drastic conditions, owing to the high inertness of kaolinite. On the other hand, calcinations above 550 °C convert kaolin into metakaolin by removal of structural OH groups, rearrangement of Si and Al ions and formation of penta and tetracoordinated Al ions at the expense of hexacoordinated Al ions. Above 900 °C, metakaolin recrystallizes into γ-Al_2O_3 [7]. Metakaolin has for a long time been used to produce zeolites and pozzolans. [9] found mine kaolin and washed, mechanically modified samples obtained from it to have pozzolanic value. Finally, the zeolites are modified by added 10% of rice husk ash (RHA) and dried at 120^o C overnight in oven.

Furthermore, in this study the synthesis zeolite was treated by acetic acid to improve the adsorption capacity as polymers in this experiment. In this study 20% of acetic acid was used for treated of synthesized zeolite. According to Gang et al. (2003) acetic acid was one of the most important intermediates in chemical industry. Certain naturally occurring hydrated metal aluminium silicates are called zeolites. The synthetic adsorbents of the invention have compositions similar to some of the natural zeolites. The most common of these zeolites are sodium zeolites. Zeolites are useful as detergent builders, cracking catalyst and molecular sieves.

Zeolite consists basically of a three-dimensional framework of SiO_4 and AlO_4 tetrahedral. The electrovalence of each tetrahedral containing aluminum is balanced by the sodium ion. This balance may be expressed by the formula Al/Na=1. The spaces between the tetrahedral are occupied by water molecules prior to dehydration. In this study, the zeolites are modified by addedd 10% of rice husk ash (RHA) and dried at 120^o C overnight in oven.

Jar Test

Jar testing is a pilot scale test of the treatment chemicals used in a particular water plant. It stimulates coagulation-flocculation process in a water treatment plant. The Jar Test Model JLT 6 is selected to run this method. By using using the 1000 mL graduated cylinder, the 500ml of river water was added. Then, coagulant (bentonite and their combination) is added into the beaker to be coagulated to each of the jar test beakers. However, the pH adjustment was done before this process.

After dosing each beaker, operate the stirrer at 80 rpm (rapid mixing) for approximately 10 minutes. This simulates the static mixer. Then, slow to the 50 rpm which matches the turbulence created in your flocculators and allow stirring for 15 minutes while observing the floc formation. At the end of the 15 minutes turn the stirrer off and allow settling.

Optimum Dose Mixture

To obtain the optimum mixture ratio of bentonite and theirs combination is the aim of this experiment for removal of selected parameters. Therefore, the media ratio used based on volume per volume percentage were presented in ratio of 10:90, 20:80, 30:70, 40:60, 50:50, 60:40, 70:30, 80:20 and 90:10. The volume used in this ratio was obtained from optimum dosage for each adsorbent. The removal percentage was determined by using equation 1.

$$\text{Removal (\%)} = \left[\frac{(Ci - Cf)}{Ci}\right]100 \qquad (1)$$

Where, C_i = Initial concentration of groundwater

C_f= Final concentration of groundwater after the adsorption process

X- Ray Diffraction

The composition of synthesis zeolite that used for adsorption is determined by using X-Ray Diffraction (XRD). There are about 95% of all solids materials can be described as crystalline. [10].

The X-ray diffraction pattern of a pure substance is like a fingerprint of the substance. The powder diffraction method is ideally for characterization and identification of polycrystalline phase. [11] The main use of powder diffraction is to identify components in a sample by a search and match procedure. Furthermore, the areas under the peak are related to the amount of each phase present in the sample.

Results and Discussion

Sungai Muda river water are collected at Rumah Pam, Bumbong Lima, Penang between January 2011 and February 2011. The several selected parameters such as turbidity, suspended solids, colour, ferrous iron and as well as pH value. The initial value is 36 NTU, 17.3 mg/L, 151 unit points at 465nm (True colour), 1.34 mg/L Fe and 7.41 for pH. Based on INWQS, this water sample fall in Class IIA where the conventional treatment needed for water supply.

Zeolite As Adsorbent

Adsorption is the most preferable process for water treatment technology. The numerous adsorbents such as granular activated carbon, polymeric resin and high silica zeolites have been used to treat contaminated groundwater and drinking water [12].

In this study zeolites had been tested on the optimum dosage for efficiency of turbidity removal. The result from figure 4.1 indicates that by increasing the zeolites dose, the removal efficiency was also increased respectively. The optimum dose was recorded at 600mg/L with 96% of removal efficiency. While, the 1000mg/L and 1200mg/L showed the small decreasing of removal efficiency with 91% and 88% for turbidity removal. Even though, there was a slightly decreased of removal efficiency, but the rate of removal was still considered good.

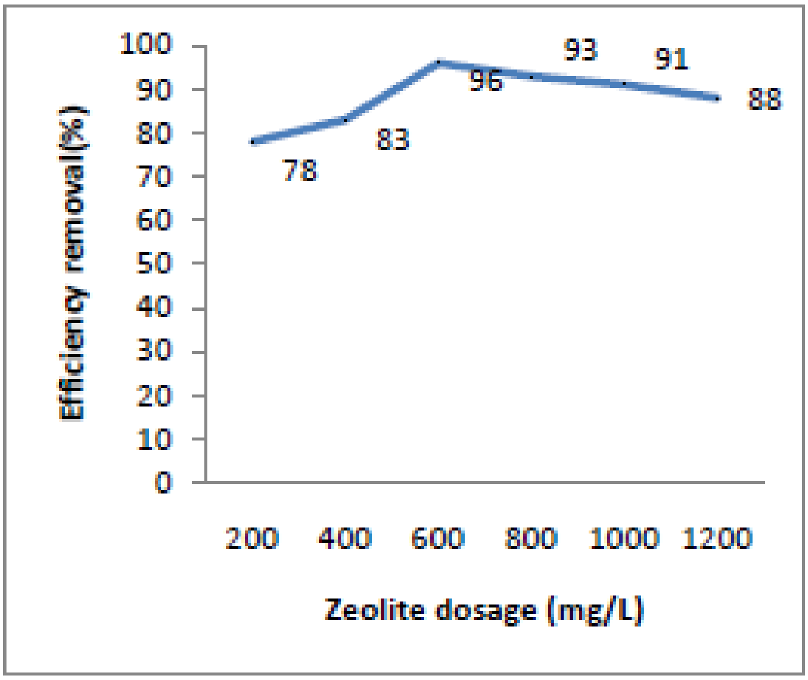

Figure 4.1: The optimum dose for zeolite.

The initial pH of the sample was 7.41. After the addition of zeolite dosages at a concentration of 600mg/L, the pH increased (2 to 6). The optimum rate of pH occurs where the removal efficiency is in the range of 88-86% of removal and pH value from 6 to 8. In his previous study, [13] was identified the extend of pH value not only depends on the types and concentration of coagulant but depends on the characteristics of water itself. In acidic conditions, the amount of cationic was increased therefore resulting high activity since charge neutralization effect was maximized.

The results on the removal of turbidity, suspended solids, colour and ferrous iron was shown in figure 4.2 below. By using 600mg/L concentration of zeolite, the efficiency of removal was in the range of 70-90%. Turdidity, suspended solids, colour and ferrous iron removal was recorded with 95.3%, 81%,80% and 75% respectively. Its indicate that, zeolite was an effective coagulants in this study. The removal efficiency of zeolites was being found to be an effective adsorbent based on several studies done.

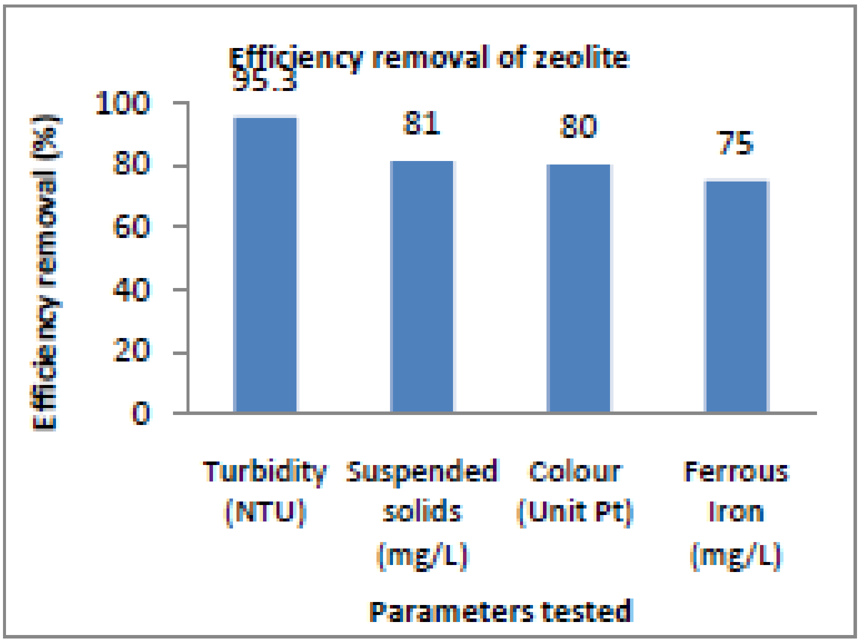

Figure 4.2: Efficiency rmoval of zeolite

Bentonite Mixed Zeolite

The smaller particles were more effective than larger particles because of the greater surface area available and increasing the adsorption capacity. [13]. Therefore, in this experiment, 63µm size was chose for this mixture to ensure the higher adsorption process occurs in the coagulation.

Figure 4.3 shows the results of removal efficiencies by applications of dose optimum (800mg/L) as well as optimum dose ratio (60:40), optimum pH (6-8) and optimum settling time (60 minutes) for this mixture.

Based on the three parameters tested, the graph showed the result of removal efficiency was range from 80-100%. Turbidity removal efficiency was stated at 93.4 %, followed by colour (86%) and suspended solids (83%). It indicates that, this combination as a good adsorbent for turbidity, colour and also suspended solids removal because the efficiency was achieved more than 70% of removal. This behaviour is compatible with what have been reported in the study of adsorption regarding the effects of bentonite concentration which have been done by [14].

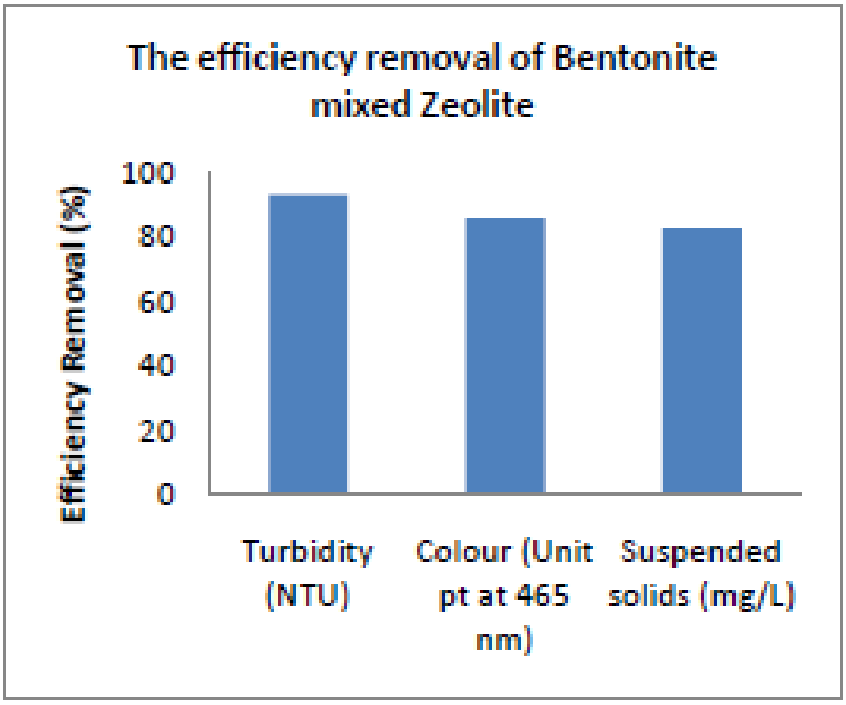

Figure 4.3: Removal efficiency of bentonite mixed zeolite.

Ferric Chloride Mixed Zeolite

The researcher, [15] found that coagulation –flocculation process using$FeCl_3$ can be used effectively for water and wastewater treatment. The removal efficiencies of COD (99%) and turbidity (88%) were obtained respectively. Therefore, coagulation – flocculation is a commonly used process in water and wastewater treatment in which compounds such as ferric chloride or polymers are added to in order to destabilize the colloidal materials and cause the small particles to agglomerate into larger steerableflocs. $FeCl_3$ is an important coagulant in water and wastewater treatment and can be used for colour removal and treatment of municipal wastewater [16].

According to the figure 4.4, the turbidity removal was stated at 95% of efficiency then followed by the colour removal, suspended solids and ferrous iron with 76%, 88% and 88% respectively. It was concluded that the efficiencies removal of selected parameters are ranged from 70-100%.

The highest turbidity removal efficiencies by using zeolites and its combination had proved that clay minerals may be used as coagulant aids in flocculation step of binding already formed small flocs into larger particles when aluminium or ferric chloride have been used as the primary coagulant.[17]

Furthermore, a study done by [18] reported that when bentonite, alum and ferric chloride were used as coagulant, the turbidity removal resulted in 93.4%, 77% and 95% respectively. This results show that ferric chloride can be noteworthy option and provide higher removal efficiencies than others combination.

While for suspended solids and ferrous iron removal are consider as a good performance for this combination. The results are still under Interim Water Quality Standards for Malaysia (2010)which is 50mg/L and 0.3mg/L in Class IIA/B.

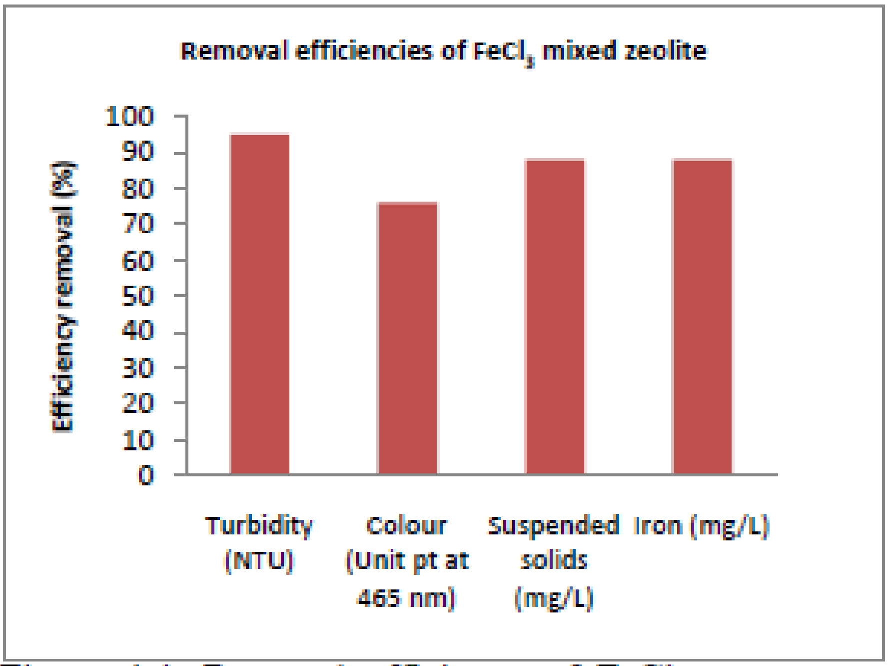

Figure 4.4: Removal efficiency of $FeCl_3$ mixed zeolite

Bentonite Mixed Modified Zeolite

This combination was done by using the optimum conditions for dosage, dose ratio, pH and settling time based on bentonite mixed zeolite. The objective was to determine the removal efficiencies of modified zeolite as coagulants.

Bentonite and modified zeolite (MZ) combination have been tested on three selected parameters. The results obtained from the experiment proved that efficiencies removal are improved slightly compared to previous combination performance with the range 80-100%.

Figure 4.5 explained the removal efficiency for turbidity, colour and suspended solids were 85%, 83-84% and 82-83% respectively. [19] the RHA is found to consist of mesopores predominantly. This is what desirable for the liquid phase adsorptive removal of metal ions. The 99% are attributed to mesopores during desorption indicates the predominance of mesopores in adsorption process.

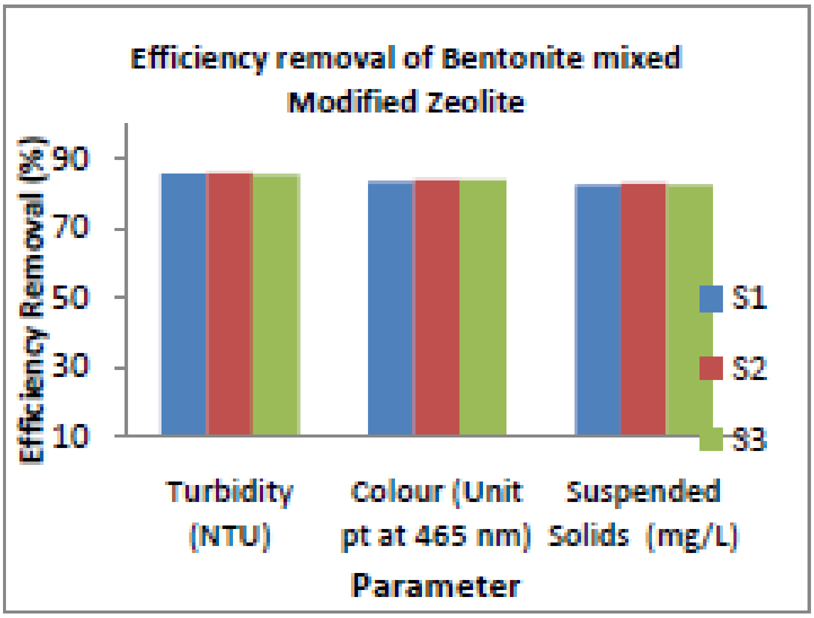

Figure 4.5: Removal efficiency of bentonite mixed modified zeolite

Ferric Chloride Mixed Modified Zeolite

Effects of this combination on the turbidity removal efficiency are shown in figure 4.6. Coagulation-flocculation was performed between 200mg/L with 20:80 ratio of combination. The highest removal turbidity efficiencies were recorded at 68%. This showed that the result was slightly decreased compare to the previous combination($FeCl_3$ mixed with zeolite). While for suspended solids and ferrous iron removal efficiencies also decreased.However, this condition was still comply with the INQWS, 2010 which all the value is still under the range stated.

Comparisons of Different Combinations of Zeolite

This study has been completed of four combinations coagulants. There are bentonite mixed zeolite, bentonite mixed modified zeolite, ferric chloride mixed zeolite and ferric chloride mixed modified zeolite. All these combination used for tested on efficiencies removal of turbidity, colour, suspended solids and ferrous iron.

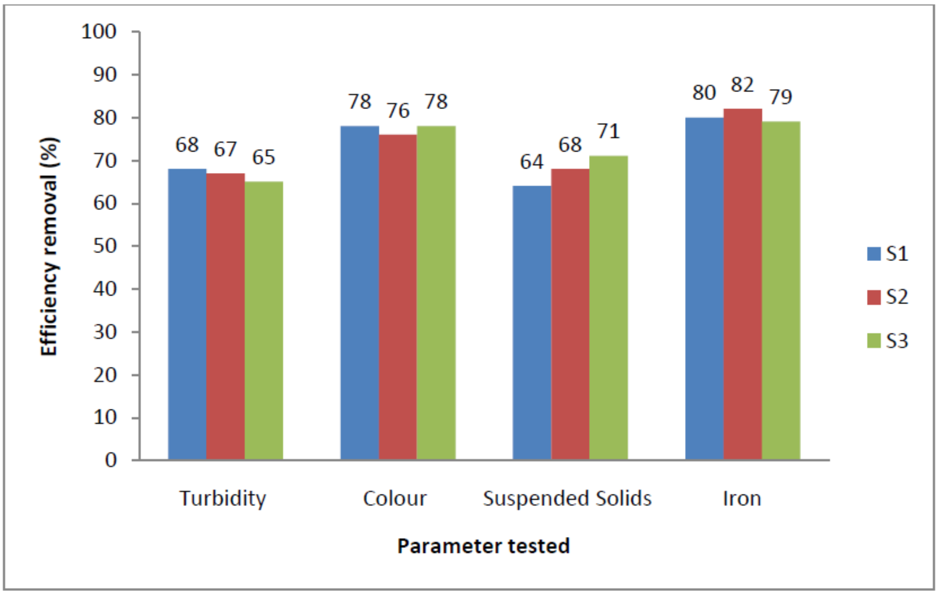

Figure 4.6: Removal efficiency of $FeCl_3$ with modified zeolite

The result of different combinations of coagulants from experiment was showed that combination of zeolite-FeCl3 is the highest removal efficiency for turbidity compared to other combination. These results have been agreed by [18] alum and ferric chloride were used as coagulant, the removal efficiency of ferric chloride was the highest.This results shows that ferric chloride can be noteworthy and provide higher removal efficiencies.

Next, the suspended solids efficiencies removal was also stated that the combination of zeolite-ferric chloride was the greatest among the others with 88% of removal. In other hand, the removal efficiency was decreased for the combination of modified zeolite-ferric chloride with only 68% of removal. The others combinations is considered in a good efficiencies removal rate. In most rivers, total suspended solids are primarily composed of small particles. TSS is often referred as turbidity and it's frequently measured. Higher TSS (> 1000 mg/L) may greatly affect water use by limiting light penetration and can limit the reservoir life through sedimentation of suspended matter.

Furthermore, the highest removal efficiency of colour in Sungai Muda River was recorded at 83% of removal forzeolite-bentonite combinations. Zeolites offer an attractive and inexpensive option for removal of both organic and inorganic contaminants. [4].

The further study was done by using modified zeolite as a coagulant. Zeolite modification with 10% RHA has proved to be a good coagulant in coagulation-flocculation. Therefore, the efficiency of turbidity removal was increased with 94% of removal which is higher than using a natural zeolite. Thus, RHA was found to have a wide pore size distribution which giving wide distributionof surface area. [18] had an agreement based on his previous study. He also admits that this structure indicates the predominance in adsorption process.

However, for combination of modified zeolite-ferric chloride was showed the decreasing of the efficiencies removal for turbidity, colour and suspended solids. In other hand, the removal efficiency for iron was increased compared with a single zeolite coagulant.

X- Ray Difraction

Based on this study, the kaolinite ($Al_2Si_2O_5(OH)_4$) experienced chemical transformation by dehydroxylation between 450^oC and 600^oC, which results in the formation of metakaolinite ($Al_2Si_2O_7$). The decomposition of metakaolinite at a temperature between 800^o C and 950^oC produces an armorphous material from which mullite crystallized, that is consistent with the result obtained from XRD.

The quantitative analysis according to the X-Ray Diffraction showed that the synthesized and modified zeolite used in this study was mainly composed of clinoptilolite mineral (63 %) of Na, K and Ca and the others as shown in table 4.2 below. The result proved that the main elements such as SiO_2, Al_2O_3, and TiO_2 in synthesized zeolite were increased compared to the natural zeolite composition.Therefore, the synthesized and modified zeolite were used in the experiment.

Table 4.2: The mineralogy composition of synthesized and modified zeolites

Elements	Percentage (%) synthesized	Percentage (%) modified
SiO_2	78	84
Al_2O_3	13.2	13
TiO_2	0.7	0.6
Fe_2O_3	1.4	2.7
MgO	0.5	0.8
CaO	2.2	3.0
Na_2O	1.2	1.0
K_2O	0.7	2.5
ZnO	-	6.6

In the XRD patterns the thermally and mechanically modified zeolite samples showed the typical mineralogical composition. The main elements of zeolite, Si_2O increased more higher than natural and synthesized zeolite with 84% of composition. Then, the others elements such as Al_2O_3, Fe_2O_3, CaO, K_2O and ZnO also showed the increased percentage of composition as presented in table 4.2 above.

Based on the result, it was found that the starting materials consisted primarily of kaolin and a little amorphous component but the crystalline of ZnO-PO_4^{3-} modified product was relatively high and some new lines observed that can be attributed to ZnO-PO_4^{3-} modified structure formed respect to the original kaolin. In addition, the RHA mineralogy compositions itself play an important role in this transformation.

Conclusion

Result presented show that zeolite synthesized and its combination indicates as a good environment friendly adsorbent through the coagulation-flocculation process for river water treatment. The optimization recorded, pH range were identified from 6-8 with the 200mg/L and 800mg/L doses followed by 80:20 and 60:40 for zeolite-ferric chloride combination and zeolite bentonite combination respectively. This ratio increased with increased coagulation addition and with increasing the removal efficiency. This trend points to the likely feasibility of removal efficiency of employing higher coagulant addition for more efficient removal.

Next, based on removal efficiency of zeolites resulted with 90% of turbidity, 81% of suspended solids, 80% of colourand 84% of ferrous iron of removal. This result indicates zeolite may be a good adsorbent based on efficiency of turbidity removal ability which exceeds more than 80% of removal. In addition, the combination of zeolite-ferric chloride was proved to be as a good coagulant with 95% of turbidity, 88% of suspended solids, 75% of colour and 88% of iron removal compared to combination of zeolite-bentonite which the results showed slightly decreased. The study also found, the modified zeolite combination with ferric chloride as well as bentonite gave good results in removal efficiency percentage. The removal efficiency was achieved more than 80% of removal.

Apart from this study, the one hour (60 minutes) settling time was determined for both types of combinations. In general, raising the pH from the very acidic levels which resulted from the coagulant addition near to the neutral levels had a strong positive effect on removal of colour and turbidity. Similar effects have been previously noted in water purification studies. Similarly, a sharp reduction in colour and turbidity was observed at pH 6-8 following ferric chloride precipitation. In the study, the efficient removal of colour observed at all levels of coagulant dose and the identification of a single variable which is most important for colour removal. Therefore, removal of up to 80% of colour was effected through precipitation using ferric chloride. A maximum colour removal was observed only at 76% with increased addition of this coagulant dose resulted in significantly higher colour levels in the supernatant. This was possibly due to the coagulant dose coagulant demand with the excess ferric chloride remaining in solution, thereby contributing to colour.

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