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Removal of fluoride by using *Passiflora Foetida* fruits as natural coagulant

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ABSTRACT

There is increasing interest in developing low cost water and wastewater treatment processes particularly in poor countries. The use of natural coagulants such as the *Passiflora foetida* fruits to replace expensive imported synthetic coagulants is particularly appropriate for agro-based developing countries such as India, because local production of these coagulants will also contribute to the rural and national economy. There is also widespread recognition that the presence of toxic chemicals. This study was conducted to investigate the potential of *Passiflora foetida* fruits for treatment of wastewater contaminated with fluoride. The potential of *Passiflora foetida* whole fruit, in removing fluoride ions from synthetic contaminated water was investigated at initial fluoride ion concentrations of 1 mg to 8 mg/L by means of jar tests. Fluoride ion removal was observed ranging from 80.86 ± 2.22% to 96.40 ± 0.00% at optimum dosage of 1gm/L. Earlier work by the same group has shown that *Citrus limon* peel powder as natural coagulant to reduce wastewater fluoride by over 95% and reduce turbidity at optimum dosage of 1gm/L. The present study also carried out with coagulation parameters such as initial fluoride ion concentration, coagulant dosage and impact of water pH on removal of fluoride by *Passiflora foetida* fruits. The results concluded that removal of fluoride was high at lower fluoride concentration with constant coagulant dosage. The fluoride removal efficiency of *Passiflora foetida* fruits is high at acidic and neutral pH medium and gradually decreased with increase base/alkalinity of medium. The results obtained in present study tested with coagulation kinetic parameters and observed that removal of fluoride by *Passiflora foetida* fruits is following pseudo second order kinetic model at lower fluoride concentration.

Keywords: Plant based coagulant, Fluoride removal, Coagulation, *Passiflora foetida* fruits, coagulation kinetic models.

1. INTRODUCTION

The industrial waste water from more than a few industries like electroplating, semiconductor, glass and oil refineries etc. contains a very high concentration of different organic and inorganic chemicals. These toxic chemical compounds are very hazardous and have adverse effect on each the aquatic lifestyles as well as the terrestrial life. Fluorides are the hazardous inorganic pollutant widely located in underground water and industrial waste water. Although the

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low concentration of fluorides are healthy and safe but beyond the permissible limits its consumption is very dangerous. The permissible vary of fluoride concentration in drinking water has been set as 0.5-1.5 mg/l by using many organizations like Central Pollution Control Board, World Health Organization and United States of European Public health Authority etc. In India, around one million human beings are affected by indigenous fluorosis [1, 2].

Fluoride is extensively distributed on earth in many forms like fluoride minerals, in underground water, foods and tea leaves [3]. Fluoride dissolved into the groundwater due to the presence of fluoride minerals/rocks like fluorite, cryolite, phosphorite, fluorapatite, theorapatite etc. at the aquifer bottom [4]. Further, a number industrial techniques such as steel production, glass manufacturing, electroplating, phosphatic fertilizer production, ceramic industry and coal combustion etc. extensively contributed in increasing the fluoride contamination stage in water. Thus the treatment of waste water is necessary before its discharge. At present, many technologies are handy for the removal of fluoride from water such as co-precipitation, coagulation, ion exchange, adsorption, dialysis, electro-dialysis, reverse osmosis, membrane technologies etc. [5-11].

All these technologies have their indigenous benefits and disadvantages such as secondary sludge production, less efficiency and especially sensitized. In these methods coagulation is a most simple and attractive process due to its low price and excessive efficiency. But now a day, plant based coagulants and its strategies are determined to be extra easy and environment friendly due to their sludge free operation and regeneration potential [12]. Coagulation-flocculation techniques contain quite a few without difficulty available plant based components like Fruits of *Opuntia Ficus indica*, Fruits of *Jatropha gossypifolia*, peel of *Borassus flabellifier* for removal of chromium [13] *Citus limon* peel powder [14], *Moringa oleifera* extract [15] clearing nuts [16], *Pithecellobium Ducle* Seeds [17] and latex exudates from *Ipomea Carnea* and *Calotropis giganteana* [18] etc. for the removal of poisonous fluoride ions from waste water. In most of the available literatures on the fluoride elimination using various plant based natural coagulants, the fluoride concentration is between 1.5-5 mg/l available in groundwater at normal conditions. However in industrial waste water the fluoride awareness is higher [19] and its removal is turn out to be necessary to stop its adverse effects on environment. Therefore, in this paper the fluoride elimination possible of *Passiflora foetida* fruits has been investigated. Here, the effect of more than a few parameters on the share removal of fluoride has been studied and the mode of action, removal nature has been also determined in phrases of kinetic models.

Plant-Based Coagulants (PBC)

PBC coagulants are required to treat water showing up at medium turbidity go (50– 500) NTU. It is unexpected that an absolute conclusive examination of existing PBC is so far nonexistent in this 21st century. The importance of PBC to natural framework finally regard a minute zone for researcher in fortifying the examination to find basic resources. Among PBC portrayed specifically, Peanut Seeds (*Pisum sativum*), powder and Cactus. Anionic polyelectrolytes are polymers of *Aloe vera* whose particles get obscured in water by through bury atom traverse. The seed removes contains – COOH and – OH clusters that are competent augmentation the coagulation competency since lipids, sugars and alkaloids. Galactomannani and galactanii are mix of polysaccharide division that are removed from *Strychnos potatorum* seeds which is capable in reducing turbidity up to 80%. In all perspectives, the galactomannans are set up 1,4-associated β-dmannopyranosyli residuum character end α-d-galactopyranosyl units related with 0–6 point of some mannerresidue. Shelled nut Seeds , Peanut Seeds powder and its related coagulation process yet not been inspected, the proximity of limitless proportion of – OH packs along chains of galactomannan and galactan gives sadly anyway ample adsorption that over the long haul lead to the past coagulant interparticle interfacing sway more research should be possible in this edges [20].

Cost of Plant-Based Coagulants

It has been elucidated in past portions that utilization of plant based coagulants gives biological points of interest and different lab-scale mulls over have shown that they are in certainty feasible for little scale point of use. Regardless, in regards to commercialization, in particular it will reliably be established in a general sense on whether the scale-up system can proceed equivalent treatment execution at for all intents and purposes indistinguishable (or diminished) cost with the normal coagulants when differentiated and set up compound coagulants. There are two or three described reports that give the costs of unrefined materials of the coagulants anyway facilitate connections to the extent coagulant types, Processing stages and expenses in different geological regions are an outstandingly confounded endeavor given the particular exchange rates, extension factor and fluctuating exactnesses of the costing regards. An intensive audit drove reveals that costing examination of Peanut Seeds, Peanut Seeds powder has been given need over other regular coagulants and this is evident given the especially progressed good conditions of the plant [20].

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Aims & Objectives

The fundamental point of the present examination was to assess the coagulation limit of chosen plant based coagulant *Passiflora foetida* fruits for the removal of fluoride in coagulation process. So as to accomplish these accompanying targets are set and are examined.

1. Assurance of impacts of various exploratory parameters like, contact time, initial ion concentration, coagulant dosage and pH of the solution on coagulation and flocculation system
2. Assurance of coagulation kinetic parameters, utilizing model equations accessible in writing on coagulation process.
3. Assessment of fluoride expulsion limit of *Passiflora foetida* fruits from industrial waste water.

2. MATERIALS & METHODS

Selection of Coagulant for removal of fluoride

The high amounts of industrial effluents being released into a few water assets, with no pre treatment comprise of high convergence of fluoride [21]. Frequently in abundance of poison focus can't be evacuated by straightforward essential treatment and can't be expelled 100 % by other ease strategies. This high level of toxins evacuation can frequently be accomplished by chemical coagulation process as it were. In this way, huge numbers of the specialists gave an account of electro-coagulative expulsion of chromium from various industrial wastewaters [22 - 26] and removal of fluoride by salts of Al, Fe, Mg from drinking water [27-28]. These synthetic compounds utilized in coagulation studies may accomplish their objectives yet the examinations inferred that the utilization of concoction coagulants may changes the other water quality parameters. A few examinations inferred that the overabundance measure of Al salt adequately expelling fluoride from drinking water yet it is changes the nature of water by expanding electro conductivity and saltiness of the treated water [29-31]. As indicated by WHO, high electro conductivity and saltiness containing water does not suits for drinking and other use. A few methodologies have been accounted for to maintain a strategic distance from the synthetic precipitation. A less information is accessible on utilization of modest, ease, compelling and natural coagulants, for the expulsion of fluoride from aqueous solutions. The benefits of the normal coagulants over the synthetic and electro coagulants are as per the following.

- ✓ Eco items got from plants and inexhaustible sources, adding to a maintainable and affordable water treatment.
- ✓ Plant based coagulants decline the volume of slime and don't modify the pH of the water under treatment process.
- ✓ Non poisonous and non destructive eco items, subsequently contributes for long time maintenances of types of instruments used for treatment. Application on an extensive pH extends (4 to 9), without adjustment of the gushing's pH.
- ✓ These plant based eco friendly coagulants impressively lessen the utilization of acidic and antacid operators bringing about tremendous sparing of synthetic compounds.
- ✓ The effluents conductivity stays unaltered. This is vital if there should be an occurrence of assimilation process and in the event of shut circuit waters.

The material created for this reason to expel fluoride from industrial waste to manufactured wastewater, biomass and different suspended solids. In this examination *Passiflora foetida* fruits were used for removal of fluoride from industrial wastewater.

Passiflora foetida

Kingdom : Plantae – Plants
Subkingdom : Tracheobionta – Vascular plants
Superdivision : Spermatophyta – Seed plants
Division : Magnoliophyta – Flowering plants
Class : Magnoliopsida – Dicotyledons
Subclass : Dilleniidae
Order : Violales
Family : Passifloraceae – Passion-flower family
Genus : *Passiflora* L. – passionflower
Species : *Passiflora foetida* L. – fetid passion flower

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Figure 1 *Passiflora foetida* fruits

Passiflora foetida belongs to passifloraceae. They are mostly vines and some are shrubs and some are herbaceous. The *Passiflora foetida* fruits are round or elongated edible fruits (Figure-1). It consists Beta carotene, vitamins, dietary fibre and iron in significant quantities. Due to high nutrient quality, government of Andhra Pradesh started growing fruits in chintapalli forest in Vishakapatnam and they found widely in forest of Assam and Niligiri. From the literature it is concluded that *Passiflora foetida* fruits were rich in phosphorus and iron. The fluoride present in aqueous solution may react with these two minerals and forms FeF_3 and PF_3 as stable compounds. Taking that factor into consideration these fruits were tested for removal of fluoride from aqueous solution by coagulation process.

Preparation of coagulant

The fruits of *Passiflora foetida* collected from local fields of Nalgonda district of Telangana state and fruits were cleaned with running tap water allowed for dry under room temperature for 15 minutes. The air dried fruits were grounded into fine paste by using a blender and the fresh *Passiflora foetida* fruits paste used as natural coagulant for removal of fluoride from industrial waste water.

Experimental Method

Preparation of fluoride standard curve

Into a series of 50 mL stand flask different fluoride solutions were taken and to this solution 5 mL of SPADNS reagent was added and the final volume was made up to the mark using distilled water. The Optical Density (O.D.) values were taken at 570 nm using spectrophotometer [32-40].

Determination of Optimum Coagulant Dosage

Assurance of ideal coagulant measurement Jar test is generally utilized procedure for assessing and advancing coagulation-flocculation process. This investigation comprises of cluster tests including rapid mixing (to scatter coagulant portion) and moderate blending (to improve floc arrangement and sedimentation for example settling of framed floc). In the present investigation all trials were performed by utilizing jar apparatus with aqueous solution of fluoride and distinctive dose of coagulant, are included into a progression of 6 jars situated on jar aparatus. The fluoride aqueous solutions are stirred rapidly for 20 minutes at standard 100 rpm speed pursued by 30 minutes moderate mixing at standard 40 rpm speed for the arrangement of flocs. At long last the flocs were permitted to agree to 40 minutes before pulling back the samples for investigation [41– 42]. In the present study all experiments were performed by using jar apparatus with 4 mg/L fluoride solution and different dosage of coagulant (0.2, 0.5, 1.0, 1.5, 2.0 and 3.0 gm) are added into a series of 6 jars positioned on jar apparatus. The samples are stirred rapidly for 20 min. at standard 100 rpm speed followed by 30 min. slow stirring at standard 40 rpm speed for the formation of flocs. The final fluoride concentration after adsorption was directly measured with spectrometer by using SPADNS dye method. To estimate the percentage removal of fluoride from aqueous solution the following equation was used.

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$$\% \, Removal \, of \, Fluoride = \frac{C_{initial} - C_{final}}{C_{initial}} \, X \, 100$$

Effect of Initial Fluoride Concentration

To discover the impact of fluoride concentration on coagulant, the tests are done by taking distinctive concentrations of fluoride solutions (extending from 1 mg/L- 8 mg/L) into a arrangement of measuring jars situated on jar apparatus. To the all different concentrations of solutions containers 1.0 gm of settled measure of coagulant is included. At that point the tests are mixed quickly for 20 min. at standard 100 rpm speed pursued by 30 min. moderate mixing at standard 40 rpm speed for the development of flocs. At last the flocs were permitted to make due with 40 min. previously pulling back the samples for examination.

Effect of pH on Coagulation Process

To check out the optimum pH for the removal of fluoride the tests were carried out at different pH values. In this study the coagulation were examined at the following pH values i.e. 3, 4, 5, 6, 7, 9, 10 and 11. The experiments are carried out by taking constant and fixed amount of fluoride and coagulant dosage i.e. 4 mg/L of fluoride and 1.0 gm of coagulant in all beakers then the samples are stirred rapidly for 20 min. at standard 100 rpm speed followed by 30 min. slow stirring at standard 40 rpm speed for the formation of flocs.

All the reactive and chemical substances used in this investigation were analytical grade and double distilled water used to be used in all the experiments to put together working solutions. As a precautionary exercise standard procedures were followed for sample dealing with and series [43]. All cleaned glassware were soaked in 10% HNO_3 in a single day for fluoride evaluation and washed with distilled and deionized water before they had been used. The evaluation of the samples were carried out quickly after series and stored in a fridge for further analysis. The calibration standards of metals have been prepared in accordance to the Standard Methods for Examination Water and Wastewater evaluation [43].

Coag-Flocculation Kinetics and Functional Parameters Response

Coag-flocculation is a core purification process, which finds wide range of application in water and waste water treatment facilities. Conceptually, Coag-flocculation is the process of adding substances to aqueous effluent to make suspended particles to bind together (coagulate) and subsequently aggregating into visible flocs (flocculation) that settle out of the water. This is achieved when the stabilized particles are aided to overcome their repulsive forces to form blobs of flocs [44 - 48]. In particular, coag-flocculation in conjunction with other treatment processes is regarded as a viable option for the treatment of aqueous effluent. Coag-flocculation can be achieved by any of the common coagulants such as alum, lime etc. The Coag-flocculation behaviors of these common compounds have been well investigated with little or no attention given to the Coag-flocculation potentials of bio derivatives. To this end, a focus is hereby given to the study on selected plant based coagulants, as a potential source of Coag-flocculation derivative for the removal of chromium and fluoride.

Theoretical Principles and Model Development for coagulation kinetics [49-52]

$$\mu_i = G_i = \left[\frac{dG}{dn_i}\right] P.T.n = a \, constant \, \ldots\ldots\ldots .1$$

Thus

$$d\mu_i = 0 \ldots\ldots\ldots . 2$$

For each of the species *I* present

G is the total Gibbs free energy

n_i is the number of moles of component (i), for dilute solutions

$$\mu_i \approx \mu_i + RT \ln C_i \ldots\ldots\ldots .3$$

A Shift from the equilibrium generates diffusional process represented by

$$f_d = \frac{d_\mu}{d_x} \ldots\ldots\ldots .4$$

Recall K_B = R/N such that for sing N, K_B = R. Hence

$$\mu_i \approx \mu_i^0 + K_B T \ln C_i \ldots\ldots\ldots .5$$

Where KB is Boltzmann Constant

μ_i is Chemical Potential

R is Universal gas Constant

C_i is Concentration

N is Avogadro's Constant

x is diffusion distance

Combining equation 4 and 5 yield

$$f_d = \frac{d}{dx}(\mu_i^0 + K_B T \ln C_i \dots\dots\dots 6$$

$$f_d = \frac{K_B}{C_i}\frac{dC_i}{dx} \dots\dots\dots 7$$

The viscous drag force on the particles due to surrounding fluid is

$f_d = BU_d$ ……….8 and

$J_i = CU_d \dots\dots\dots 9$

Where B is friction factor

U_d is terminal diffusing velocity

J_i is flux of diffusing material

f_d is drag force

From Fick's law

$$D' = \frac{-J}{({}^{d_c}/_{d_x})} \dots\dots\dots 10$$

Where D' is diffusion co-efficient. Combining equations 8, 9 & 10 yield

$$D' = \frac{-f_d}{B}\frac{c}{({}^{d_c}/_{d_x})} \dots\dots\dots 11$$

Comparing equations 7 and 11 generate Einstein's equation

$$D' = \frac{k_B T}{B} \dots\dots\dots 12$$

For similar phase, the rate of successful collisions between particles of sizes *I* and *j* to form particle of size *k* is

$$N_{i\,j} = \varepsilon_p \, \beta\,(i,j) n_i n_j \dots\dots\dots 13$$

Where $N_{i\,j}$ is the rate of collision between particles of size I and j (mass concentration/time)

ε_p is collision efficiency

$\beta\,(i,j)$ is collision factor between particles of size *I* and *j*.

$n_i n_j$ is particle concentration for particles of size *I* and *j* respectively.

Assuming mono disperse, no break up and bi particle collision, the general model peri kinetic coag – flocculation is given as

$$\frac{dn_k}{d_t} = \frac{1}{2}\sum_{i+j=k}\beta\,(i,j)n_i n_j - \sum_{i=1}^{\infty}\beta\,(i,k)n_i n_k \dots\dots\dots 14$$

Where $\frac{dn_k}{d_t}$ is the rate of change of concentration of particle size k (concentration/time)

β is function of coag- flocculation transport mechanism. The appropriate value of β for Brownian transport is given by

$$\beta_{BR} = \frac{8}{3}\varepsilon_p \frac{k_B T}{n} \dots\dots\dots 15$$

Where K_B is Boltzmann's constant (J/K)

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T is Absolute temperature (k)

The generic aggregation rate of particles (during coagulation/ flocculation) can be derived by the combination of equation 2 and 3 to yield

$$-\frac{dN_t}{d_t} = KN_t^{\propto} \ldots\ldots\ldots.16$$

Where Nt is total particle concentration at time t

K is the α^{th} order coag – flocculation constant

A is the order of coag – flocculation process.

And $K = \frac{1}{2}\beta_{BR} \ldots\ldots\ldots.17$

Where β_{BR} is collision factor for Brownian transport, also

$$\beta_{BR} = \varepsilon_p K_R \ldots\ldots\ldots.18$$

Combining equations 4. 5 and 6 produce

$$-\frac{dN_t}{d_t} = \frac{1}{2}\beta_{BR} N_t^{\propto} \ldots\ldots\ldots.19$$

$$= \frac{1}{2}\varepsilon_p K_R N_t^{\propto} \ldots\ldots\ldots.20$$

Where K_R is the Von Smoluchowski rate constant for rapid coagulation.

However

$$K_R = 8\pi\, R\, D' \ldots\ldots\ldots.21$$

$$R_p = 2a \ldots\ldots\ldots.22$$

Where D' is particle diffusion coefficient

A is particle radius.

From Einstein's equation

$$D' = {K_B T}/{B} \ldots\ldots\ldots.23$$

From stroke's equation

$$B = 6\pi n a \ldots\ldots\ldots.24$$

Where B is the friction factor and n is viscosity of the fluid.

Combining equation 21 to 24 gives

$$-\frac{dN_t}{d_t} = \frac{4}{3}\varepsilon_p \frac{K_B T}{n} N_t^{\propto} \ldots\ldots\ldots.25$$

Comparing equations 16 and 25

$$K = \frac{4}{3}\varepsilon_p \frac{K_B T}{n} \ldots\ldots\ldots.26$$

For peri kinetic aggregation, α theoretically equals 2 as would shown below

From Fick's law

$$J_f = D' 4\pi R_p^2 \frac{dN_t}{d_R} \ldots\ldots\ldots.27$$

Integrating equation 27 at initial conditions $N_t = 0$, R = 2a

$\frac{J_f}{D' 4\pi}\int_0^{R_p}\frac{\delta R_p}{R_p^2} = \int_{N_0}^{N_t}\delta N_t \ldots\ldots\ldots.28$. There fore

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$J_f = 8\pi D' aN_0 \ldots\ldots\ldots.29$

$= \frac{1}{2} K_R N_0 \ldots\ldots\ldots.30$

For central particle of same size undergoing Brownian motion, the initial rate of rapid coag – flocculation is

$-\frac{dN_t}{d_t} = J_f \varepsilon_p N_0 \ldots\ldots\ldots31$

$\frac{1}{2} = K_R \varepsilon_p N_0^2$

$= \frac{4}{3} \varepsilon_p \frac{K_B T}{n} N_0^2 \ldots\ldots\ldots.32$

$= \frac{4}{3} \varepsilon_p \frac{K_B T}{n} N_t^2 at\ t > 0$

Hence, from equation 32, α =2. However in real practice, empirical evidence shows that in general $1 \leq \alpha \leq 2$. Based on this, what is required to evaluate 'K' is to determine the line of better fit between α = 1 and 2, while the experimental data are fitted into linearised form of equation. Hence for α = 1, equivalence of equation 16, yields

$\frac{dN}{dt} = -kN \ldots\ldots\ldots.33$

Integrating within the limits produces

$\int_{N_0}^{N} \frac{dN}{dt} = -\int_0^t K dt \ldots\ldots\ldots34$. Hence

$\ln(\frac{1}{N}) = k_t - \ln N_0 \ldots\ldots\ldots35.$

Plot ln (1/N) vs t gives a slop of K and intercept of (-ln N_0).

For α = 2, equivalence of equation 16 yields

$\frac{dN}{dt} = -kN^2 \ldots\ldots\ldots.36$

$\int_{N_0}^{N} \frac{dN}{dt} = -k \int_0^t dt \ldots\ldots\ldots.37$

$\frac{1}{N} = k_t + \frac{1}{N_0} \ldots\ldots\ldots.38$

Plot ln (1/N) vs t gives a slop of K and intercept of $1/N_0$

For the evaluation of coagulation period ($\tau_{1/2}$), from equation 38

$N = \frac{N_0}{1 + N_0 k_t} \ldots\ldots\ldots.39$

$= \frac{N_0}{1 + \frac{t}{(1/N_0 K)}} \ldots\ldots\ldots.40$

Where

$\tau = \left(1/N_0\, K\right)$

$N = \frac{N_0}{1 + (t/\tau)} \ldots\ldots\ldots.41$

When t = τ the equation 41 becomes

$N = \frac{N_0}{2} \ldots\ldots\ldots.42$

Therefore as N0 → 0.5 ; $\tau = \tau_{1/2}$

$\tau_{1/2} = \frac{1}{0.5\, N_0 K} \ldots\ldots\ldots.43$ for second order and

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$\tau_{1/2} = \frac{\ln 2}{K}$ ……….44 for first order

3. RESULTS

Determination of optimum coagulant dosage

Effect of *Passiflora foetida* dosage on coagulation process was shown in Fig-2. From the figure it is observed that the removal efficiency increased with increase in coagulant dosage. When the removal efficiency reached the summit, the increase in coagulant dosage did not increase but remained stable, it indicates the mechanism of inter particle bridging adsorption on the active sites of colloids and destabilisation of colloidal particles to form large settable particles. If the dosage of coagulant is increased beyond the optimum dosage the active sites were occupied and wrapped up by the *Passiflora foetida* so that destabilisation of colloids takes place.

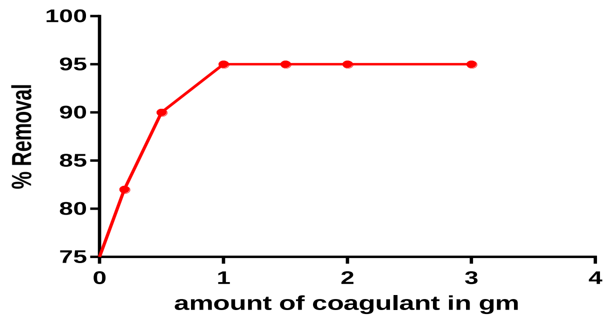

Figure 2 Determination of optimum coagulant dosage for removal of fluoride using *Passiflora foetida* fruits

Effect of Initial Fluoride Concentration

Concentration effect on coagulation of fluoride experiments were done by using different initial fluoride concentrations (1 to 8 mg/L), with optimum dosage of coagulant (1 g/L). From the Fig-3 it is concluded that the percentage removal of fluoride is increased with increase in initial fluoride concentration.

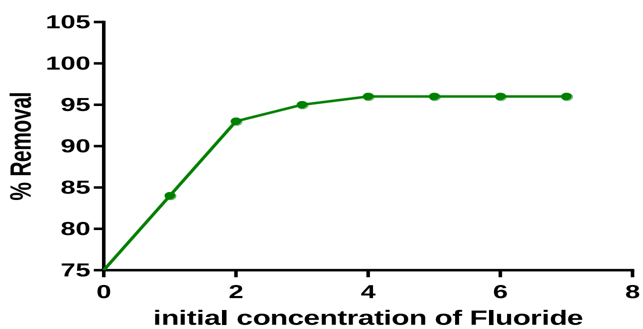

Figure 3 Effect of initial Fluoride ion concentration on *Passiflora foetida* fruits

Effect of pH on Coagulation Process

pH effect on coagulation of fluoride by *Passiflora foetida* was done by taking 4 mg/L fluoride solution and optimum coagulant dosage of 1 g/L. The pH of the solutions was maintained in the range of 3-11. From the Fig-4 it is observed that as the pH of the solution increased, the percentage removal of fluoride is decreased. The highest percentage removal of fluoride was observed to occur at pH 4 to 5 with 95 % removal. From the investigations, it is concluded that the pH of the aqueous solution may alter the overall percentage removal of fluoride and observations indicate better percentage removal under acidic conditions. As pH of the solution is increased the zeta potential may have decreased. This will results in the increase in surface zeta potentials, which will enhance the negative charges. At pH 4 and 5 the surface charge was neutralised and resulted in high fluoride removal.

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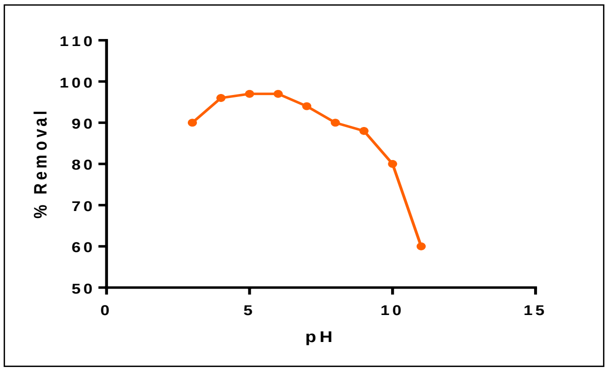

Figure 4 Effect of pH on removal of fluoride using *Passiflora foetida* fruits

Suggested Mechanism of action

The suggested mechanism of coagulation property of *Passiflora foetida* is due to the presence of iron, potassium and mucilage protein present in the fruits. The iron and potassium bind with fluoride molecules, the positive charge of proteins present in fruits will bind with the complex through electrostatic interaction. This leads to development of negatively charged and positively charged areas on the particle surface. The formation of neutralised complex leads to formation of flocs.

Efficiency (%) versus time (T) plots

The effect of contact time on removal of fluoride by *Passiflora foetida* was shown in Fig-5. From the figure it is observed that the percentage removal of fluoride increased with increase in contact time at all coagulant dosage. The same experiment conducted at a constant coagulant dosage by varying intital fluoride concentration from 1 -4 mg/L and the results were shown in Fig-6. From the figure it is observed that the percentage removal of fluoride is increased with increase in contact time at all different intial fluoride concentrations. Form the figure it can be concluded that the percentage removal is high at higer concentrations compare to lower fluoride concentrations. A better efficiency was observed at coagulant dosage 1.0 g to 3.0 g. Note that starting from *t=10* min, there is virtually no variation in E (%) values at 1. gm to 3.0 g with the least E>90 %, it confirms the effectiveness of *Passiflora foetida* to remove fluoride from the aqueous solution. At E (%)> 90 %, it justifies the theory of fast coagulation which validates the real life application of coag-flocculation.

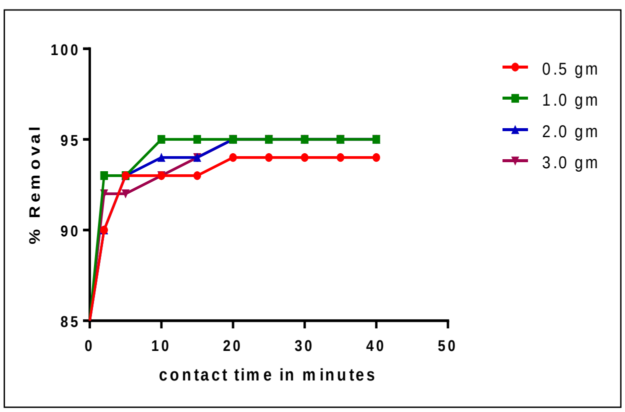

Figure 5 Effect of contact time on % removal of fluoride at different *Passiflora foetida* dosage

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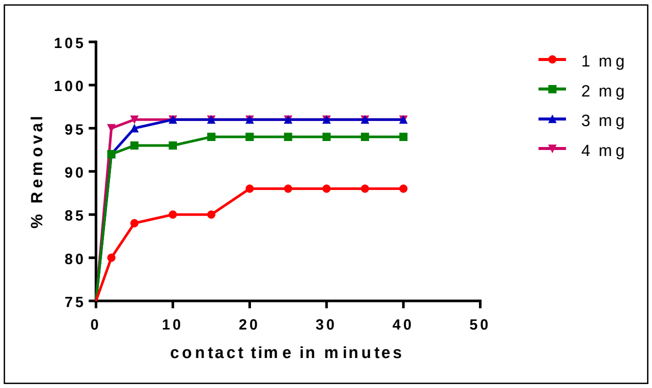

Figure 6 Effect of contact time on % removal of fluoride at different initial fluoride concentration at optimum *Passiflora foetida* dosage

Coagulation Kinetics

First Order Kinetic Model

The plot *log (a-x)* versus *time* representing the first order kinetic model for removal of fluoride by *Passiflora foetida* at different coagulant dosage and at different initial fluoride concentrations, are shown in Fig-7 & 8. From the figures it is observed that the lines were deviated from the linearity The correlation coefficient (R^2) values are in between 0.378 to 0.729 at different coagulant dosage and 0.262 to 0.706 at different initial fluoride concentrations, which proving that the removal of fluoride from aqueous solution using *Passiflora foetida* by coagulation method is not applicable to first order kinetic model. The calculated constant values for first order kinetic model were shown in table-1 & 2.

Table 1 Coag-floculation functional parameters for varying dosage at constant fluoride concentration

Parameter	0.5 gm/L	1.0 gm/L	2.0 gm/L	3.0 gm/L
	First Order Kinetic Model			
R^2	0.378	0.505	0.482	0.729
K	2.0×10^4	1.0×10^4	3.0×10^4	1.0×10^4
β_{BR}	4.0×10^4	2.0×10^4	6.0×10^4	2.0×10^4
ε_P	2.034×10^{20}	1.017×10^{20}	3.052×10^{20}	1.017×10^{20}
$\tau_{1/2}$	3465	6931	2310	6931
K_R	1.966×10^{24}	1.966×10^{24}	1.966×10^{24}	1.966×10^{24}
	Second order Kinetic Model			
R^2	0.425	0.505	0.566	0.746
K	3.83×10^2	3.00×10^2	5.80×10^2	5.29×10^2
β_{BR}	7.66×10^2	6.00×10^2	11.6×10^2	10.58×10^2
ε_P	3.896×10^{18}	3.052×10^{18}	5.900×10^{18}	5.381×10^{18}
$\tau_{1/2}$	18.09	23.10	11.95	13.10
K_R	1.966×10^{20}	1.966×10^{20}	1.966×10^{20}	1.966×10^{20}

Table 2 Coag-floculation functional parameters for varying fluoride concentration at constant dosage of coagulant

Parameter	1 mg/L	2 mg/L	3 mg/L	4 mg/L
	First Order Kinetic Model			
R^2	0.706	0.637	0.308	0.262
K	9.0×10^4	5.85×10^2	1.0×10^4	8.6×10^3
β_{BR}	18.0×10^4	11.7×10^2	2.0×10^4	17.2×10^3
ε_P	9.156×10^{20}	5.951×10^{20}	1.017×10^{20}	8.749×10^{20}
$\tau_{1/2}$	770.16	11.84	6931	80.59
K_R	1.965×10^{24}	1.966×10^{24}	1.867×10^{24}	1.965×10^{24}
	Second order Kinetic Model			

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R^2	0.785	0.590	0.316	0.262
K	6.60 x10^2	1.34 x10^2	2.63 x10^2	1.26 x10^2
β_{BR}	13.2 x10^2	2.68 x10^2	5.26 x10^2	2.52 x10^2
ε_P	6.714 x10^{18}	1.363 x10^{18}	2.675 x10^{18}	1.281 x10^{18}
$\tau_{1/2}$	10.50	51.72	26.35	55.011

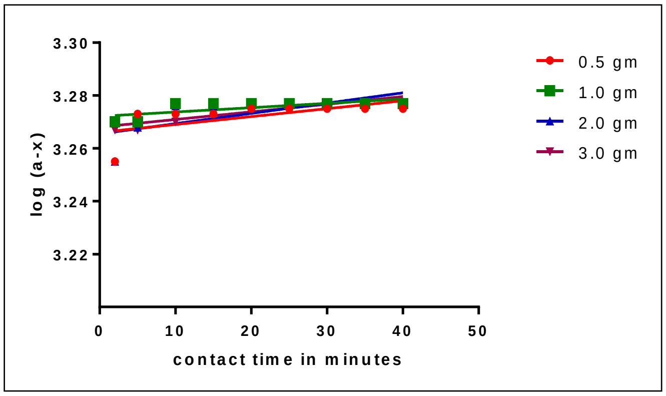

Figure 7 First Order kinetic model at different *Passiflora foetida* dosage

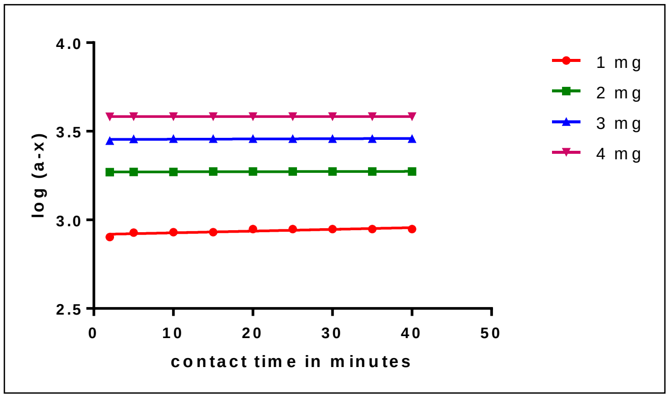

Figure 8 First Order kinetic model at different fluoride concentrations using *Passiflora foetida*

Second Order Kinetic Model

The second order kinetic models for present investigations (different coagulant dosage and different fluoride concentrations) are shown in Fig-9 & 10. From the figures it is observed that the removal process shown linearity at lower fluoride concentration and at higher coagulant dosage compare to higher concentration and lower coagulant dosage respectively. The R^2 values of second order kinetic models at higher coagulant dosage and lower initial concentrations were compared with correlation values of first order kinetic model, which indicates that coagulative removal of fluoride by *Passiflora foetida* is approximately fit into second order kinetic model. The second order rate constant K_2 and $1/N_o$ values were determined from the slope and intercept of the plot.

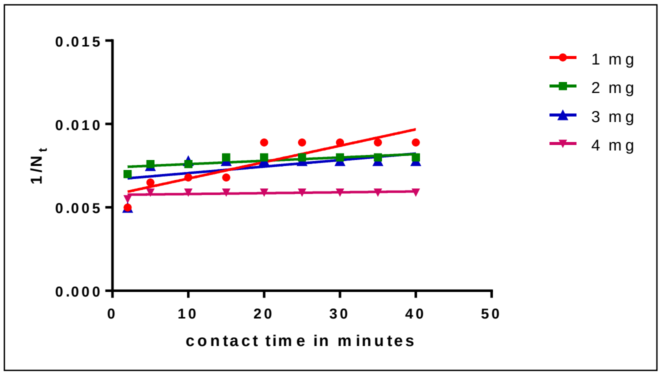

Figure 9 Second Order kinetic model at different fluoride concentrations

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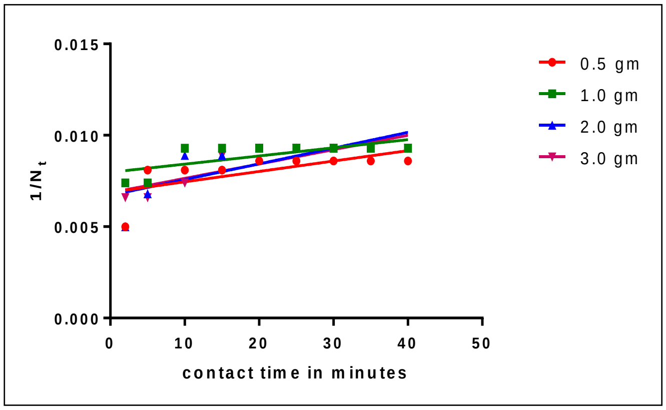

Figure 10 Second Order kinetic model at different coagulant dosage

FTIR report of *Passiflora foetida* before and after coagulation

As shown in Fig-11 and Fig-12, the spectrum displays several vibrational bands, indicating the complex nature of *Passiflora foetida*. There are three broad and intense absorption peaks in the range of 3412-3582 cm^{-1} indicating the existence of bounded hydroxyl groups. The number of absorption peaks decreased and intensity of peak decreased and there is slight change in the wave number indicating participation of bounded hydroxyl groups in coagulation of fluoride by *Passiflora foetida*. There are intense absorption peaks observed in the range of 1638-1698 cm^{-1} which represents the presence of C=O (aldehydes, ketones, carboxylic acid, ester groups) before coagulation. However only one peak common to before and after coagulation is represented by 1655.82 cm^{-1}, which indicates that coagulation has occurred by breaking C=O carboxyl group. Peaks at 1376 cm^{-1} (lignin region), 1396 cm^{-1} and 1458 cm^{-1} represents the presence of C-OH group which were absent after coagulation, indicating the participation in coagulation process.

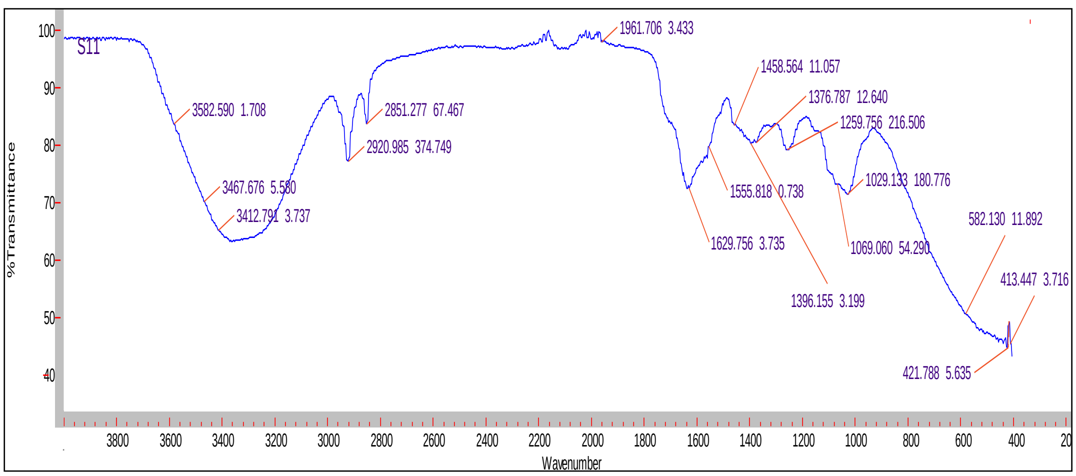

Figure 11 FTIR spectrum of *Passiflora foetida* before coagulation

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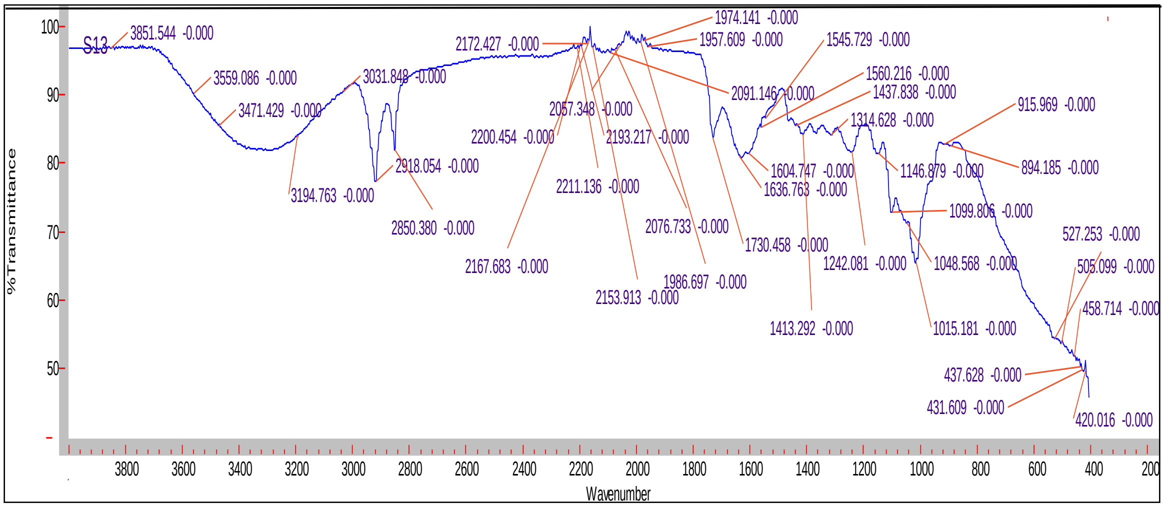

Figure 12 FTIR Spectrum of *Passiflora foetida* after coagulation

4. DISCUSSION

Coagulants are manufactured synthetic substances that water needs to help the strategy of precipitation of small scale particles that can't settle without any other substances. Consistently, industrial treatment used inorganic coagulants, for instance, alum, PAC, ferric chloride, ferric sulfate and cation polymer in their waste water treatment. Inorganic coagulants are more dominant than natural coagulants, anyway in high doses, they may cause quickens that are difficult to treat. This reason settles on natural coagulant as a decision to replace inorganic one.

Protein rich seeds and vegetative parts can be used as coagulant in light of the way that the seeds protein content that goes about as a polyelectrolyte. According to Dobrynin & Michael (2005), [52] polyelectrolytes are polymers that pass on positive or negative charges of ionized floc formation. In a polar dissolvable, for instance, water, the group may isolate, leaving the charge on its polymer chain and releasing the limiting molecule in solution. The extension of a polyelectrolyte center will result in diminished colloidal steadfastness and will decrease the rejecting power between particles to help the precipitation methodology. Coagulation is the creating of fine particles that happens as a result of its effects. Compound reactions are normally divided. Coagulation changes with various components like pH, temperature and portion. This procedure isn't feasible in many developing countries in view of high settled capital and detachment of manufactured/chemical coagulants. There are incredible arrangements of issues developing in view of the use of manufactured /synthetic or chemical coagulants. Alum treated water contain residual aluminum that prompts authentic medicinal issues, for instance, the headway of Alzheimer's contamination (AD) and weak dementia. Additionally, using of inorganic coagulants ensnares the dealing with and safe transfer undertakings. On account of these downsides of chemical coagulants, natural bio and plant based coagulants are taking lift from condition's point of view. Besides, plant based coagulants produces biodegradable and least volume slop 20-30% that of substance coagulant. Bio based or plant based coagulants have been used for household for an extensive timeframe in standard water treatment in tropical provincial regions.

Due to the lack of proper water treatment strategies and methodologies in these rural and under developed communities. Its application fuses departure of separated substance species and turbidity from water by methods for development of normal chemical based coagulants, explicitly, alum ($AlCl_3$), ferric chloride ($FeCl_3$) and poly aluminum chloride (PAC). While the ampleness of these engineered synthetic chemicals as coagulants is very much perceived, there are, regardless, loads related with use of these coagulants, for instance, deficiency in low-temperature water, reasonably high securing costs, negative effects on human prosperity, age of far reaching sludge volumes and the manner in which that they in a general sense impact pH of treated water.

Merits of Plant-Based Coagulants

The crucial good conditions of using ordinary plant-based coagulants as POU water treatment material are self-evident; they are monetarily clever, implausible to make treated water with incredible pH and significantly biodegradable. In the time of ecological change, utilization of earth's standard resources and expansive natural debasement, use of these coagulants is a vital effort as per the worldwide pragmatic enhancement exercises. Use of plant-based coagulants for turbid water treatment and lethal toxin evacuation returns to in excess of a couple of hundreds of years earlier and up to this point, biological specialists have had the ability to recognize a couple of plant types hence. While it is legitimate that the coagulants are inferred as essential nearby POU development, there have in like manner been different examinations focused on their use for treatment of modern wastewaters. It is fundamental for huge take holders to totally understand the subtleties included while contemplating the coagulants for commonplace, family unit or mechanical water treatment. To address this, this paper given a prudent answer for fluoride expulsion and the trademark coagulant sources, techniques and instruments included with the objective that natural bosses can tailor its usage for a crowd of water contaminants for explicitly fluoride evacuation. To give a dynamically connected with talk, regular coagulants got from non-plant sources, for instance, chitosan (comprehensively made from exoskeleton of shellfish) and isinglass (conveyed from fish swim bladders) are banned from this overview.

Natural Plant-Based Coagulants and Coagulation Mechanisms

Various examinations concerning normal coagulants insinuated them as 'polyelectrolytes' regardless of the way that an extensive parcel of these examinations did not by any stretch of the imagination guide in depth manufactured depiction to choose their ionic activity. Everything considered, this term should be used circumspectly, and be associated just after ionic development is set out to be accessible in the coagulant. It is essential to totally understand the crucial coagulation instruments related with these basic coagulants so all out trade in the going with portions. Cognizance of their use can be made sense of it. Gathering of Particulates in an answer can occur through four excellent coagulation frameworks: (a) twofold layer weight; (b) clear flocculation; (c) adsorption and charge equalization; and (d) adsorption and cover atom traverse. Adsorption and charge balance suggest the sorption of two particulates with oppositely charged particles while cover atom traverse happens when a coagulant gives a polymeric chain which sorbs particulates. Polymeric coagulants are generally associated with frameworks (c) and (d) as their since a long time prior binded structures (especially polymers with high nuclear burdens) remarkably increase the amount of empty adsorption areas. It gives that these two instruments give fundamental gauges to the inner tasks of plant based coagulants as well and they are the point of convergence of trade in the going with portions. The nearness of establishment electrolytes in watery medium can empower the coagulating effect of polymeric coagulants since there is lesser electrostatic repulsiveness between particles. But many plant-based coagulants have been represented; only four sorts are generally well known inside set up scientists, to be explicit, Nirmali seeds (*Strychnos potatorum*), *Moringa oleifera*, Tannin and Cactus, Peanuts, *Aloe vera*, mung Bean.

Table 3 Removal efficiency of fluoride from Electro plating industrial wastewater with chemical coagulants

S.No	Name of the Coagulant	Coagulant dosage (mg)	Initial Fluoride Concentration (mg/L)	Residual Fluoride (mg/L)	Fluoride removal (%)
1	$AlCl_3$	100	4.4	3	31.8
2	$MgCl_2$	100	4.4	2.70	38.6
3	$FeCl_3$	100	4.4	3.35	23.8
4	*Passiflora foetida*	100	4.4	2.44	44.5

Application

The developed method was applied for electroplating industry wastewater (which has 4.40 mg/L fluoride) was treated with 0.1 g of *Passiflora foetida* and removal efficiency was found to be promising when compared with chemical coagulants like $AlCl_3$, $MgCl_2$ and $FeCl_3$. Results were shown in table-3.

Conflict of interest

The authors declare that they have no conflict of interest.

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Ethical approval

This article does not contain any studies with human participants performed by any of the authors.

Data and materials availability:

All data associated with this study are present in the paper.

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