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RESEARCH ARTICLE

A Comparative Assessment Study on Natural Coagulant and Regenerated Coagulant

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Abstract

Surface water brings impurities in the form of suspended and colloidal solids. Coagulation and flocculation followed by filtration and disinfection process involved in the treatment of surface water. This process generates large quantity of waste generally known as water treatment sludge (WTS). A typical Water Treatment Plant (WTP) produces about 100,000 tons/year of sludge whereas, on a global scale, available literature estimates that at present the daily production of sludge exceeds 10,000 tons (Babatunde and Zhao, 2007). Increase in demand for milk and dairy products, dairy industries in most of the countries of the world, including India will lead to enhanced productivity, as a result increase in wastewater generation and environmental pollution. The objective of the present study is to determine WTS characteristics obtained from WTP and to investigate the reuse of recovered/regenerated coagulant in synthetic dairy effluent and compare with natural coagulant (Moringa Oleifera Seed Powder (MOSP)). The characterization of WTS shows the maximum amount of SiO_2 at 67.75% by using Energy-Dispersive X-Ray Fluorescence (ED-XRF). The other oxides also observed by ED-XRF such as Al_2O_3 (16.76%), Fe_2O_3 (5.52%) and MgO (3.33%). For the optimum recovery of the coagulant, WTS acidified with the different normality of sulfuric acid. The RC produced similar turbidity removal efficiency (94%) at 25ml/l dose as compared to natural coagulant at 60 ml/l dose. It also has been observed that the removal of TSS, TDS, BOD and COD efficiently by RC. CPCB [3] reported that some of the WTPs are cleaned once in a year and in the lack of sludge disposal management it disposed off on nearby open lands or drain. Proper handling WTS from WTPs in an economical and environmentally friendly manner remains a very important issue. The recovery, recycling and reuse may provide a sustainable solution to the WTS management.

Keywords: *Water Treatment, Sludge, Recovery of coagulant, MOSP, SEM, ED-XRD, FTIR, and XRD.*

Introduction

In the purification of surface water, coagulation-flocculation process is mostly used for the removal of turbidity and other impurities. A large quantity of water treatment sludge (WTS) is generated which is presently being disposed of drains or land. Such practices cause contamination of surface and groundwater resources. A sustainable sludge management, therefore, requires the possibility of minimizing sludge generation or whatever generated is reused, and recycled. Such strategy must be explored for recovery of chemicals as by- products from WTS which can be re-used in the treatment of water and wastewater. Increasing demand of milk and milk products has led to the growth of dairy industries in most of the countries of the world. India is also among the leading producers of milk and other dairy products in the world and contributes about 35% of the total Asian milk [1, 11]. It is generally characterized by high COD (80-95,000 mg/L) and BOD (40-48,000 mg/L), relatively large load of suspended solids (24-45,000 mg/L) and significant variation in pH (4.2-9.4) [7, 13]. The impurities in the form of colloidal suspensions in dairy wastewater are composed of inorganic and organic substances and contains at least one dimension lying within the range of ten angstrom to one micron undergo Brownian motion. Colloidal particles are negatively charged. They do not agglomerate its own. Hence, particles remain suspended for long periods of time without settling [14]. It can be treated by the coagulation-flocculation process. Many coagulants are widely used in water treatment, based on their chemical characteristics. Coagulants are classified mainly in inorganic, composite of inorganic, synthetic organic polymer and natural coagulants.

Aluminum salts namely alum and polyaluminum chlorides (PAC) are the most common synthetic chemical coagulants used. The efficiency of ferrous sulphate and alum as coagulant were studied in chemical treatment of dairy wastewater [8]. The removal of arsenite and arsenate removal using Ferric chloride as a coagulant and chitosan as a coagulant aid had from synthetic water was reported by [15]. Natural coagulants are economically and environmentally more acceptable than chemical coagulants. The seeds from Moringa Oleifera are found to be one of the most effective primary coagulants for water treatment. Moringa Oleifera is a tropical plant belonging to the family of Moringaceae [3].

It is used as coagulant on a small scale in developing countries for water purification. Moringa Oleifera is a potential alternative to chemical coagulants for water treatment in rural and urban areas [16]. In a report, optimum MO seed powder as coagulant was found to be 0.6gm/100ml and optimum time required for the reduction turbidity and optical density of absorbance and transmittance agitation was found to 1 hour [2].

The advantages observed beside its natural and wide availability, cost effectiveness, their high turbidity removal rate ranging from 89 - 99% and the alum saving by 40 - 60% is the other advantage [17]. The Moringa Oleifera has high coagulation activities for high turbidity water. The coagulation mechanism of the Moringa Oleifera coagulant protein has been described as adsorption, charge neutralization and inter particle bridging. It is mainly characteristic of high molecular weight polyelectrolyte.

The results showed that Moringa seeds were capable of adsorbing heavy metals tested in some water samples [12]. The maximum percentage of turbidity was found in oil free Moringa coagulants [10]. The composition of WTS states that oxide of silica was the major constituent. In the water treatment, mostly Al or Fe based has been found while physicochemical analysis, which contains heavy metals in trace.

In general, SiO_2 founds as a major portion of the sludge and followed by Al_2O_3 and Fe_2O_3. Other oxides such as CaO, MgO, Na_2O, K_2O, P_2O_5 and TiO_2 were found in small percentages. The present study was undertaken to (i) determine the physio-chemical characteristics of water treatment sludge, (ii) recovery of coagulant from WTS for reuse, and (iii) the compare removal efficiency of turbidity by the regenerated coagulant (RC) with natural coagulant at various pH (2-12) conditions. The *total dissolved solids (TDS), total suspended solids, BOD and COD* tests were also performed at neutral pH (7) of raw and supernatant of dairy wastewater using both coagulants.

Materials and Methods

In present the study, WTS was collected from the Delhi Jal Board's Chandrawal Water Treatment Plant located in west Delhi, which uses poly aluminum chloride (PAC) as coagulant. The characterization of WTS and MOSP has been investigated by using Energy-Dispersive X-Ray Fluorescence (ED-XRF). ED-XRF setup involving a low power (100 W) tungsten anode X-ray tube (Kevex, 50 kV, 2.0 mA, water cooled) as a source of excitation. The X-ray tube was operated at 35 kV and 1.7 mA.

Wastewater Preparation, Sludge Collection, and their Characteristics

For the preparation of Synthetic dairy wastewater (SDW), 7ml milk (full cream manufactured by Mother Dairy Fruits and Vegetable Ltd.) in one-liter distilled water and added one gram of kaolin ($H_2Al_2Si_2O$ $8H_2O$) (laboratory Grade) powder and mixed to get a uniform solution as per the standard methods. For the experiment works, SDW were prepared freshly whenever required to keep composition constant during the study. The SDW sample was analyzed for pH, colloidal suspension, removal in terms of turbidity, and other parameters as per the prescribed standard method of APHA.

Recovery of Coagulant from Water Treatment Sludge (WTS)

The WTS was acidified with the different normality of H_2SO_4 at 1.0 N to 4 N (interval of 0.5N) at a ratio of 0.02 ml H_2SO_4 /ml of sludge i.e. for the experimental purpose 100 ml sludge was mixed with 2ml H_2SO_4 of different normality.

A varying concentration of product in solution as regenerated coagulant (RC) are produced with varying volume from 5 ml to 30 ml and normality and used to treat 1 liter of SDW.

Preparation of Moringa Oleifera Seed Powder and Solution

Dry Moringa Oleifera seeds collected from Munirka, New Delhi. Seed shells were removed, and Sun dried for 48 hours. Hulls and wings from the kernels were removed manually. Kernals grounded in a domestic mixer grinder and sieved through 600 micrometer stainless sieve. The fine powder of Moringa seeds is stored in airtight container and kept in refrigerator to prevent the loss of its action. The optimum results were obtained by mixing of 6gm of Moringa Oleifera seeds powder in one liter of distilled water with the help of magnetic stirrer and filtered through the 20µm paper filter.

Experimental Methodology

Jar tests were carried out to simulate the conventional coagulation-flocculation process. Six beakers containing 1000 ml SDW sample were placed on a standard jar test apparatus. A flash/rapid mixing at 100 rpm for 2 minutes was provided after adding the coagulant dose, then the slow mixing was carried out at 20 rpm for 25 minutes. Thereafter, jars were kept standstill for 20 minutes to settle down the flocs. A series of jar tests were performed to determine the effect of coagulant nature, coagulant dose and initial pH on turbidity removal from SDW.

Removal of colloidal suspension from the SDW was investigated over a wide pH range of 2-12. Initial pH of SDW was maintained to the required level using H_2SO_4 or Noah before adding different coagulants. Batch experiments were carried out at each pH condition for variable dosage of RC and MOSP. The supernatant from each jar was withdrawn and analyzed for colloidal suspension removal in terms of turbidity through Nephelometric turbidity.

Results and Discussion

SDW Characteristics

SDW had slightly acidic pH of 6.5-6.8 and high COD value of 1250 mg/L (Table 1). Turbidity and TSS values were 372 NTU and 348 mg/L respectively, with a high TDS value of 1187 mg/L, thus total solids present in the SDW were about 1535 mg/L. Kushwaha *et al* [7].Reported higher optimum dose of different coagulants in their study. Therefore, to make the solution uniform and lower the required coagulant dose, kaolin was added while preparing the SDW. The addition of kaolin lowered the optimum coagulant dose in case of each coagulant applied in this study. It may due to adsorption of colloids onto the surface of kaolin particles.

Table 1: Characteristics of SDW

Parameter	Value	Unit
pH	6.5 - 6.8	-
Turbidity	372	NTU
BOD	454	mg/L
COD	1250	mg/L
TSS	348	mg/L
TDS	1187	mg/L

WTS Characteristics

Chemical composition of the WTS is shown in Table 2. SiO_2 (67.75%) and Al_2O_3 (16.76%) where the major oxides present in the WTS however, Fe_2O_3 was also found in small quantity. Since PACl is used at the water treatment plant, WTS had a higher percentage of aluminium in the dried mass of WTS. Therefore, conditioning of WTS would regenerate the aluminium and iron present in the sludge which could work as coagulant in the treatment of dairy wastewater. Some trace elements, including toxic substances shown in Table 2 were also found in the WTS as these metals are present in the raw water or present as impurities in the coagulants which get accumulated in the small mass of sludge [18]. Hence, proper handling of such waste is needed for sustainable development.

Table 2: Chemical composition and trace Elements of WTS

Composition	% W/W	Elements	ppm
SiO_2	67.75	Ni	21.97
Al_2O_3	16.76	Cu	38.31
Fe_2O_3	5.52	Zn	83.08
MgO	3.33	Ga	13.48
K_2O	3.03	As	18.00
CaO	2.37	Br	7.76
TiO_2	0.51	Rb	136.26
MnO	0.09	Sr	68.41
Cl	0.30	Y	22.50
		Zr	108.59
		Nb	10.76
		Ba	485.94
		W	147.67
		Pb	24.00

MOSP Characteristics

Chemical composition of the Moringa Oleifera seed powder is shown in Table 3. H_2O is the major component (95.95% W/W), however, Potassium (K, 1.05%W/W) and sulfur (S, 2.36%W/W) chemical. Some trace elements shown in Table 3 were also found in the WTS and trace of iron (Fe) and tungsten (W) are also detected (100 ppm) were also found in small quantity.

Table 3: Chemical Composition of MOSP

Composition	% W/W	Elements	ppm
H_2O	95.95	Mn	19.32
Mg	0.06	Fe	99.55
Ca	0.18	Cu	16.6
K	1.05	Zn	35.41
S	2.3	Rb	4.05
P	0.44	W	99.44

Recovery of Coagulant from WTS

Turbidity removal at a variable dosing of WTS acidified with the different normality of H_2SO_4 is shown in Fig. 1. It can be seen from Fig. 1 that, as the normality of H_2SO_4 was increased from 1 N to 3 N, turbidity removal was also increased, whereas it decreased when WTS was acidified with H_2SO_4 of normality above 3 N. Conditioning of WTS with acids releases the aluminum and iron metals from the sludge matrix which act as a coagulating agent. Qasim and Mane [11] recovered coagulants from WTS through the acidification process and applied in the wastewater treatment. The optimum normality of H_2SO_4 was found to be 3 N which gave highest turbidity removal at all the dosage of acidified WTS. Hence, the collected WTS was acidified with 3 N H_2SO_4 at a rate of 50 ml/L of sludge to RC which was used as a coagulant in the treatment of SDW at variable condition.

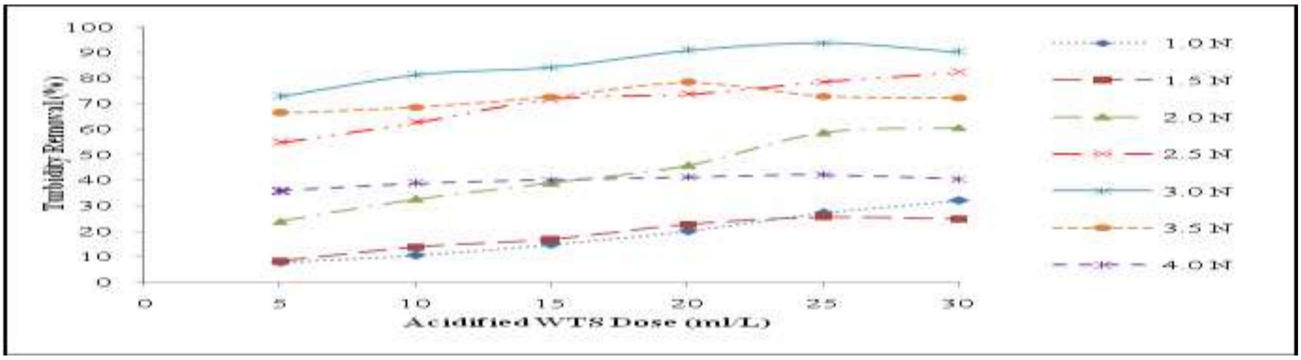

Fig. 1: Turbidity removal at variable dosage of acidified WTS

Effect of Initial pH and Coagulant Dose

Turbidity removal from SDW in the pH range of 2 to 12 at a variable dosing of RC and natural coagulant is shown in Fig. 2 and 3 respectively.

It could be observed from Fig. 2 that the effect of initial pH of the SDW and RC dose on the turbidity removal from SDW (Table 4). In case of regenerated coagulant (RC) also the turbidity removal increased with pH and had increased up to pH 7, after that it decreased rapidly as the RC dose was increased from 10 ml/L to 40 ml/L. At pH 5, 6 and 7 turbidity removal was increasing trends with RC dose and attained maximum removal at the dose of 25 ml/L and then decreased. RC was found to be more effective at neutral pH, gave maximum turbidity removal of 93.60% from SDW at the optimum dose of 25 mg/L. Fig. 3 present the turbidity removal from SDW at a variable dose of MOSP at different initial pH of SDW.

The turbidity removal increases with increase of pH and upto pH 7. It could be further inferred that the turbidity removal increased with MOSP dose and attained the maxima at about 60 ml/L dose in case of all pH conditions. Maximum turbidity removal of 94 % was achieved at the alum dose of 60 ml/L when the initial pH of SDW was kept at pH 7 (Table 5). In fact, the colloidal removal or the coagulation mechanism is determined by the interrelations existing between coagulant dose, pH and colloidal concentration. Formations of metallic cations are dominant at lower pH and favours colloidal removal through adsorption by charge neutralization. However, at higher pH and increasing the dose, formation of hydroxide precipitate leads to colloidal solids removal mainly by sweep floc mechanism and physical adsorption [19, 6, 11].The casein (milk protein) present in the dairy wastewater mainly imparts colloidal nature of the wastewater [3].

Selmer-Olsen *et al* [13].Reported that dairy wastewater has the isoelectric point (pH_{iso}) around 4.2. At pH > pH_{iso} milk proteins present in the dairy wastewater carries a negative charge and hence, can be removed through charge neutralization by positively charged coagulant species. RC was found efficient in removing colloidal suspensions from dairy wastewater having pH around 7 and the results were comparable with the efficiency of natural coagulants in this pH range.

Table 4: Turbidity Removal by RC with variable pH conditions

Dose	pH 2	pH 3	pH 4	pH 5	pH 6	pH 7	pH 8	pH 9	pH 10	pH 11	pH 12
5	18.57	21.05	68.42	59.43	71.95	72.70	62.35	65.88	70.68	73.54	67.24
10	16.86	19.39	65.14	61.43	80.51	81.30	56.82	75.28	69.84	82.58	79.67
15	14.86	17.45	55.43	62.29	83.48	84.20	43.25	65.37	51.25	66.33	60.63
20	12.86	15.51	46.86	63.43	86.04	90.70	32.18	54.24	39.42	44.56	42.15
25	10.00	12.74	42.00	65.71	88.07	**93.60**	24.69	20.43	19.68	18.35	17.25
30	8.57	11.36	37.43	62.00	85.28	90.10	22.14	18.21	17.34	15.36	14.22

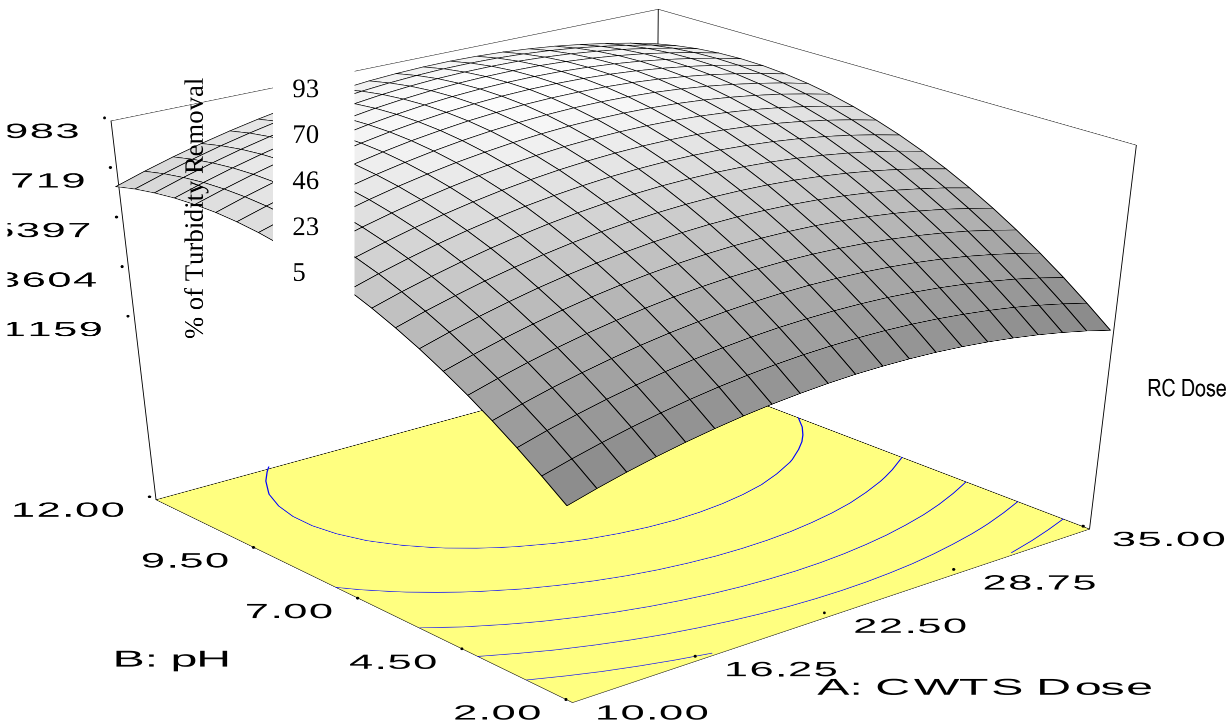

Fig. 2: Turbidity removal at variable dosage of Regenerated Coagulant

Table 5: Turbidity Removal by Natural coagulant with different pH conditions

Dose	pH 2	pH 3	pH 4	pH 5	pH 6	pH 7	pH 8	pH 9	pH 10	pH 11	pH 12
10	18.38	24.21	28.5	32.16	35.67	37.25	30.24	34.52	36.12	44.28	47.52
20	29.87	31.95	34.55	42.60	48.83	49.02	44.68	47.79	50.39	55.32	56.88
30	35.06	35.06	42.86	50.13	52.99	50.59	51.95	50.13	54.29	57.66	60.52
40	42.60	40.26	45.45	57.92	67.27	60.78	67.53	61.04	67.27	75.84	76.62
50	52.99	53.25	52.73	74.81	73.51	84.71	77.40	75.06	85.71	87.79	88.31
60	55.58	63.64	68.31	82.08	90.39	**94.12**	91.69	81.82	78.70	77.66	83.90
70	50.65	58.44	63.64	77.92	84.16	85.88	82.60	78.44	74.29	70.91	81.04
80	47.31	55.92	58.32	69.43	78.45	80.39	76.32	72.15	62.36	63.69	78.36

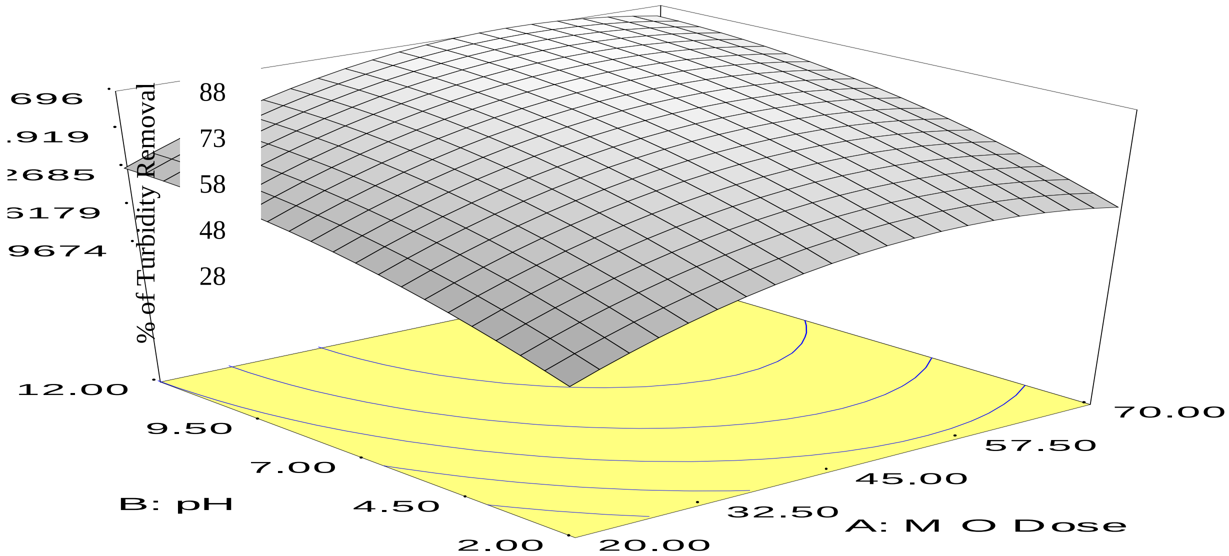

Fig. 3: Turbidity removal at variable dosage of acidified WTS

Quality of Treated SDW at Optimum Condition

During the coagulation-flocculation process, apart from turbidity other pollutants associated with the colloidal particles also get removed. Therefore, SDW obtained after the coagulation-flocculation process at the optimum conditions in case of both coagulants were analyzed for COD, BOD, TSS and TDS removals. COD, BOD, TSS and TDS removal physicochemical treatment are shown in Table 6.

RC had the highest COD removal percent (66%) as compared with MOSP (63%); BOD removal by RC was also comparable to the BOD removal by MOSP under investigation. Similar BOD removal of 68% was achieved with both coagulants TSS and TDS removals were also higher than that of conventional coagulants. TSS and TDS removal from the SDW were 85% and 86% respectively at the optimum condition of pH 7 and 25 ml/L RC [9]. However, the TSS and TDS removals were 81% and 81% respectively for MOSP. Unlike other pollutants, TDS removal efficiency seems to be unexpectedly high in case of all the coagulants. Such higher removal efficiency values were observed may be due to the constraint of gravimetric method used to determine the TDS removal. The cation-anions along with the ultrafine flocs were removed in the filter paper giving higher TDS removal efficiency in all the cases. However, it was a comparative study and except for BOD, RC was found to perform even better than the natural coagulants at removing COD, TSS and TDS from SDW.

It is evident from the results that the RC is efficient in removing pollution load from the dairy wastewater. Hence, from the above discussion, it can be inferred that WTS can be applied in the primary treatment of dairy wastewater and could substitute the conventional coagulants partially or fully in the dairy waste water treatment plants. WTS application would provide safe and sustainable sludge disposal option. At the same time, it could provide cost effective physicochemical treatment to dairy wastewater by reducing the requirements of chemical coagulants [5].

Table 6: Removal of pollutants from SDW at the optimum condition

Coagulant	Pollutant Removal (%)					
	Turbidity	COD	BOD	TSS	TDS	Optimum Condition
Regenerated Coagulant	94	66	68	85	86	pH 7, 25ml/L dose
MOSP	94	63	68	81	81	pH 7, 60 ml/L dose

Conclusions

The current practice of sludge disposal poses danger to the environment and community health. The reuse of WTS offers both economic and environmental sustainability. Prior characterizations of sludge in terms of its chemical properties are necessary for better reuse and recycle as safe disposal alternatives. WTS application in water /wastewater treatment will provide some significant chemical savings through resource recovery, re-use, and sludge volume reduction.

Currently, the developing and many developed countries have introduced specific legislation related to the sound management WTS with a focus on waste reduction and reuses guidelines. Ministry of Environment, New Delhi emphasized the need of waste minimization-waste reuse and recycling in the policy Statement for Abatement of pollution, 1992 and in E (P) Act 1986. RC prepared from WTS acidified with 3 N H_2SO_4 at a rate of 50 ml/L sludge performed even better than the MOSP for the treatment of SDW. Maximum turbidity removal of 93.60% was achieved at pH 7 at an RC dose of 25 ml/L. Along with the turbidity, pollutants associated with the colloidal particles, also get removed. Significant removal of COD (66%), BOD (68%), TSS (85%) and TDS (86%) from the SDW was achieved at the optimum condition. Removal of all the pollutants from SDW by RC was comparable to MOSP.

Moringa Oleifera is a natural product, it is safe to the environment for disposal and non-toxic will be an alternative to conventional and expensive coagulants. Hence, it can be concluded from the present study that RC and natural coagulant could be potentially utilized to treat the dairy wastewater. Recycling of waste from water treatment plants into dairy wastewater treatment plants would provide constructive utilization and sustainable disposal of WTS, at the same time providing cost effective physicochemical treatment of dairy wastewater [20].

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