



Decolorization of aqueous coffee and tea infusions by chemical coagulation

Sri Chandana Panchangam^{a,*}, Karthikeyan Janakiraman^b

^aDepartment of Civil Engineering, Annamacharya Institute of Technology and Sciences, Kadapa 516003, India
Tel. +91 8562 201003; Fax: +91 8562 201005; email: pscl7@rediffmail.com

^bDepartment of Civil Engineering, Sri Venkateswara University, Tirupati 517502, India

Received 5 May 2013; Accepted 12 August 2013

ABSTRACT

The production and consumption of coffee and tea results in the generation of huge quantities of colored wastewaters, and these are accountable for certain problems like color, BOD, toxicity, and odor when discharged into the aquatic environment. In this study, the color from the aqueous infusions of coffee and tea was removed by chemical coagulation, using coagulants like ferrous sulfate, alum, and lime, and the chemical coagulation was also used for the evaluation of both the process efficiency and process parameters, such as pH, dose of coagulant, effect of coagulant type, etc. A coagulant dosage of 1.0 g L⁻¹ was considered as appropriate, since a major amount (70–97%) of color removal was accomplished from different brands of coffee and tea infusions. Higher efficiency of decolorization was obtained from slightly acidic to neutral pH (4.0–6.0). Higher percentage of color removal was achieved from tea infusions compared with coffee infusions. Average color removal efficiencies of individual coagulants for coffee and tea infusions were compared in order to assess the overall efficiency of the coagulant toward the infusions. Ferrous sulfate is used as an effective coagulant for coffee with 74% removal of color, and lime is used for tea infusion with 88% removal of color. The color of the infusions has been reduced from brown and brownish red to relatively clear liquid to an extent that it can be released into the aquatic environment without any detrimental effect.

Keywords: Aqueous coffee infusion; Aqueous tea infusion; Chemical coagulation; Decolorization; Polyphenols

1. Introduction

Coffee and tea are frontrunners in the list of most popular beverages consumed throughout the world. Tea is a pleasant beverage that is consumed by two-thirds of the world population as a part of culture, trend and to alleviate the senses. India is the leading country on global scale to produce and export various varieties of tea and sixth in position with three

hundred million kilograms of coffee being produced annually. Coffee (beans) and tea (leaves) contain an assortment of chemicals; some of them like tannins, alkaloids (caffeine), polyphenols, various acids, hydrocarbons, alcohols, furans, mercaptans, polysaccharides, carbohydrates, etc. which become toxic and exert detrimental effects when discharged into the aquatic environment. Some of these chemicals are responsible for the color of coffee and tea. Coffee industries generate huge quantities of wastewaters, and their amount,

*Corresponding author.

however, differs depending upon the type of processing of coffee beans (dry, wet or semi-wet process). Those process are also used for the processing of tea. Consequently, a momentous percentage of tea and coffee effluents can be anticipated in the wastewaters. The tinted effluents tend to inhibit the growth of micro-organisms responsible for biological degradation of organic materials in the wastewaters leading to anaerobic conditions emanating obnoxious odors. The main color-producing compounds in coffee are the melanoidins, which are the by-products of Maillard reaction, and in tea, the principal color-producing compounds are theaflavins and thearubigins that are red and red-brown in color, respectively, in addition to the phenols, tannins, lignins, and melanins [1,2]. Compounds like melanoidins have a tendency to chelate with the metal ions leading to direct intoxication of aquatic biota that are responsible for re-oxygenation [1,2]. Though the color of the caffeinated beverages is one of its attributes in food industry, it is aesthetically unacceptable in the case of environmental pollution. Color is said to be a singular monitor of pollution. Therefore, decolorization describes an essential objective in search for a method to eliminate color from the effluents in consort with their pollutant properties.

Macromolecules that are responsible for imparting color to the aqueous coffee and tea infusions like lignins, tannins, and humic acids are recalcitrant and are not amenable for degradation by conventional biological processes and hence are not remarkable for the reduction of color and COD of coffee and tea wastewaters [3–6], and therefore, dark color will be present in spite of significant removal of COD. Reprehensibly, the color of the effluent is alleviated due to the re-polymerization of the color compounds though the multistage biological treatment can reduce the organic loading of molasses wastewater [7]. Several investigations have been carried out on decolorization of the colored effluents from different matrices like pulp and paper mills, Maillard reaction products, phenols, tannins, textile mill effluents, etc. employing different techniques like adsorption, advanced oxidation processes, membrane bioreactors, etc. [8–13]. In recent years, a credible amount of attention is drawn by food industry wastewater and in particular coffee industry effluents; as a result, a significant number of groups have studied the treatment of coffee wastewater for the reduction of COD [14] and color by processes other than biological. Color reduction from coffee wastewaters were studied by technologies like electrochemical oxidation [15], Photo-Fenton process [16], and coagulation/flocculation in conjunction with advanced oxidation processes [17]. Despite of higher efficiency in color reduction, the methods reported so

far are cost-intensive, and hence, there is a necessity for cost-effective technologies. The venerable yet valuable technique for decolorization of effluents is coagulation that is always easy on pocket. Several studies were dedicated to assess the competence of this physicochemical process for color removal [18,19].

The vital objective of this study is to explore the potentiality of coagulation to remove color from aqueous coffee and tea infusions. Efficiency of three different coagulants ferrous sulfate, alum, and lime were evaluated for color removal. The effect of dosage of coagulant and pH on the coagulation process for the decolorization was also investigated.

2. Materials and methods

The coagulants used were ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), lime (CaO), and alum ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) and were obtained from Wako pure chemical industries Ltd., Taiwan. No coagulant aids were employed. About 1M NaOH and 1M H_2SO_4 reagent grade solutions were used to adjust the pH and were obtained from Acros organics, New Jersey.

Aqueous infusions of coffee and tea were prepared by adding 1.0 g of readily available commercial grade coffee and tea powders purchased from local market into one liter of boiling DI (De-ionized) water and brewing the suspension for 30 min with occasional stirring. The solid matter from the brew was removed by filtration with Advantec 5C quantitative ashless filter paper. Freshly prepared infusions were used for every experiment. The filtrate was measured for color, total dissolved solids (TDS), and pH. The pH of the solutions was measured with Suntext Microprocessor pH meter 2000A, Suntext Instruments Co. Ltd., Taiwan. A UV-vis spectrophotometer (JASCO V-550) was used to measure the absorbance of the solution. The absorbance of the solutions was measured at 455 nm wavelength against water as blank. A wavelength of 455 nm was used, since no specific peak was observed within 400–700 nm wavelength range, and particularly, 455 nm was chosen because it marks the beginning of the visible wavelength range. All the glassware used was of Pyrex quality. The commercial brands of coffee and tea powders used in this work are listed in Table 1.

Jar test apparatus was used to conduct coagulation experiments with 100 mL of 1 g L^{-1} coffee or tea infusions in each of the six beakers. A flash mixing of 10 min at 80–85 rpm was done, followed by a slow-agitation for 15 min at 30–35 rpm. The reaction mixture was allowed to settle, and then, the supernatant was measured for the residual color. All the experi-

Table 1
Commercial brands of coffee and tea used in this work

	Brand	Acronym	TDS, mg/L	pH	Color, Pt-Co units
Tea powder	AVT supreme	AVT	290	4.83	1,982
	Yellow Label tea	YLT	287	4.90	866
	Red Label tea	RLT	295	4.80	933
Coffee powder	Double Roast coffee	DRC	210	4.66	1,035
	R.K Green Label coffee	RGLC	215	4.65	1,018
	Nescafe coffee	NCC	209	4.81	908

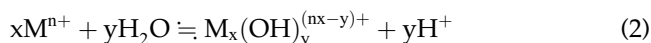
ments were carried out at room temperature ($25 \pm 2^\circ\text{C}$). The effect of pH on coagulative removal was studied by varying the pH of the test infusions from 2.0 to 12.0 at increments of 2.0 and employing a constant dosage of coagulant of 1.0 g L^{-1} . At constant pH (pH at which maximum color removal was observed), the effect of dosage of coagulant on color removal from aqueous coffee and tea infusions was studied using different dosages of 1.0, 2.0, 3.0, and 4.0 g L^{-1} . Color measurements in this study were reported as true color assayed at 455 nm against DI water as blank using a spectrophotometer and following Standard Method for the Examination of Water and Wastewater [20], Method no. 2120C and reported as platinum–cobalt (Pt–Co) color units. According to this method, a unit of color is produced by $1.0 \text{ mg platinum/L}$ in the form of the chloroplatinate ion. Color removal was evaluated by measuring color concentrations before and after the experiment with the help of calibration curve and represented using the following formula.

$$\text{Removal of color (\%)} = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where C_i is the initial color concentration in Pt–Co units, and C_f is the final concentration of color of aqueous infusion in Pt–Co units.

3. Results and discussion

The effectiveness of the coagulation process is influenced by the coagulating agent, the coagulant dose, the solution pH, and the ionic strength. Furthermore, concentration and the nature of the organic compounds also exert some influence on the color removal process. When the coagulants are dissolved in water, the metal ion (M) is hydrated to form monomeric and polymeric species. The hydrolysis reaction of metal coagulants may be represented as follows:



The resulting metal hydroxide polymers are amorphous in structure with a positive charge and possess a very large surface area that aids in the removal of color molecules from the infusions.

3.1. Effect of pH

The coagulation process is largely affected by pH, and the dependence is ascribed to the balance of two competitive forces: one between H^+ and metal hydrolysis products for organic ligands and the other for the competition between hydroxide ions and organic anions for metal hydrolysis products. The color removal efficiency from coffee and tea infusions of the three coagulants alum, ferrous sulfate, and lime at different pH values are shown in Figs. 1(a–c), respectively.

The trend in color removal adopts the same fashion for all the types of coffee infusions with alum and ferrous sulfate as coagulants: an increase in color removal with increase of pH, having maximum removal at around slightly acidic to neutral pH range (4.0–6.0) and decrease in the removal of color at alkaline pH values. At highly acidic pH ($\text{pH} < 4$), the acids present in tea and coffee are not likely to dissociate

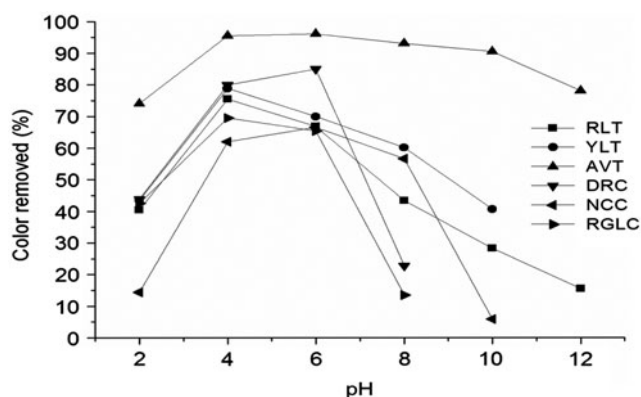


Fig. 1(a). Effect of pH on color removal from 1 g/L initial concentration of aqueous coffee and tea infusions by 1 g/L dosage of FeSO_4 at room temperature ($25 \pm 2^\circ\text{C}$).

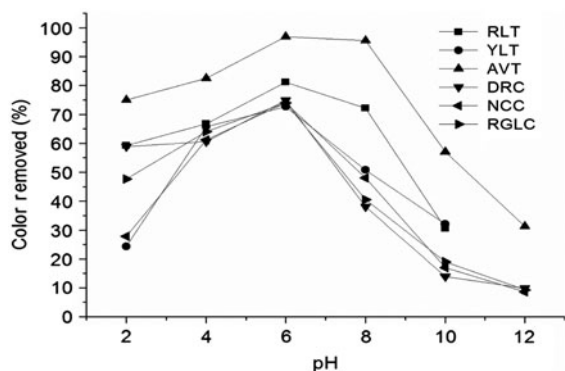


Fig. 1(b). Effect of pH on color removal from 1 g/L initial concentration of aqueous coffee and tea infusions by 1 g/L dosage of alum at room temperature ($25 \pm 2^\circ\text{C}$).

and, in addition, metal ions are not expected to form hydroxides in that pH range. An increase in the color removal efficiency at slightly acidic pH can be expected due to the formation of a large number of positively charged flocs, but a decrease in the color removal at alkaline pH is perhaps due to the repulsion of negatively charged flocs and a negatively charged color colloid [21]. Further, under highly alkaline pH values ($\text{pH} > 12$), the coagulating species become less positively charged diminishing their attraction to the anionic organic compounds [22]. In case of lime, the trend (Fig. 1(c)) follows a different pattern with a trivial increase in the color removal efficiency with an increase in the pH value. The marginal increase in the color removal at high alkaline pH when lime is used as a coagulant may be due to sorption onto the hydroxide flocs of lime. These observations are in

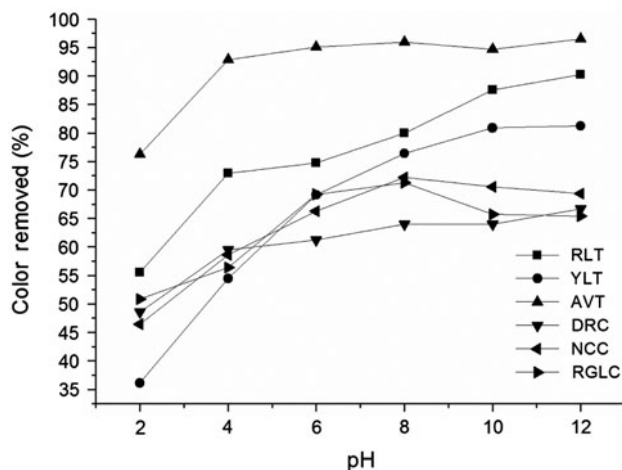


Fig. 1(c). Effect of pH on color removal from 1 g/L initial concentration of aqueous coffee and tea infusions by 1 g/L dosage of lime at room temperature ($25 \pm 2^\circ\text{C}$).

agreement with the findings of Orozoco [23], who found that calcium oxide when added to coffee wastewater is precipitated as calcium pectate if it contains a huge amount of mucilage pectin. These melanic pigments are soluble in the alkaline media but are believed to be insoluble in the water, acid, and in the organic solvents [2]. A contrast effect is observed when alum and ferrous sulfate were used as coagulants. In the case of color removal from AVT tea infusion using alum as a coagulant, 97% removal is observed at the neutral pH range which decreased in both acidic and basic pH environment. The hydrolysis product of alum is aluminum hydroxide, which is having amorphous structure; has its solubility both in acidic and basic pH—while it is slightly dissolvable at pH of 6.2; therefore, at the neutral pH range, most of the aluminum hydroxide is precipitated as Al-coagulant that can adsorb the colored molecules [24]. Usually, at lower pH, the protons outcompete the metal hydrolysis products for organic ligands and poor removal occurs while at acidic pH, hydroxyl ions competes with organic compounds for absorption sites resulting in charge-charge repulsions and consequently less removal is observed. In few instances in this study, the intensity of color of coffee infusions (DRC and RGLC) increased due to the addition of ferrous sulfate especially at higher doses, and this result indicates an increase in the color concentration compared with the stock infusion. Though this may be construed as negative color removal, it is considered as no color removal.

3.2. Effect of coagulant dosage

An optimum dosage of coagulant is an important parameter as the coagulant dose is a key determinant of organic removal efficiency and mechanisms. The effect of the coagulant dosage on the removal of color from coffee and tea infusions was studied at optimum pH, that is, pH at which the maximum color reduction was observed for a given type of infusion. The results of the experiments are depicted graphically in Figs. 2(a-c).

It is apparent from the Figures that the efficiency of decolorization increases, though less significantly, with an increase in the dosage of the coagulant from 1.0 to 4.0 g L⁻¹. The effect of a dosage of 1.0 g L⁻¹ of the coagulant is plausible as a major amount of color (70–97%) is removed, and the variation in efficiency of color removal is not striking beyond a coagulant dosage of 1.0 g L⁻¹; increment in maximum color reduction being 14% for a fourfold increase in the dosage of the coagulant. Though there is no significant variation in the trend, apparently there are some interesting pat-

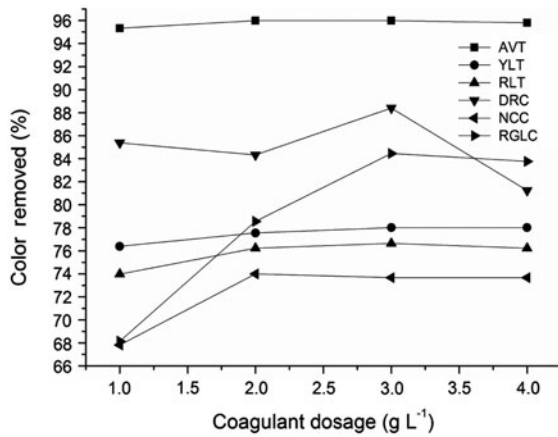


Fig. 2(a). Effect of coagulant dosage on color removal from aqueous coffee and tea infusions with FeSO_4 as coagulant.

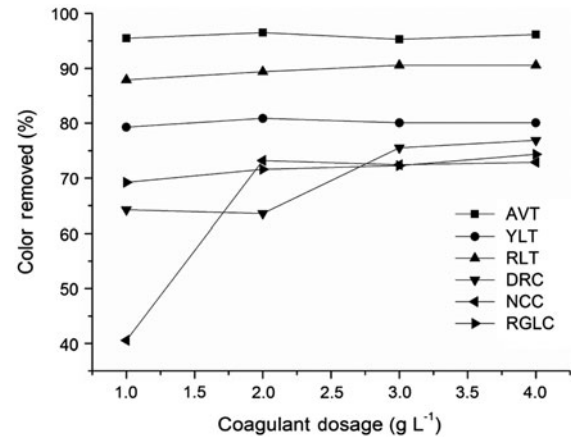


Fig. 2(c). Effect of coagulant dosage on color removal from aqueous coffee and tea infusions with lime as coagulant.

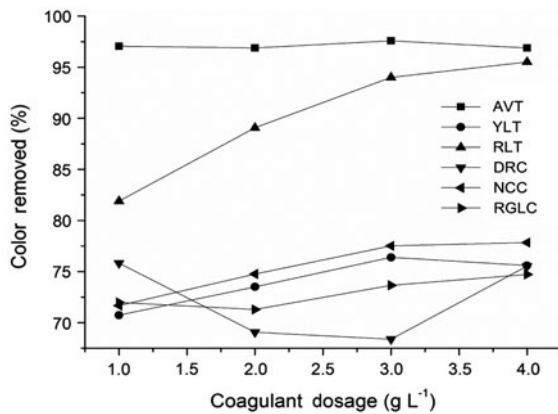


Fig. 2(b). Effect of coagulant dosage on color removal from aqueous coffee and tea infusions with alum as coagulant.

terms in behavior of coagulants with change in the dosage. In case of alum as a coagulant for DRC coffee infusion, a decrease in the color removal with an increase in the dosage of alum from 1.0 to 3.0 g L^{-1} (75.83–68.39%) was observed, while a further increase in the dosage of coagulant to 4.0 g L^{-1} has caused the color removal percentage (76%) to reach almost the same level of efficiency as that of 1.0 g L^{-1} dosage of the coagulant. This pattern of color removal is typical of physiochemical coagulation process indicating destabilization and restabilization of the colloidal matter. The restabilization of the colloidal matter occurs when coagulants were used at dosages higher than the optimum value [18]. A higher percentage of color removal is observed in tea infusions (e.g. 97% color removal from AVT and 76% from DRC with alum as coagulant) when compared to the coffee infusions. This can be presumed due to the difference in chemical structures and the solubility patterns of the two

infusions (coffee and tea). A coagulant dosage of 1.0 g L^{-1} was considered as an appropriate dose in this study as on an average, and 72–85% of color removal was accomplished from both the coffee and tea infusions, respectively. A comparison of color removal efficiency among the tea and coffee infusions reveal that a higher color removal of value greater than 95% was achieved for AVT in case of tea infusions, while a removal efficiency of between 65 and 85% was achieved for DRC among coffee infusions.

3.3. Effect of type of coagulant

The type of coagulant also affects the coagulation process. In this study, three coagulants, two of them divalent (Fe^{2+} and Ca^{2+}) and the other a trivalent (Al^{3+}), metal species were investigated. Average value of the color removal efficiencies from aqueous coffee and tea infusions at optimum dosage of coagulant (dosage = 1 g L^{-1}) and at optimum pH (pH = 6.0) condition was calculated in terms of percentage average decolorization efficiency (η_{Avg}) as:

$$(\eta_{\text{Avg}})_{\text{Tea}} = (\eta_{\text{AVT}} + \eta_{\text{YLT}} + \eta_{\text{RLT}})/3 \quad (3)$$

$$(\eta_{\text{Avg}})_{\text{Coffee}} = (\eta_{\text{DRC}} + \eta_{\text{NCC}} + \eta_{\text{RGLC}})/3 \quad (4)$$

where η_{Avg} is the average color removal efficiency as percentage of coffee/tea infusion for a particular coagulant at optimum dosage of coagulant and optimum pH and η_{suffix} is the efficiency of color removal from a brand of coffee/tea at the optimum conditions for a particular coagulant. Values of average color removal efficiency for different types of coagulants are given in Table 2.

Table 2

Comparison of performance of different coagulants by average decolorization efficiency

	Average color removal from tea infusion (η_{Avg}^{Tea} (%))	Average color removal from coffee infusion (η_{Avg}^{Coffee} (%))
Ferrous sulfate	82	74
Alum	84	73
Lime	88	68

Note: Average color removal from coffee/tea infusion with optimum dosage of coagulant (1 g/L) and at optimum pH (pH = 6) in % = Average of color removal percentages of individual coffee/tea infusions at corresponding conditions.

Interestingly, the order of efficiency for the removal of color from coffee infusion is ferrous sulfate > alum > lime, and the order is reverse in the case of tea infusion as ferrous sulfate < alum < lime. Ferrous sulfate is having higher efficiency for coffee color removal and least for tea infusion, while lime is having higher color removal efficiency for tea infusion with least efficiency toward coffee infusion. Though the difference in values of efficiency of the different coagulants was not significant, the trend seems to be interesting following a particular order. The main color-producing compounds present in coffee effluents are melanoidins. It was shown by Tamaki et al. [25] that the Fe^{2+} binding ability of melanoidins is strong. A significant decolorization was achieved by ferrous-initiated reaction when a model coffee effluent was subjected to decolorization with Photo-Fenton process [16]. Also, it is a well known fact and probably an experience of many, which when tea is brewed in hard water containing lime, the infusion looks dull because of the formation of scum especially when the tea used is low in concentration. When the pH of the polyphenols occurs rendering the tea infusion a dark brown color. Reliably, under the influence of calcium and bicarbonate ions, tea solubles are further oxidized at the liquid/air interface to form higher molecular weight products, and the most of the scum consists of these products, mutually with some reformed $CaCO_3$ (<http://www.norbentea.com/quality.html>). Further, calcium ions have ability to destabilize the negatively charged melanoidins [26]. This indicates the affinity of the color-producing compounds of tea toward lime, and this may be the probable reason for the higher color removal percentage.

4. Conclusions

Ferrous sulfate is effective for the removal of color from aqueous coffee infusion (75%), whereas lime is found to be effective for the removal of color from tea infusion (87.54%). The percentage values for the removal of color from tea infusions (70–97%) are higher than that of coffee infusions (64–85%), and this may be due to the difference in its chemical composition and the affinity of the color-producing compounds toward the coagulants. Astill et al. [27] have shown that the concentrations of the components present in tea beverages as consumed are dependent on the method of preparation including the quantity of tea and water used, infusion time, and the amount of agitation. Extrapolating further, this kind of dependency can be assumed for coffee beverages also, and this is in addition to the type/nature of tea/coffee and the method of their manufacture. Maximum efficiency of color removal was achieved in most of the cases at pH values 4.0–6.0. An optimum coagulant dosage of 1.0 g L^{-1} was adopted in all the cases considering the engineering and economical aspects of coagulation process. Overall, the color of the infusions has been reduced from brown and brownish red to clear liquid, thus achieving the goal of removing color to an extent that it can be released into the aquatic environment without any detrimental effect.

Abbreviations

AVT	— AVT supreme
YLT	— Yellow Label tea
DRC	— Double Roast coffee
RGLC	— R.K. Green Label coffee
NCC	— Nescafe Coffee
Mn^+	— metal ion
$M_x(OH)_y^{(nx-y)+}$	— metal hydroxide polymers
C_i	— initial color concentration
C_f	— final color concentration
$(\eta_{Avg})^{Tea}$	— average color removal efficiency as percentage of tea infusion for a particular coagulant at optimum dosage of coagulant and optimum pH
$(\eta_{Avg})^{Coffee}$	— average color removal efficiency as percentage of coffee infusion for a particular coagulant at optimum dosage of coagulant and optimum pH
$\eta_{AVT}, \eta_{YLT}, \eta_{RLT}, \eta_{DRC}, \eta_{NCC}, \eta_{RGLC}$	— efficiency of color removal from a brand (represented by suffix) of coffee/tea at the optimum conditions for a particular coagulant

References

- [1] S.A. Wiseman, D.A. Balentine, B. Frei, Antioxidants in tea, *Crit. Rev. Food Sci. Nutr.* 37 (1997) 705–718.
- [2] V. Sava, S. Yang, M.Y. Hong, P.C. Yang, G.S. Huang, Isolation and characterization of melanic pigments derived from tea and tea polyphenols, *Food Chem.* 73 (2001) 177–184.
- [3] R. Bello-Mendoza, M.F. Castillo-Rivera, Start-up of an anaerobic hybrid (UASB/Filter) reactor treating wastewater from a coffee processing plant, *Anaerobe* 4 (1998) 219–225.
- [4] R.M. Dinsdale, F.R. Hawkes, D.L. Hawkes, The mesophilic and thermophilic anaerobic digestion of coffee waste containing coffee grounds, *Water Res.* 30 (1996) 371–377.
- [5] F.J. Morales, Assessing the non-specific hydroxyl radical scavenging properties of melanoidins in a Fenton-type reaction system, *Anal. Chim. Acta* 534 (2005) 171–176.
- [6] J. Dahiya, D. Singh, P. Nigam, Decolourisation of synthetic and spentwash melanoidins using the white-rot fungus *Phanerochaete chrysosporium* JAG-40, *Bioresour. Technol.* 78 (2001) 95–98.
- [7] M. Pena, M. Coca, G. Gonzalez, R. Rioja, M.T. Garcia, Chemical oxidation of wastewater from molasses fermentation with ozone, *Chemosphere* 51 (2001) 893–900.
- [8] P. Sri Chandana, J. Karthikeyan, Sorptive removal of color from aqueous coffee and tea infusions, *Desalin. Water Treat.* 50 (2012) 338–347.
- [9] S. Arda, A. Bruce, V. Gokmen, Adsorption of Maillard reaction products from aqueous solutions and sugar syrups using adsorbent resin, *J. Food Eng.* 82 (2007) 342–350.
- [10] V.V. Basava Rao, S. Ram Mohan, Adsorption studies on treatment of textile dyeing industrial effluent by flyash, *Chem. Eng. J.* 116 (2006) 77–84.
- [11] K.C. Namkung, A.E. Burgess, D.H. Bremner, H. Staines, Advanced Fenton processing of aqueous phenol solutions: A continuous system study including sonication effects, *Ultrason. Sonochem.* 15 (2008) 171–176.
- [12] T.S. Anirudhan, M. Ramachandran, Adsorptive removal of tannin from aqueous solutions by surfactant-modified bentonite clay, *J. Colloid Interface Sci.* 299 (2006) 116–124.
- [13] M. Brik, P. Schoeberl, B. Chamam, R. Braun, W. Fuchs, Advanced treatment of textile wastewater towards reuse using a membrane bioreactor, *Process Biochem.* 41 (2006) 1751–1757.
- [14] Y. Aguilera, R. Consuegra, M. Rapado, Radiation technology for conservation of the environment, in: *Proceedings of a Symposium, Zakopane, Poland, September 8–12, 1997.*
- [15] R.S. Bejankiwar, K.S. Lokesh, T.P. Gowda, Colour and organic removal of biologically treated coffee curing wastewater by electrochemical oxidation method, *J. Environ. Sci.* 15 (2003) 323–327.
- [16] M. Tokumura, O. Ayanto, H.T. Znad, K. Yoshinori, UV light assisted decolorization of dark brown color coffee effluent by photo-Fenton reaction, *Water Res.* 40 (2006) 3775–3784.
- [17] Z.P. Teresa, G. Gunther, H. Fernando, Chemical oxygen demand reduction in coffee wastewater through chemical flocculation and advanced oxidation processes, *J. Environ. Sci.* 19 (2007) 300–305.
- [18] A.A. Hamidi, S. Alias, Md. Nordin Adlan, A.H. Faridah Asaari, Md. Shahrir Zahari, Colour removal from landfill leachate by coagulation and flocculation processes, *Bioresour. Technol.* 98 (2007) 218–220.
- [19] Z. Song, C.J. Williams, R.G.J. Edyvean, Treatment of tannery wastewater by chemical coagulation, *Desalination* 164 (2004) 249–259.
- [20] APHA, AWWA, WEF, *Standard Methods for the Examination of Water and Wastewater*, 20th ed., American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC 1998.
- [21] D. Karadag, S. Tok, E. Akgul, K. Ulucan, H. Evden, A. Mehmet Kaya, Combining adsorption and coagulation for the treatment of azo and anthraquinone dyes from aqueous solution, *Ind. Eng. Chem. Res.* 45 (2006) 3969–3973.
- [22] H.W. Ching, T.S. Tanaka, M. Elimelech, Dynamics of coagulation of kaolin particles with ferric chloride, *Water Res.* 28 (1994) 559–569.
- [23] R.A. Orozoco, *Proceedings of ASIC conferences. 6th Colloquium: Chemistry in relation to technology, Bogota, Columbia, 290–296 (1973).*
- [24] C.L. Yang, J. McGarrah, Adsorption and desorption of phenol on activated carbon and a comparison of isotherm models, *J. Hazard. Mater.* B127 (2005) 40–47.
- [25] M. Tamaki, M. Murata, S. Homma, Characterization of brown pigment in sautéed onion by metal-chelating Sepharose 6B column chromatography and microbial decolorization, *LWT—Food Sci. Technol.* 40 (2007) 144–150.
- [26] Y. Satyawali, M. Balakrishnan, Removal of color from biometanated distillery spentwash by treatment with activated carbons, *Bioresour. Technol.* 98 (2007) 2629–2635.
- [27] C. Astill, M.R. Birch, C. Dacombe, P.G. Humphrey, P.T. Martin, Factors affecting the caffeine and polyphenol contents of black and green tea infusions, *J. Agric. Food Chem.* 49 (2001) 5340–5347.