Ecological Engineering 94 (2016) 352–364

Contents lists available at ScienceDirect

Ecological Engineering

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

Performance of conventional starches as natural coagulants for turbidity removal

Sook Yan Choy[a], Krishnamurthy Nagendra Prasad[a], Ta Yeong Wu[a,c], Mavinakere Eshwaraiah Raghunandan[b,c], Ramakrishnan Nagasundara Ramanan[a,c,*]

[a] Chemical Engineering Discipline, School of Engineering, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia
[b] Civil Engineering Discipline, School of Engineering, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia
[c] Advanced Engineering Platform, School of Engineering, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia

ARTICLE INFO

Article history:
Received 5 July 2015
Received in revised form 25 May 2016
Accepted 28 May 2016
Available online 20 June 2016

Keywords:
Coagulation-flocculation
Alum
PACl
Natural coagulant
Starch
Floc characteristics

ABSTRACT

The efficiency of rice, wheat, corn and potato starches in removing turbidity was performed and compared to alum and polyaluminium chloride. Using kaolin suspensions, the effects of pH, dosage and need for starch gelatinization was studied. Autoclaved rice starch with larger polymer chain length outperformed other starches to yield 50% turbidity removal with an optimized dosage of 120 mg/L and initial pH of 4 at a sedimentation time of 30 min. The integrated use of autoclaved rice starch and chemical coagulants as a two step coagulation process has further increased the efficiency of turbidity removal by at least 30%. This process has also reduced the amount of chemical-based sludge by 60% which was highly favourable. SEM images of starch treated flocs revealed distinct bridging of kaolin particles in-line with properties of respective coagulation mechanisms specifically adsorption and bridging. This study further conduces to the understanding of starches as coagulants and puts forward a basis for the characterization of resulting flocs.

1. Introduction

Coagulation-flocculation which can be achieved with the addition of coagulants; both of chemical and natural-based remains as one of the most efficient and simplest method adopted in water treatment industries. Often characterized by its anionic nature, colloidal matters which include kaolin particles would repel one another and remain suspended to form a stable suspension (Kim et al., 2001; Sincero and Sincero, 2002). Adsorption and charge neutralization on these particles induced by positively charged chemical coagulants would result in system destabilization. A secondary mechanism known as adsorption and bridging would then promote agglomeration of neighbouring particles by linking the microflocs formed to form macroflocs through the addition of polymers such as starches (Caskey and Primus, 1986). The starch solution which acts as a layer of netting would entangle multiple microflocs before binding them together over the course of stirring. With ongoing size increment until a steady state is reached, gravitational sedimentation of these macroflocs can be enhanced.

The application of chemical coagulants specifically inorganic coagulants is more widespread and heavily depended on owing to their superiority in treating turbid water, wide availability and cost-effectiveness (Duan and Gregory, 2003). Published records on the adverse impacts on human health have surfaced since the 1960s (Simate et al., 2012) with more worrying revelation on the potential development of Alzheimer's disease (AD) (Flaten, 2001; Gauthier et al., 2000; McLachlan, 1995; Rondeau et al., 2000; Walton, 2013) and senile dementia (Rondeau et al., 2001) with the use of aluminium salts namely alum.

Countries such as Malaysia classified aluminium-based sludge as Scheduled Waste (SW204) wherein 2 million metric tonnes of sludge per year was generated from a total of 462 water treatment plants (SPAN, 2010). This large amount of sludge is currently being treated in the country's only prescribed premise in Bukit Nanas, Negeri Sembilan which has resulted in excessive strain to the facility and also additional expenditures up to $ 6 million per day for sludge disposals only (SPAN, 2010). Thus, there is a need to consider other potential alternatives to minimize the use of aluminium salts. Natural coagulants such as starches could be the proposed solution.

* Corresponding author at: Chemical Engineering Discipline, School of Engineering, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia.
E-mail address: ramanan@monash.edu (R.N. Ramanan).

http://dx.doi.org/10.1016/j.ecoleng.2016.05.082
0925-8574/

Natural coagulants are largely non-toxic, eco-friendly and results in less sludge volume in some instances (Ndabigengesere et al., 1995). Research studies have proven the efficiency of *Moringa oleifera* seeds in the treatment of raw surface and synthetic turbid waters. Established coagulants such as tannins have also been used in real time applications. Besides fruit wastes (Choy et al., 2014) and other various vegetables and legumes (Choy et al., 2015), biopolymers such as starch is also gaining increasing interests for applications as potential natural coagulants. Being one of the most abundant biopolymer after cellulose with a projected global annual production of 85 million tonnes by 2015 (ISI, 2014), starch is attractive due to its biodegradability, non-toxicity and is also one of the lowest costing biopolymer.

Starch is commonly modified into various types of starch-grafted polymers notably cationic, anionic and amphoteric (Yang et al., 2014). The studies performed on other starch derivatives have also been discussed in a more recent review (Oladoja, 2015). Such chemical modifications are achieved via graft copolymerization whereby synthetic polymers such as acrylamide are combined with starch to enhance the overall molecular weight of polymer (Willett, 2009). Although enhanced turbidity removals can be achieved, negative impacts on both environmental and human health would be of great concerns. Instead, unmodified starches could be used as an aid in tandem with chemical coagulants to minimize the harmful impacts caused by these proprietary coagulants. Besides the removal of metal cations through ultrafiltration (Baharuddin et al., 2014), the use of unmodified starches has also been studied in the treatment of semiconductor wastewater (Mohd Omar et al., 2013) and applied as an aid to ferric chloride (Shahriari et al., 2012) with promising results that were worth further investigations. However, research areas utilizing native, heat treated-starches as natural coagulants particularly in the removal of turbidity remained scarce with the exception on studies conducted using palm oil mill effluent where total suspended solids removal above 80% have been reported (Teh et al., 2014a,b). The rheological properties of starches can easily be modified through gelatinization; a process defined by the rupture of starch granules under sufficient moisture and supplied heat leading to increased solution viscosity (Biliaderis, 2009). The release of starch fractions to the surroundings would facilitate the linking of colloidal particles through the bridging mechanism. As such, autoclaved starch solutions would also present enhanced viscosity which is one of the key factors leading to their potential use in treating turbid water. Similar to that of the earliest known Chinese water clarifying agent, glue made from animal hides were also used in the ancient times and their efficiencies were largely governed by the degree of viscosity and molecular mass (Jahn, 2001).

Motivated by the positive results reported, this research work aims to evaluate the performance of various conventional starches as low-cost natural coagulants and its suitability as a primary or coagulant aid to aluminium salts. Moreover, integrated use of starch with alum and polyaluminium chloride (PACl) was accessed by considering three different modes of coagulant additions; aid, together and two step coagulation. The respective coagulation mechanism involved was then identified. Additionally, characterization of starches used and flocs formed was performed using various microscopy techniques namely inverted routine microscopy and Scanning Electron Microscopy (SEM), Fourier Transform infrared spectroscopy (FTIR) and X-Ray Diffraction (XRD). This research study also aimed to elucidate the importance of starch gelatinization which resulted in enhanced agglomeration of kaolin particles leading to water clarification. Knowledge on the physicochemical properties of conventional starches would have significant contribution towards the understanding of the respective coagulation behavior and the explanation on the different coagulation activities observed.

2. Materials and methods

2.1. Materials

Kaolin heavy powder (Sigma-Aldrich) was used as a model suspension for the jar test studies. Four native starch samples of rice, wheat, corn and potato purchased from Sigma-Aldrich were studied for their feasibility as natural coagulants. For comparison purposes, aluminium salts were used as chemical coagulants in this study. Aluminium sulfate hydrate ($Al_2(SO_4)_3{\cdot}H_2O$) sourced from Sigma-Aldrich and liquid polyaluminium chloride 10% supplied by Holy Mate Sdn. Bhd., Malaysia were then prepared as solutions of 3% strength. The characteristics of these aluminium salts were summarized in Table S1. Distilled water was used to dilute hydrochloric acid (Merck) and to dissolve sodium hydroxide pellets (Merck) for the preparation of 1 M solutions.

2.2. Preparation of synthetic turbid water

Twenty grams of kaolin powder was added into 2 L of distilled water. The solution was mixed thoroughly and allowed to stand overnight for complete hydration. The top and bottom 400 mL was drained away before collecting the filtered stock kaolin solution using a strainer into a separate beaker. Model turbid synthetic water with medium initial turbidity of 165 ± 5 NTU was then prepared by diluting approximately 700 mL of kaolin stock solution with 3500 mL of distilled water. The turbid water was stirred continuously to prevent gravitational settling and a series of analysis − turbidity measurement, pH and zeta potential was performed. Similar model turbid water was prepared and used in all jar testing experiments.

2.3. Preparation of starch solutions

Starch solutions with 3% concentration were prepared using distilled water. To determine the effect of gelatinization on the feasibility of starches as natural coagulants, non-autoclaved and autoclaved starches were prepared. These solutions were prepared fresh when needed to avoid potential biodegradation leading to inconsistent results. For non-autoclaved starches, the prepared solutions were used directly in jar tests according to the desired coagulant dosage. As for autoclaved starches, the preparation was similar to previously published protocol (Teh et al., 2014b).

2.4. Coagulation-flocculation experiments

The conventional 4 jar apparatus (VELP Scientifica JLT4, Italy) was utilized in all the coagulation-flocculation experiments. A placebo (without the addition of any coagulants) was also prepared at the same starting conditions of the jar test. Any pH adjustments required were performed with the addition of 1 M HCl and 1 M NaOH solutions to obtain the desired pH values within ± 0.1 pH units. The beakers filled with 500 mL of synthetic turbid water were placed on the flocculator and agitated at the pre-selected rapid mixing intensity of 100 rpm. Subsequently, varying dosages of the studied coagulants (0–600 mg/L) were added depending on the respective mode of addition.

Rapid mixing was then continued at an intensity of 100 rpm for 2 min followed by slow mixing at 40 rpm for 20 min (Ndabigengesere et al., 1995). At the last 2 min of slow mixing, 5 mL of sample was collected for the measurement of zeta potential. Upon slow mixing, the beakers were removed and transferred to a flat, safe environment for sedimentation. After 30 min, 30 mL