

## **19. Studies on The Treatment of Tannery Effluent by Using Animal Coagulant – Chitosan Extracted from Crab Shell**

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### **Abstract:**

*Tannery industry effluent is ranked as the highest pollutants among all industrial waste water. There are two types of tanning system which are vegetable tanning, which does not contain chromium, and chrome tanning. Tannery wastewater has the characteristics of pH-5.5, green colored, high COD, high pH, and high dissolved solids. These impurities mixed with water which are discharged from the tanneries and reached the ground water and pollute permanently and make the ground water is unfit for drinking, irrigation and general consumption. Therefore, there lies an urgent need to determine the pollution levels in the effluent from these industries. Hence, an attempt has been made to treat the vegetable tannery effluent by using natural material such as chitosan. The tannery effluent was collected from a typical tannery industry and analyzed the physicochemical impurities such as color, odor, temperature, pH, electrical conductivity, acidity, alkalinity, chemical oxygen demand, total dissolved solids, total solids, total suspended solid, chloride, calcium, nitrate, sulphate, fluoride and phosphate. The results obtained were compared with the CPCB Standards for the discharge of effluents into water.*

*The values of total dissolved solids, chemical oxygen demand, and chloride were higher when compared with the standard, the rest of the parameters were within limit. Hence, it was treated with Chitosan polysaccharide extracted from Crab shell. About 77% of Chloride, 67% of TDS, 60% of COD removal took place when the tannery effluent was treated with 0.75g of chitosan. GC-MS studies also confirmed the reduction of pollutant from the effluent. The study proved the pollutant removal capability of the natural material chitosan.*

### **Keywords:**

*Tannery Effluent, Chitosan, Natural Coagulants, COD removal.*

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## **19.1 Introduction:**

Water of good drinking quality is the basic importance to human physiology. Life's existence depends very much on the availability of water. An average human being, requires about 3 L of water in liquid and food daily for wellbeing. Water abundance, good quality drinking water is not readily available to man. Water quality is essential for all people's wellbeing. The quality of water can be affected by different pollutants such as waste from industries like chemical, physical and biological contaminants such as bacteria, viruses, heavy metals, and nitrate salts, where they have found their way into the water supplies. Water pollution occurs when a water body are adversely affected by the addition of huge number of materials to the water. The sources of water pollution are categorized as being a point source or nonpoint source of pollution. Point sources occur when there is runoff of pollutants into a water way (Manohan, 2000).

Due to the human population, industrialization, use of fertilizer in agriculture and other man-made activity are causing heavy metal pollution and varied pollution in aquatic environment, leading to the depletion of water quality and aquatic biota. It is therefore necessary that the quality of drinking water should be checked at regular time interval because, due to the use of contaminated drinking water, human population suffers from a variety of water borne diseases. (Manjare *et al.*, 2010)).

Tannery industry is one of the oldest industries in the world. Tanning processes were organized to fulfill the local demands of leather goods like musical instruments, drums and footwear. Leather is one among the major foreign exchange earners in India. Although tanning industries exist for a long time. The problems of environmental pollution receive serious consideration only in recent years. The pollutants from the large number of tanneries in the country have caused considerable damage in drinking water supply and irrigation, untreated wastewater when allowed to stagnate as in being done in most cases, gives rise to odour nuisance, unsightly, appearance, creating water pollution (Durai and Rajasimmam, 2010).

A total number of 2,161 tanneries are located in India, and spread across the states of Tamilnadu, West Bengal, Maharashtra, Punjab, Karnataka, Andhra Pradesh, Bihar and Uttar Pradesh. There are about 568 tanneries located in Tamilnadu. Among these, Dindigul is an important centre for leather processing with 61 tanneries. The effluent discharged from tanneries contains high values of pH, EC, chlorides, sulphides, sulphate, and carbonate chromium. BOD, COD, oil and dyes. The production processes in a tannery can be split into main categories 1) 5 Hide and skin storage and beam house operation 2) Tan yard operation 3) Post tanning operation 4) Finishing operation (Durai and Rajasimmam., 2010). The presence of heavy metals in drinking water can be hazardous to consumers because it can damage nervous systems, liver and bones and block functional group of vital enzymes. Such metals cannot be degraded or detoxified by biologically and have affinity to accumulate in living organisms. Although some heavy metal ion plays a pivotal role in living systems, they are very toxic and hence capable of causing serious environmental health problems. Chromium is a heavy metal used various industry processes like in chrome plating; wood preserving, textile dyeing, pigmenting, chromium chemical production, pulp and paper industrial and tanning process (Leta et al.,2004).

In general, tannery wastewater is imparted with a dark brown colour and have a high content of organic substances that vary according to the chemicals used (Kongjao et al., 2007). Jamal et al., (2004) has collected the tannery effluent in Ambur and has analyzed the physico-chemical parameters of the effluent. The physico-chemical parameters such as color, odor, pH, electrical conductivity, TSS, TDS, BOD, COD, Total hardness, calcium, magnesium, chloride, sulphate and total chromium were determined. The results revealed that almost all the parameters were found to be high and exceeded the CPCB limits.

Conventional methods for tannery effluent treatment consist of various combinations of biological, physical and chemical methods. Coagulation and flocculation process have many advantages including low cost, compactness, flexibility, easy working, toxic compounds removal and minimum energy usage. This method is a good choice for the removal of various pollutants like, heavy metals, dyes and oil compounds from various industrial waste sources. There are many inorganic coagulants used in industries, however, they are having some drawbacks such as producing large amounts of sludge, high cost and adverse effects on human health which can be resolved by using natural coagulants (Tawakkoly et al., 2019).

Chemical coagulants such as  $\text{FeCl}_2$  or alum is commonly used for the treatment. They are high cost, less active in low temperature wastewater, which cause serve adverse effects to the huge quantity of solid waste after the treatment. They also disturb the pH stability of the water. Bio based coagulants are low cost, it treats water at extreme pH and highly degradable. One such natural coagulant is chitosan. Chitosan is a  $\beta$ -1-4- linked polysaccharides obtained by the partial deacetylation of chitin. chitin is a biopolymer present in the shell of crustaceans, the cuticles of insects and the cell wall of fungi.

They are non-toxic, and bio-decomposability (Jin-hsing chang *et al.*, 2012). Because of these advantages, they are widely used in environmental pollution remediation to treat tannery effluent, food processing waste and conditioning of sludge and also be used for the adsorption and binding of heavy metals. It is a natural cationic polymer and act as polyelectrolyte used for waste water treatment (Brain Bolto & John Gregory, 2007).

## **19.2 Materials and Methods:**

Physico-chemical characterization of Tannery effluent the physico-chemical characteristics of tannery effluent includes colour, odour, pH, temperature, Total Solids (TS), Total Dissolved Solids (TDS), Total Suspended Solids (TSS), Total hardness, Alkalinity, Chloride and Chemical Oxygen Demand (COD) were determined using standard analytical methods (APHA, 2012).

### **A. Comparison of Physicochemical Characteristics of Drinking Water Samples With (CPCB) Standard:**

The physicochemical characteristics of tannery effluent were analysed and compared with CPCB effluent standard. BOD, TDS, and COD of tannery effluent were found to be higher and hence, an attempt was made in this study to reduce these factors using animal based natural coagulants as eco-friendly treatment technique.

**Collection of Crab Shell waste:** The crab shell wastes were collected from Local market of Kodaikanal. The crab shell was washed thoroughly with tap water and distilled water to remove the sand particles and other dirt. Cleaned crab shell was stored in the refrigerator for further use.



**Figure 19.1: Crab shell**

**Preparation of Chitosan (Sumaila et al., 2020):** Chitosan, a product of chitin is having positive charges, is an important natural biopolymer. The four main steps such as Deproteinization, Demineralization, Decolorization, and Deacetylation are involved in chronological order to produce chitosan from crab shell:



**Figure 19.2: Chitosan powder**

### **B. Effect of Chitosan Treatment on Chloride, TDS, and COD Removal from Tannery Effluent:**

About 100ml tannery effluent was taken in six conical flasks. Chitosan of practical grade isolated from crab shells (commercial) was used. Different concentrations of 0.25g, 0.50g, 0.75g, 1g, 1.25g, 1.50g of chitosan was added in each flask. It was mixed and the solution was undisturbed for 3 hours. After, it was filtered with Whatman No.1 filter paper of 110mm of diameter used for further analysis. The filtrate was subjected to colour, TDS, chloride and COD.

### **C. GCMS Analysis of Tannery Effluent:**

Raw and chitosan treated effluent were subjected with GC-MS analysis. It was carried out using a system comprising an AOC-20i auto sampler and gas chromatography interfaced to a mass spectrometer (GCMS-QP-2010). The interpretation of the mass spectrum GC-MS was inferred using the database of National Institute Standards and Technology (NIST) having more than 62,000 patterns. The name molecular weight and structure of the components of the sample was ascertained using NIST Ver.2.1 MS data library.

### **D. FTIR Characterization of Chitosan Treated Tannery Effluent Sludge:**

Chitosan active functional groups matter participated in the TDS, COD, and removal from tannery effluent samples were identified through FTIR Spectral analysis (before and after treatment) by using FTIR Instrument (Model: spectrum Perkin RXT) by directly placing the potassium bromide crystals over the sample. The mass spectrum obtained in the mid IR region of 450-4000cm<sup>-1</sup> was recorded using ATR (Attenuated Reflectance Technique) in transmittance mode

## **19.3 Results & Discussion:**

### **A. Physicochemical Characterization of Tannery Effluent:**

Various physical and chemical parameters such as color, odor, temperature, pH, electrical conductivity, total solids, total dissolved solids, total suspended solids, total alkalinity, acidity, calcium, magnesium, total hardness, chloride, nitrate, sulphate, phosphate, fluoride were analyzed. Color in fresh water made by the presence of minerals such as manganese and iron or algae and weeds. Color tests indicate the efficacy of the water treatment system. The color of tannery effluent sample was green color, and it may be contributed by the dissolved, suspended solids present in the effluent. Similar such results have been reported by many researchers. Odor pollution of tannery effluent is caused by chemical agents like alcohols, free chlorine, hydrogen sulphide, ammonia, phenols, and esters. The sample contained objectionable odor. This may be due to the presence of organic, inorganic compounds produced during the process of tanning. Similar results have been reported by Saritha and Manikandan, (2013). pH is the negative logarithm of the hydrogen ion concentration of water and the range of pH is given 0 to 14. The pH of the tannery effluent was within the accepted limit as per CPCB waste water discharge regulation.

The pH of tannery effluent was 7.5. Conductivity shows significant connection with ten parameters such as temperature, pH value, alkalinity, hardness, calcium, total solids, total dissolved solids, chemical oxygen demand, chloride and iron concentration of water. Patil et al., (2012). The effluent had 7.25cm of electrical conductance. Similar results have been reported by Masilamani et al., (2017). Turbidity is caused by the presence of suspended solids. Suspended solids may consist of clay, silt, airborne particles, and colloidal organic particles. TDS values are mainly due to carbonates, bicarbonates, chlorides, sulphates, phosphates, nitrate etc. (Esmail and jahal,2005). It is composed primarily of carbonate and bicarbonate; alkalinity acts as a stabilizer for pH. It is an important parameter which indicates the alkalinity of water to neutralize acids from waste water. The amount of alkalinity in tannery effluent was 305mg/L. Similar results have been reported by Amaliya and Sugirtha., (2013).

Chloride is associated with salty water. Higher amount of chloride contents are corroding the metallic pipes as well as for agricultural crops. Intrusion of sea water is main source of chloride in natural fresh water. The effluent had 3998 mg/L of chloride. Similar results have been reported by Sabur *et al.*, (2013).

COD is the amount of oxygen required to completely oxidise the chemical species present. An empirical relation is available between chemical oxygen demand and BOD. Chemical Oxygen Demand is the measure of amount of oxygen required to breakdown both organic and inorganic matters. The effluent had about 8100mg/L of COD. Similar results have been reported by Sabur *et al.*, (2013).

Hence, an attempt was made to comprehend the physico and chemical properties of tannery effluent and treatment of the same with natural material - chitosan. The characterization of waste water showed the higher concentration of all pollutants. It is not attained within the prescribed limits of CPCB standard. Hence, it is meant to be treated.

**Table 19.1: Physico Chemical Parameter of Tannery Effluent**

| Sr. No | Parameters             | Values mg/L   |
|--------|------------------------|---------------|
| 1      | Colour                 | Green color   |
| 2      | Odour                  | Objectionable |
| 3      | pH                     | 7.58          |
| 4      | Temperature            | 20-35°C       |
| 5      | Totalsolids            | 4,680         |
| 6      | Totaldissolvedsolid    | 3,560         |
| 7      | Totalsuspendedsolid    | 1,120         |
| 8      | Alkalinity             | 305           |
| 9      | Acidity                | nil           |
| 10     | Chloride               | 3998          |
| 11     | Dissolved oxygen       | nil           |
| 12     | Chemical oxygen demand | 8100          |
| 13     | Nitrate                | 17.4          |

| Sr. No | Parameters | Values mg/L |
|--------|------------|-------------|
| 14     | Phosphate  | 0.5679      |
| 15     | Fluoride   | 0.0169      |
| 16     | Chromium   | nil         |

### B. Effect of Chitosan Treatment on Chloride, TDS, and COD Removal from Tannery Effluent:

Different dosages of Chitosan were used to treat wastewater for the removal of chloride, TDS and COD. About 77% of Chloride, 67% of TDS, 60% of COD removal took place when the tannery effluent was treated with 0.75g of chitosan. After that, there was no major reduction noticed in the TDS, chloride and COD from the tannery effluent. Hence, this could be the best (optimum) dosage (Table 3).

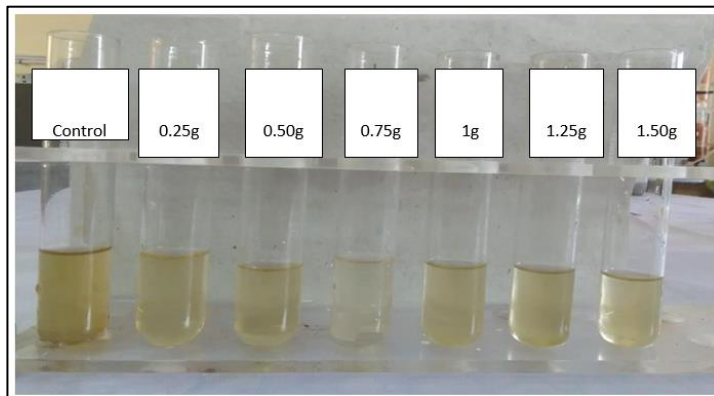
Hossain *et al.*, (2018) affirmed that the chitosan employed effective results in decolorization and COD removal from the industrial effluents. Chitosan is a copolymer of glucosamine and N-acetylglucosamine, and it has an amine functional group that is strongly reactive with metal ions (Zubair *et al.*, 2020). In recent years, considerable research has been done on the uptake of metal cations by chitosan (Gavhane *et al.*, 2013).

Researchers have reported that the COD and turbidity reducing ability of Chitosan due to high charge density property of chitosan resulted quickly destabilization of colloidal equilibrium in the waste water (Nechita *et al.*, 2015). Chitosan has the ability of biosorption as ion exchange, complexation, micro-precipitation and electrostatic attraction for the removal of metal from effluent (Nechita (2017)).

Chitosan biopolymer has high cationic charge density, long chains with high molecular weight and primary amino groups which made the biopolymer more effective coagulant or flocculent for the removal of impurities especially suspended and dissolved state from the waste water.

Nouj *et al.*, (2021) used liquid chitosan prepared from shrimp shell for the treatment of fish processing waste water successfully. Hence, the chitosan involved in the biosorption and coagulation process for the removal of color, TDS and COD. The results are depicted in the following table 2

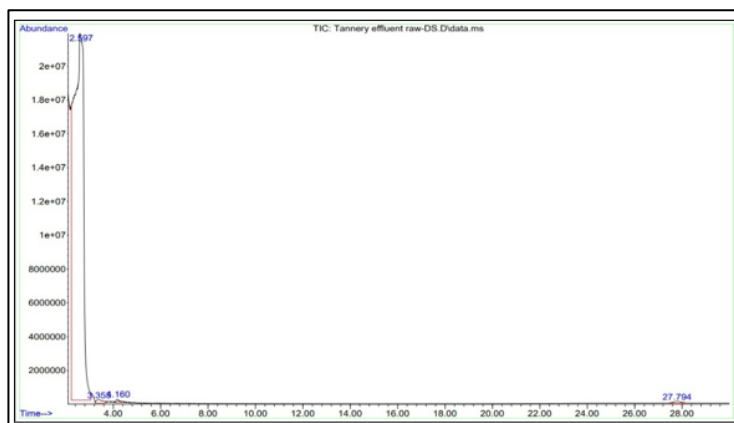
| Sr. no | Concentration (gm) | Chloride reduction (%) | TDS reduction (%) | COD reduction (%) |
|--------|--------------------|------------------------|-------------------|-------------------|
| 1      | 0.25               | 60                     | 40                | 45                |
| 2      | 0.50               | 65                     | 53                | 50                |
| 3      | 0.75               | 77                     | 67                | 60                |
| 4      | 1                  | 70                     | 65                | 54                |
| 5      | 1.25               | 65                     | 60                | 53                |
| 6      | 1.50               | 63                     | 60                | 50                |



**Figure 19.3: Effect of chitosan treatment on chloride, TDS, and COD removal from Tannery effluent**

### C. GC-MS Analysis of Raw Tannery Effluent:

GC-MS chromatogram of raw Tannery effluent shows four peaks indicating the presence of four chemical constituents are illustrated in table 5. The identified compounds includes Benzoic acid, 3,5-dichloro-4-hydroxy-, methyl ester, 2-Pyridinecarboxylic acid, 2,2,4,4-Tetramethyl-2,4-disilathietane, Propene. Similar results also revealed by Adinolfi et al., (2000).



**Figure 19.4: GC-MS analysis of Raw Tannery effluent**

**Table 19.3: GC-MS analysis of Compound Present in The Raw Tannery Effluent**

| Sr. No | RT     | Compound Name  | Chemical formula                               | Molecular Weight |
|--------|--------|--|--|------------------|
| 1      | 28.023 | Benzoic acid, 3,5- dichloro-4-hydroxy-, methyl ester | C <sub>20</sub> H <sub>28</sub> O <sub>3</sub> | 316              |
| 2      | 4.194  | 2-Pyridinecarboxylic acid                            | C <sub>2</sub> H <sub>5</sub> NO <sub>2</sub>  | 123              |



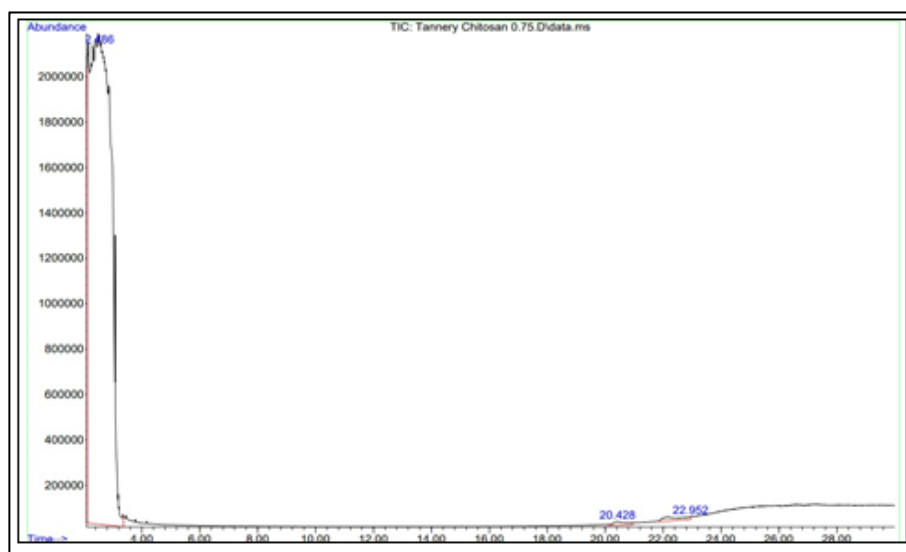
| Sr. No | RT    | Compound Name                           | Chemical formula                                | Molecular Weight |
|--------|-------|---|---|------------------|
| 3      | 3.421 | 2,2,4,4-Tetramethyl- 2,4-disilathietane | C <sub>5</sub> H <sub>14</sub> SSi <sub>2</sub> | 162              |
| 4      | 2.546 | Propene                                 | C <sub>3</sub> H <sub>6</sub>                   | 42               |

#### D. GCMS Analysis of Chitosan Treated Tannery Treatment:

GC-MS chromatogram of chitosan show three peaks indicating the presence of three chemical constituents are illustrated in table 19.6. The identified compounds include Pentadecanoic acid,13-methyl-, methyl ester, Propene, Oleicacid,3-(Octadecyl)propyl ester. It proves the treatment ability of chitosan. Pollutants from raw effluent were removed by flocculation ability of chitosan. (Table 19.4)

**Table 19.4: GCMS Analysis of Chitosan Treated Tannery Treatment**

| Sr. No | RT     | Compound Name                              | Chemical formula                               | Molecular Weight | Biological activity                                       |
|--------|--------|--|--|------------------|---|
| 1      | 20.513 | Pentadecanoicacid,13-methyl-, methyl ester | C <sub>17</sub> H <sub>34</sub> O <sub>2</sub> | 270              | Antimicrobial activity (Orishadipeet <i>et al.</i> ,2012) |
| 2      | 2.529  | Propene                                    | C <sub>3</sub> H <sub>6</sub>                  | 42               | Antimicrobial activity. (Orishadipe <i>et al.</i> ,2012)  |
| 3      | 22.094 | Oleicacid,3-(Octadecyl)propylester         | C <sub>39</sub> H <sub>76</sub> O <sub>3</sub> | 592              | Antifungalactivity (Das <i>et al.</i> ,2012)              |



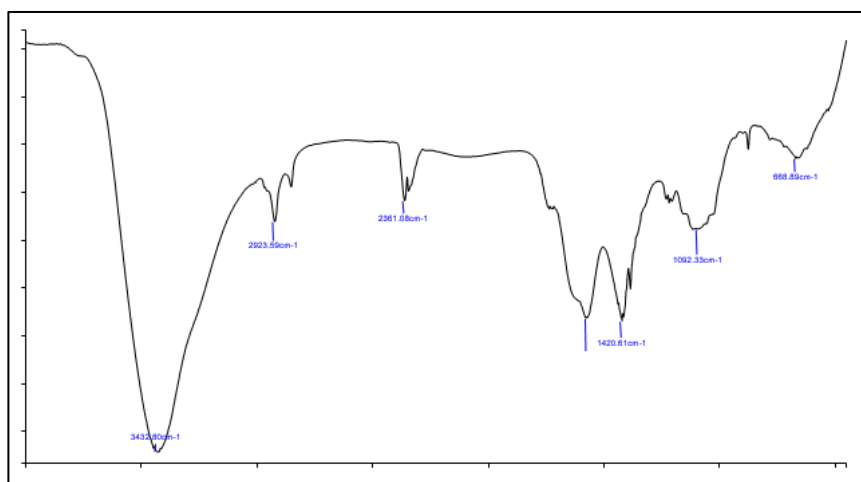
**Figure 19.5: GC MS chromatogram of Chitosan treated tannery effluent**

### E. FTIR Spectrum Analysis of Chitosan Treated Tannery Effluent Sludge:

Characterization of chitosan to tannery wastewater treatment as showed seven absorbance peaks at 3432.80 cm<sup>-1</sup> the presence of, functional group Alcohol with O-H Stretching. The peak 2923.59 cm<sup>-1</sup> and 2361.08 cm<sup>-1</sup> the presence of, functional group Alkane with C-H Stretching. The peak 1571.79 cm<sup>-1</sup> indicates the presence of functional group Amide with N-H Stretching (Table 19.5). Chitosan structure has an amine group (NH<sub>2</sub>) that has a PKa value of about 6.5. This means that protonation occurs in acidic to a neutral solution with the charge density depending on the pH value (Wang et al 2007). These amine and other groups were accountable for the taking away of pollutants from the waste water of tannery.

**Table 19.5: FTIR Spectrum Analysis of Chitosan Treated Tannery Effluent Sludge**

| Sr. No | Peak cm <sup>-1</sup>   | Functional group          | Interpretation |
|--------|-------------------------|---------------------------|----------------|
| 1      | 3432.80cm <sup>-1</sup> | Alcohol                   | O-H Stretch    |
| 2      | 2923.59cm <sup>-1</sup> | Alkane                    | C-H Stretch    |
| 3      | 2361.08cm <sup>-1</sup> | Alkane                    | C-H Stretch    |
| 4      | 1571.79cm <sup>-1</sup> | Amide                     | N-H Stretch    |
| 5      | 1420.61cm <sup>-1</sup> | Alkenes                   | C-C Stretch    |
| 6      | 1092.33cm <sup>-1</sup> | Carboxylic acids, esters. | C-O Stretch    |
| 7      | 668.89cm <sup>-1</sup>  | Alkyl halides             | C-Br stretch   |



**Figure.19.6: FTIR Spectrum Analysis of Chitosan Treated Tannery Effluent Sludge**

### 19.4 Conclusion:

Among the dosages 0.75g of chitosan has shown good reduction of color, TDS and COD from the tannery effluent. The study concludes that the natural material chitosan can be used as coagulants aid in future for tannery effluent treatment.

### **19.5 Acknowledgement:**

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