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Current Review on the Coagulation/Flocculation of Lignin Containing Wastewater

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Abstract

Agro based industries including pulp and paper mill, palm oil mill, textile, dairy parlour etc. are characterised by its water intensive nature. The pollution problems related to the agro-based industries wastewater are colour, odour and toxicity. Lignin is the major colorant in the agro based industry wastewater. This review is dedicated to explore the lignin and its removal from wastewater via coagulation/flocculation. Over the years, several studies have been conducted for lignin removal using coagulation/flocculation. Aluminium based and iron based coagulants are widely used for this purpose. However, their usage could deteriorate the performance of biological post treatment. Among others coagulant, calcium based coagulant is seems to be an alternative. Hence, there is interest in developing the use of calcium based coagulants alone or hybrid calcium-other metals coagulant that might fully dismiss or minimize the adverse impact of the aluminium and iron based coagulants. On the whole it looks for the possibility of calcium based salt as a potential coagulant.

Keywords: Lignin; Coagulation; Agro based industry wastewater; Calcium

Introduction

Agro based industries such as pulp and paper mill, palm oil mill, textile and dairy parlour are characterised by water intensive nature [1]. The industry consumes considerable quantities of water for their processes including cleaning and washing purposes and then end up as coloured wastewater. The coloured wastewater might be due to the presence of plant component i.e. lignin, tannin [1] as well as its biodegradation product e.g. melanoidin [2].

The presence of colored compounds in surface water is aesthetically undesirable and causes disturbance of the aquatic biosphere due to reduction of sunlight penetration and depletion of dissolved oxygen. Some of the colored compounds are toxic and mutagenic and have the potential to release the carcinogenic amines [1,3]. Due to their recalcitrant properties, coloured compounds can also contribute to the failure of biological processes in wastewater treatment plants [4]. In some applications, coloured treated water is also not suitable for water reuse [2]. In addition, colour commonly increases throughout biological treatment systems, which may be due to the organic material being converted into smaller chromophoric units rather than being mineralized [5]. This review will focus on the lignin as the main colorant and its treatment using coagulation/flocculation. Prior to that, the lignin structure and class will be discussed.

Lignin

Lignin is the second most abundant natural raw material and nature's most abundant aromatic (phenolic) polymer [6,7] (as cited in Suhas [8]. Lignin is a natural polymeric product from an enzyme initiated dehydrogenative polymerization of the three primary precursors (Figure 1) [9,10]. In another study, Essington [11] proposed the lignin structure as shown in the Figure 2. There are three classes of lignin (Table 1): softwood, hard wood and grass lignin (as cited in Suhas [8]). Generally, lignin particles has negatively charge surface in water [4].

Coagulation/Flocculation

Coagulation/flocculation is one of the most important treatment steps in wastewater treatment plants. Coagulation is the destabilisation of particles using coagulant(s), which can be classified into three main categories: (1) inorganic-based coagulants, (2) organic-based flocculants and (3) hybrid materials [12]. Coagulation tends to overcome the factors that promote particles stability and form agglomerates or flocs. Flocculation is the process of whereby destabilised particles, or particles formed as a consequence of destabilisation, are induced to come together, make contact, and thereby form large(r) agglomerates [13]. The widely used coagulants for lignin containing wastewater are aluminium and iron based (Table 2). They were chosen since they are effective in removing organic substances.

Since in the normal water condition, the lignin particle is negatively charged, then the proposed mechanism of lignin removal could include [14-16]:

- Charge neutralization (inorganic-based coagulants, organic-based flocculants (e.g. poly-diallyl-dimethyl ammonium chloride), hybrid materials)
- Adsorption-precipitation (inorganic-based coagulants),
- Complex chemical reactions/chelation-precipitation (inorganic-based coagulants)
- Sweep coagulation (inorganic-based coagulants at alkali pH)
- Bridging (organic-based flocculants (e.g. poly-diallyl-dimethyl ammonium chloride), hybrid materials)
- Electrostatic patch (organic-based flocculants (e.g. poly-diallyl-

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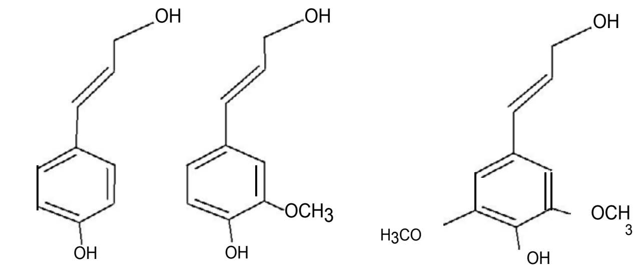

(a) p - courmary (b) coniferyl alcohol (c) sinapyl alcohol

Figure 1: Lignin precursors [9,10]

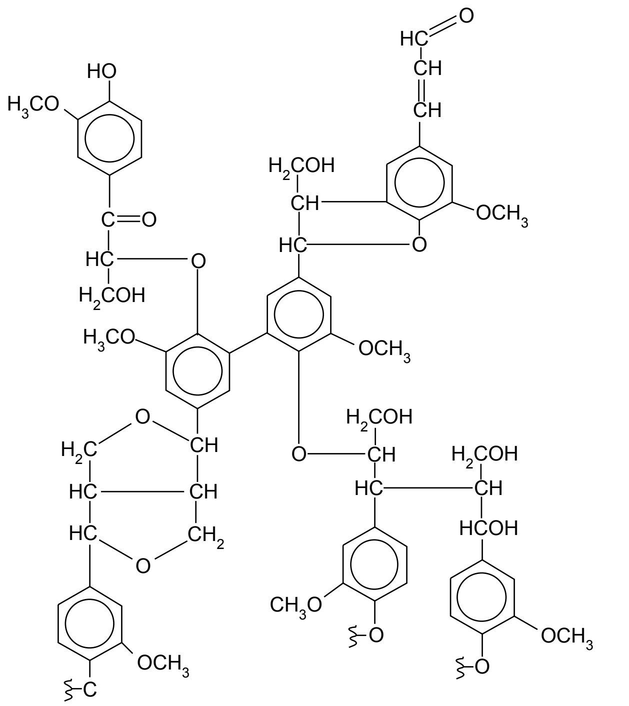

Figure 2: Possible structure of lignin [11]

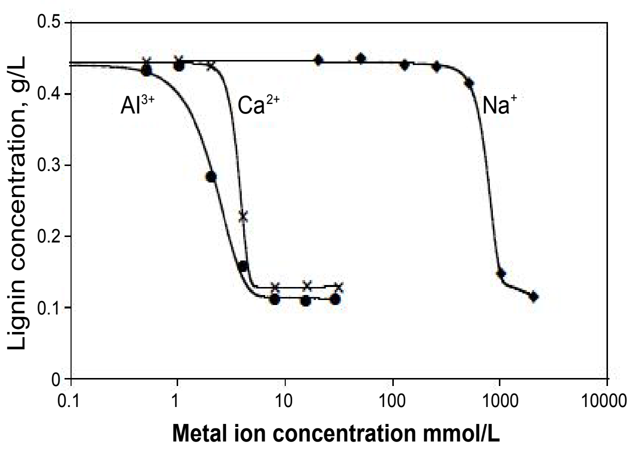

Figure 3: The influence of Al^{3+}, Ca^{2+} and Na^{+} on spruce kraft lignin in solution pH 9 and 25°C [24]

dimethyl ammonium chloride), hybrid materials)

The efficiency of coagulation or flocculation process depends on five important factors [12]:

(1) The rate of transport of coagulant or flocculant molecules to the lignin particles in the fluid,

(2) The rate of adsorption of coagulant/flocculant on the surface of lignin particles,

(3) The time scale needed for the coagulant/flocculant layer to reach equilibrium,

(4) The rate of aggregation of lignin particles having adsorbed coagulant/flocculant, and

(5) The frequency of collisions of lignin particles with adsorbed particles to form flocs

Several types of coagulants that can be used for treating lignin containing wastewater are shown in Table 2. Table 2 shows the performance of metal, polymer and hybrid metal-polymer coagulant. The most frequent metal coagulant used in treating agro based wastewater is aluminium sulphate (Table 2).

Ganjidoust [17] investigated aluminium sulphate and synthetic polymers for pulp and paper wastewater treatment. The result shows that the TOC and lignin removal is ~30% and ~80%, respectively. However, treatment using chitosan successfully remove ~90% lignin. Rohella [18] reported that aluminium chloride (10-20 mg/L) unable to remove colour efficiently. However, the application of cationic polyelectrolyte (0.2 mL/L) alone shows that the removal for colour is 82.58%.

Ahmad [19] investigated aluminium sulphate-polyaluminium chloride for palm oil mill effluent treatment. They stated that the COD and TSS removal is 95% and 98%, respectively. Ahmad [20] studied coagulation of pulp and paper mill effluent using aluminium sulphate (~1 g/L) and found that the highest percentage removal of COD, TSS and turbidity is 91%, 99.4% and 99.8%, respectively. Srivastava [21] that utilised poly aluminium chloride (PAC) followed by adsorption with bagasse fly ash (BFA). They reported that at the optimal condition (pH 3; PAC dosage 300 mg/L), it can remove about 80% COD removal and 90% colour [21].

Irfan et al. [22] investigated the performance of different coagulants and flocculants like alum, ferric chloride, aluminium

Class	Other name (lignin)	Unit	Reference
Soft wood	• Guaiacyl • Coniferous	Coniferyl alcohol	
Hard wood	• Dicotyledonous angiosperm	Coniferyl alcohol Sinapyl alcohol	Suhas [8]
Grass lignin	• Annual plant • Monocotyledonous angiosperm	Coniferyl alcohol Sinapyl alcohol p-coumaryl alcohol	

Table 1: Classes of lignin

Industry	Metal	Polymer	Operating Condition			Other	Lignin removal			Reference
			pH	Dosage (mg/L)	Temp (°C)	Removal (%)	Before (mg/L)	After (mg/L)	Removal (%)	
Metal										
Pulp & Paper mill	Aluminium sulphate	–	6	999.6		Turbidity-99.8 TSS-99.4 COD-91	–	–	–	Ahmad [20]
Pulp & Paper mill	Aluminium sulphate	–	7	30		–	–	–	88	Dilek and Bese [29]
Pulp & Paper mill	Aluminium sulphate	–	–	–		–	–	–	80	Ganjidoust [17]
Pulp & Paper mill	Ferric chloride + Aluminium sulphate	–	3	FeCl3-799.97 AlCl3-800.04		COD-18 TSS-49 Color-48	–	–	–	Irfan et al. [22]
Kraft pulp mill (washing water)	Sodium	_	9	22989.80 - 114949.00	25	_	450 - 400	150 -100	66.7 - 75.0	Sundin [22]
	Calcium	_	9	200.39 - 2805.46	25	_	451 - 400	150 -100	66.7 - 75.0	
	Magnesium	_	11	680.54 - 729.15	25	_	452 - 400	100 -50	77.8 - 87.5	
	Aluminium	_	9	134.91 - 1888.71	25	_	453 - 400	120 - 90	73.3 - 77.5	
Palm oil mill (AnPOME)	Calcium lactate		8-8.4	500	_	_	1173 - 1517	_	91	Zahrim et al. [30]
Polymer						_				
Sugar mill	_	PAC	3	300	_	COD-80	_	_	_	Srivastava et al. [21]
Oily	–	Poly-Zinc-Silicate-Sulfate (PZSS) + Anionic Polyacrylamide (APAM)	2	Zn/Si ratio - 1.00-1.50	Ambient	COD-superior TSS- 95 Turbidity- 96.3	–	–	–	Zeng and Park [31]
Paper and pulp	–	Polydadmac + Poly-acrylamide(PAM)	–	Polydadmac-1.20 PAM - 2.00	Ambient	COD-98 TSS-96.8	–	–	71.7	Ariffin [32]
Pulp mill	_	Acrylamide + Starch + 2-methyarcyloyloxyethyl trimethyl ammonium chloride(DMC)	8.35	22.30 and 22.30	Ambient	Turbidity-95.7 Water recovery-72.7	_	_	83.4	Wang [23]
Pulp mill	_	Chitosan + 2-methyarcyloyloxyethyl trimethylammonium chloride(DMC)	7.1	Chitosan - 17.80 DMC- 17.80	Ambient	Turbidity- 99.4 COD-90.7 Water recovery - 89.4	_	_	81.3	Wang [33]
Wood	–	Poly-aluminium Chloride	9.2	125	Ambient	COD-40.9 Color-83.8 LES-92.8	_	_	58.4	Brovkina [34]
Pulp	_	Poly-ethylene	2	350	Ambient	COD-15	_	_	15	Shi [35]
Pulp and paper	–	Natural polymer (chitosan)	–	–	–	TOC-70	_	_	90	Ganjidoust [17]
Metal-Polymer										
Pulp and paper	Aluminium sulphate	Poly-aluminium Chloride (PACl)	6	PACl: 500.00 Alum: 1000.00	Ambient	COD-91 TSS-99.4	–	–	–	Ahmad [36]
Pulp and paper	Ferric chloride	PACl (Poly aluminum chloride) + PAM (Poly acrylamide)	Ferric Chloride: 2 PAC: 3 PAM: 2	Ferric chloride and PACl: 200.00 PAM: 4.00	Ambient	COD - 81 TSS - 95 Colour - Not efficient	–	–	–	Irfan et al. [22]
Pulp and paper	Aluminum chloride + ferrous sulphate	Anionic PAM	Aluminium chloride and Ferrous Sulphate: 6	Aluminium chloride: 800.00 Ferrous sulphate: 800.00 PAM: 4.00		COD-76 TSS-95 Colour-95	–	–	–	Irfan et al. [22]

Palm oil mill	Aluminium sulphate	Poly-aluminium Chloride (PACl)	4.5	Alum : 8000.00 PACl: 600.00	Ambient	COD-95 TSS-98 Residue oil and suspended solids -99	–	–	–	Ahmad [19]
Palm oil mill	Aluminium sulphate + Activated Carbon	Cationic Polyacrylamide	6	Alum: 1700.00	Ambient	COD-85 TSS-99.9 BOD-86.3 Residue oil And suspended solids-95	–	–	–	Ahmad [37]
Pulp and paper	Aluminium sulphate	Syntheticpolymer (HE, PEI, and PAM)	6	–	–	TOC-30	–	–	80	Ganjidoust [17]
Pulp and paper	Aluminium sulphate	Modified natural polymer, starch-g-PAM-g-PDMC [polyacrylamide and poly (2-methacryloyloxyeth-yl) trimethyl ammonium chloride]	8.35	Alum: 871 mg/L, Flocculant dosage: 22.3 mg/L	–	Turbidity: 95.7 Water Recovery Efficiency: 83.4	–	–	72.7	Wang [23]
Kraft paper mill waste water	Aluminium sulphate	Anionic Polyelectrolyte	–	Alum: 300mg/L Anionic Polyelectrolyte : 0.05mg/L	–	Suspended Solid: 91.6 COD: 97	–	–	66.7	Pawels and Bhole [38]
Pulp and Paper	Aluminium Chloride (Alum)	Cationic Polyelectrolyte	–	Alum: 20mg/L Cationic Polyelectrolyte : 0.02mg/L	–	Turbidity: 96.26 COD: 55.65	–	–	82.58	Rohella [18]

Table 2: Coagulants Used to treat lignin containing wastewater

chloride, ferrous sulphate, poly aluminium chloride (PAC), cationic and anionic polyacrylamide polymers in individual form as well as in different combinations. They found that mixed coagulants were found to be more effective in reducing COD, TSS and colour than the individual form. Combination of ferric chloride, PAC and cationic polymer was excellent for reduction of 81% COD and 95% TSS while combination of Aluminium chloride, PAC and Anionic PAM was good in 88% colour reduction [22].

In another study, Wang [23] reported coagulation/flocculation of pulp and paper mill wastewater using aluminum chloride as coagulant and a modified natural polymer, starch-g-PAM-g- PDMC [polyacrylamide and poly (2-methacryloyloxyethyl) trimethyl ammonium chloride], as flocculant aids. They concluded that the coagulation/flocculation was able to reduce the turbidity up to 95.7% with water recovery as much 72.7% [23].

Adverse effect of iron and aluminium based coagulants

From the previous studies, it can be shown that high valence coagulants such as aluminium sulphate and ferric chloride; were being used (Table 2). This probably because as the valence electron increased, the metal ion concentration for coagulation decreases [24] (Figure 3); and this affected the choice of coagulant due to the lower cost. However, these conventional coagulants have been shown to produce less biodegradable wastewater since the coagulation process also remove the amino acids, proteins, and long-chain fatty acids from wastewater [25]. Moreover, it also have been reported that the residual alum and ferric based coagulants (initial concentration for both coagulants is 25 mg/L) inhibit the biological treatment process which is indicated by the reduction of microorganism respiration rate and low organic matter removal [26]. Besides that, the predatory growth in biological wastewater treatment plant also enhanced significantly [26].

The effects of alum addition on the treatment performance of membrane bioreactor were investigated by Zahid and El-Shafai [27]. The authors reported that the alum doses above 60 mg/L have toxic effect on the autotrophic bacteria with significant reduction in ammonia oxidation and hence the nitrogen removal [27]. Furthermore, the addition of iron and aluminium based coagulants could turn treated wastewater into acidic condition [14,28] in which the neutralization step is required prior to biological treatment.

Potential of Calcium Based Coagulant

Among others coagulant, calcium based coagulant is seems to be an alternative. Hence, there is interest in developing the use of calcium based coagulants alone [39,40] or hybrid calcium-other metals coagulant [41,42] that might fully dismiss or minimize the adverse impact of the aluminium and iron based coagulants. Herewith, we give some examples of the treatment lignin containing wastewater using various calcium based coagulants.

Olive mill wastewater (OMW) generated by the olive-oil extracted industry is a great pollutant contributor in Mediterranean countries. Aktas [43] proposed lime pretreatment proposed to reduce the polluting effect of OMW is an applicable method in practice since lime can easily be purchased anywhere and it is cheaper than other chemicals such as aluminium sulfate, ferric chloride, magnesium sulfate, etc., used for pretreatment of wastewaters [43]. After lime treatment process, chemical oxygen demand (COD) values of the wastewater samples could be reduced by 41.5–46.2% in pressing and centrifugal methods, respectively. The average removal percentage of the other parameters are 29.3–46.9% for total solids, 41.2–53.2% for volatile solids, 74.4– 37.0% for reduced sugar, 94.9–95.8% for oil-grease, 73.5–62.5% for polyphenols, 38.4–32.0% for volatile phenols and 60.5–80.1% for nitrogenous compounds, respectively [43]. The application of $Ca(OH)_2$ for the removal of soluble COD, phenolic and polyphenolic like compounds, and other organic compounds responsible for the olive mill wastewater (OOWW) colour has been studied by Boukhoubza [44]. For 1% lime concentration, the contents of COD and TSS are reduced to around 72%, 73% and 60% for polyphenols [44].

Palm oil mill industry is one of the main agro based industry in Malaysia andIndonesia. Calcium lactate has been used before by Zahrim et al. [30] for the removal of lignin in aerobic palm oil mill effluent (AnPOME) and it shows that 91% of lignin- tannin was removed from the AnPOME which initially had a range concentration of lignin-tannin 1173-1517 mg/L using 500 mg/L of calcium lactate.

Conclusion

Coagulation of lignin particles is important to ensure the effectiveness of wastewater treatment plant. Composite coagulants were found to be more effective in reducing lignin particles than the individual coagulant. Over the years, the treatment of lignin containing wastewater was dominated by aluminium and iron based coagulants. Application of these conventional coagulants should be minimised especially if the next treatment is biological. In this study, the calcium based coagulant has an advantage tend to more biodegradable treated water than the conventional aluminium and iron based coagulants. However, published literatures on this matter are lacking. In addition, the calcium based power of coagulating can be increased by the addition of suitable additive(s).

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