Industrial Crops and Products 56 (2014) 17–26

Contents lists available at ScienceDirect

Industrial Crops and Products

journal homepage: www.elsevier.com/locate/indcrop

Optimization of agro-industrial wastewater treatment using unmodified rice starch as a natural coagulant

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ARTICLE INFO

Article history:
Received 9 September 2013
Received in revised form 20 January 2014
Accepted 16 February 2014
Available online 16 March 2014

Keywords:
Alum
Coagulation-flocculation
Palm oil mill effluent (POME)
Response surface methodology (RSM)
Rice starch
Sludge

ABSTRACT

There have been increasing interests on the use of organic starch derivative coagulants such as cationic starch in coagulation-flocculation process due to environment and health concerns regarding the use of inorganic coagulants. However, studies on the potential utilization of unmodified starch in wastewater treatment remain limited. In this study, treatment performance of unmodified rice starch and alum was tested on agro-industrial wastewater produced from crude palm oil extraction, namely palm oil mill effluent (POME). Response surface methodology (RSM) showed that an addition of unmodified rice starch not only enhanced total suspended solids (TSS) and chemical oxygen demand (COD) removals; but it also significantly improved the process by reducing both the dosage of alum (−47.95%) and settling time (−58.66%), while shifting the operating pH closer to the natural pH of POME. The treatment enabled TSS and COD removals up to 86.65 and 49.23%, respectively under the optimum conditions of 0.38 g/L alum, 0.28 g/L unmodified rice starch, pH 4.45 and settling time of 5.54 min. An addition of unmodified rice starch to alum also resulted in 33.8% reduction of chemical cost as compared to the treatment using alum alone.

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1. Introduction

Over the years, agro-industries have been one of the major contributors in the world economy especially for developing nations which often rely on subsistence farming and income generation from formal and/or informal agro-industries. At the same time, this industry is also one of the main contributors to worldwide industrial pollution problem. With such a drastic rate of population growth, increasing in per-capita energy consumption and the over-exploitation of natural resources, a rise of highly complex wastes generated from agro-industries is often inevitable. Although the composition and quantity of agro-industrial waste depend on the source of raw materials, nature of the products, operations and processing steps, it generally contains large amount of organics materials with high values of chemical oxygen demand (COD) and total suspended solids (TSS) which could potentially cause severe pollution problems (Prasertsan et al., 2007). If the untreated waste is released into the environments, it is certain to cause significant environmental problems due to its accumulation in soil and water environments (Wu et al., 2013a).

Wet process of palm oil milling has been the most common method to extract crude palm oil for large scale productions. However, some of the processes such as sterilization of fresh oil palm fruit bunches, palm oil clarification and hydrocyclone operations generate a thick colloidal mixture of agro-industrial wastewater known as palm oil mill effluent (POME) (Wu et al., 2010). Crude palm oil extraction produces approximately 50 million tonnes of POME annually in Malaysia (Wu et al., 2013b) and this figure is expected to rise in the future. Improper discharge of this highly polluting wastewater is causing environmental problems due to its high polluting characteristics with considerable amount of COD (53,630 mg/L) and TSS (19,020 mg/L) (Wu et al., 2009a). Ponding system is the most typical method used in POME treatment but it has many disadvantages such as long retention time, large treatment area, accumulations of large amounts of sludge and uncontrollable release of methane gas into the atmosphere (Wu et al., 2010; Lim et al., 2014). In many countries, waste management systems are undergoing changes due to the threat of global climate change and other environmental issues (Nouri et al., 2012). Hence, continuous studies on other treatment processes will provide invaluable insights towards improving the current treatment of POME (Wu et al., 2010).

As one of the most important processes to treat both urban and industrial wastewater, coagulation-flocculation could be a

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http://dx.doi.org/10.1016/j.indcrop.2014.02.018
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potential alternative for POME treatment. However, there are concerns regarding the use of inorganic metal salts (e.g. alum) in coagulation-flocculation process of wastewater. Besides producing large volumes of sludge, overdosing of these chemicals increases metal ions content in coagulated water and can potentially cause health hazards (Renault et al., 2009). Increasing awareness of this issue shifted interest towards using natural plant-based materials such as chitosan (Ahmad et al., 2006), *Moringa oleifera* seeds (Bhatia et al., 2007), guar gum (Pritchard et al., 2009), tannin (Beltrán-Heredia et al., 2010), *Jatropha curcas* seeds (Abidin et al., 2013), mustard seeds (Bodlund et al., 2013) and other common vegetables and legumes (Choy et al., 2013) in coagulation-flocculation treatment of raw water and wastewater.

Starch, one of the most abundant natural polymers in the world, is an interesting material to be used as a coagulant. According to Kraak (1992), starch products have a special role in purification process for drinking water through flocculation. In its crude form, it consists of a mixture of two polymers of anhydroglucose units, amylose and amylopectin (Wei et al., 2008). Due to its renewability, biodegradability and low cost, starch is often modified chemically or biochemically to produce coagulant. Currently, cationic starch is one of the most commonly investigated starch derivative coagulants to be used to effectively treat organic and inorganic matters in wastewater carrying negative charge (Wei et al., 2008). However, potentially hazardous chemicals such as formaldehyde, caustic soda and various solvents are often used for starch modifications.

Until now, study on the optimized use of unmodified starch as a natural coagulant still remains scarce. Selective flocculation of dark-colored minerals (DCM) using starch by Dogu and Arol (2004) was among the few studies done in the previous years. Due to the elimination of chemical modification steps, the use of unmodified starch to treat real industrial wastewater could be a better option as compared to its modified counterparts, either as a primary coagulant or coagulant aid to reduce the risk caused by the use of metal coagulants. Thus, the main aim of this work was to improve and optimize the overall efficiency and process conditions in coagulation-flocculation treatment of POME using (1) alum, (2) unmodified rice starch and (3) combination between alum and unmodified rice starch. Accurate evaluations on the treatment performances were done by optimizing coagulant/coagulant aid dosage, initial pH and settling time for each treatment through response surface methodology (RSM). This study would provide a better understanding on the feasibility of using unmodified rice starch in agro-industrial wastewater treatment as well as the potential to combine and reduce the amount of alum during the treatment of POME.

2. Materials and methods

2.1. Chemicals, collection of POME and procedures

Aluminum sulfate octadecahydrate ($Al_2(SO_4)_3 \cdot 18H_2O$) and rice starch were purchased from Sigma–Aldrich Corporation. Both analytical-reagent-grade chemicals were used without further purification. Rice starch solution of 30 g/L was prepared by dissolving the starch in distilled water. The mixture was then autoclaved using a pressure steam sterilizer (Electric Model No.50X, All American) at 121 °C and 17 psi for 20 min. The solution was then kept at constant temperature of 80 °C with agitation speed of 400 rpm using a hot plate stirrer. The rice starch solution was prepared daily to eliminate inconsistency of results due to biodegradation. Raw POME was obtained from Seri Ulu Langat Palm Oil Mill Sdn. Bhd., Malaysia and was preserved at 4 °C to prevent biodegradation. The initial TSS and COD of raw POME were 25,750 and 65,667 mg/L, respectively.

Table 1
Experimental range and levels of independent variables for treatment using alum or unmodified rice starch alone.

Variables	Symbols		Coded levels				
	Actual	Coded	−1.682	−1	0	1	1.682
Alum alone							
Alum dosage (g/L)	X_1	x_1	0.16	0.30	0.50	0.70	0.84
Initial pH	X_2	x_2	1.14	2.50	4.50	6.50	7.86
Settling time (min)	X_3	x_3	1.59	5.00	10.00	15.00	18.41
Unmodified rice starch alone							
Unmodified rice starch dosage (g/L)	Y_1	y_1	0.00	0.20	0.50	0.80	1.00
Initial pH	Y_2	y_2	1.48	2.50	4.00	5.50	6.52
Settling time (min)	Y_3	y_3	0.31	1.50	3.25	5.00	6.19

Coagulation-flocculation experiments were carried out using a flocculator (VELP Scientifica Flocculator JLT4) in 500 mL beakers. pH of the POME (300 mL) was firstly adjusted using 1 mol/L HCl or NaOH. The beaker was then placed in the flocculator and different dosage of coagulant (alum or unmodified rice starch) was added. The sample was then stirred rapidly at 150 rpm for 5 min, followed by slow stirring at 40 rpm for 15 min. For combination use of coagulant and coagulant aid, specific dosages of alum and unmodified rice starch were added at the beginning of rapid and slow stirring, respectively. The flocs were then allowed to settle for a specific duration of time. Analysis samples were taken from approximately 2 cm below the water level for TSS and COD analysis.

TSS was measured using photometric method (HACH Method 8006), while COD was determined by using United States Environmental Protection Agency (USEPA) reactor digestion method (HACH Method 8000). The removal efficiency from the treatment process was calculated by using equation (1) as stated below:

$$\text{Removal efficiency}(\%) = \left(1 - \frac{C_{\text{final}}}{C_{\text{initial}}}\right) \times 100\% \quad (1)$$

where C_{initial} and C_{final} are the concentration of TSS or COD in POME before and after the treatment, respectively.

2.2. Experimental design

Traditionally, 'one-factor-at-a-time' technique was used to optimize the treatment process, in which one parameter would be varied while the others were kept at constant level. However, this technique not only fails to identify possible interactions between variables, but is also time-consuming and expensive because large number of experiments must be carried out (Wu et al., 2009b).

To overcome these drawbacks, RSM was used in this study for analyzing and modeling the effects of multiple variables and their interactions using a limited number of experiments. Recently, RSM is used widely in waste treatment and management (Jadhav et al., 2013; Sánchez-Martín et al., 2013; Usharani and Muthukumar, 2013). Central Composite Design (CCD) was selected in this study to optimize the variables which were affecting the treatment efficiency. Independent factors chosen for the optimization process were alum dosage or/and unmodified rice starch dosage, initial pH and settling time. The range and levels of the chosen variables were shown in Tables 1 and 2. Both TSS and COD removals were the dependent variables (responses) investigated in this study. Using Design Expert (Version 6.0.10, Stat-Ease Inc., Minneapolis, USA) software, all responses were fitted using a predictive polynomial quadratic equation to obtain a correlation of both the response and independent variables. General form of second-order polynomial

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