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# Study on the Effective Removal of Heavy Metals from the Tannery Industry Effluents using Natural Adsorbents

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## Abstract

The present investigation focuses on a sustainable and innovative strategy for the removal of heavy metals from contaminated water by the application of a low-cost and highly efficient adsorbent. The increasing volume of environmental degradation due to rapid urbanization, industrialization and unsustainable anthropogenic activities has severely impacted the water quality that leads to the depletion of freshwater resources and the accumulation of hazardous wastes. One of the most critical issues arises from the massive discharge of effluents generated by chemical based industries such as electroplating, mining, metal finishing, tanning, textile, fertilizer and paint manufacturing units. These effluents contain significant concentrations of toxic heavy metals that results the environmental pollution. Given these concerns, the systematic analysis and monitoring of surface water and groundwater for heavy metal contamination has become imperative to safeguard environmental sustainability and public health. These include the requirement of extensive land area for operation, complex and costly sludge management processes, high operational and maintenance costs, and dependence on trained technical personnel. Furthermore, many of these conventional techniques are inefficient to treat low-concentration pollutants and the production of secondary pollutants. Batch experiments are carried out for kinetic studies on the removal of heavy metals from aqueous solution. AAS is used for the determination of dissolved and total recoverable heavy metals in industrial effluent. To overcome these limitations, the present study emphasizes the development and utilization of a feasible adsorbent as an alternative remediation strategy.

## Introduction

Environmental pollution is being considered as the unfavourable alteration of water, air, soil and land. Environmental degradation is manmade due to rapid industry aviation and urbanization over the years. Heavy metals present in the industrial effluents are introduced into aquatic ecosystems not only through industrial discharges but also via domestic sewage outflows, atmospheric deposition of street dust, surface runoff from agricultural and urban lands and emissions from fossil fuel combustion. The perseverance and non-biodegradable nature of these heavy metals makes them extremely hazardous and tend to accumulate in water, soil, and biological tissues. Even at trace levels, metals such as mercury (Hg), cadmium (Cd), lead (Pb), arsenic (As), cobalt (Co), manganese (Mn), iron (Fe), and chromium (Cr) have been identified as highly deleterious to aquatic ecosystems and possess severe risks to human health through bioaccumulation and biomagnification in the food chain. Prolonged exposure to these contaminants can result in neurological disorders, kidney damage, carcinogenic effects, and other chronic health issues.

The conventional leather tanning technology is highly dangerous as it produces large amounts of organic and chemical pollutants. These pollutants are a serious threat to the environment as they can cause serious damage to soil and water bodies. The high amount of salt contained in the effluent can increase soil salinity, reduce the soil fertility and damage farming. However, existing technologies for heavy metal remediation, such as chemical precipitation, ion exchange, electrochemical treatment, and membrane filtration, often suffer from inherent drawbacks. Adsorption using activated carbon is a best method for the treatment of tannery effluents contaminated with heavy metals such as mainly Cr (II), Cu (II) and Zn (II). Adsorption is considered one of the most promising methods for heavy metal removal due to its simplicity, cost-effectiveness, high efficiency, and the possibility of adsorbent regeneration and reuse. By employing this approach, it is possible to achieve a sustainable balance between environmental protection, economic feasibility, and practical applicability for large-scale wastewater treatment.

Food crop namely Triticum spelta is used as an adsorbent for the removal of Cr (II), Cu (II) and Zn (II) from waste water. Batch experiments are carried out for kinetic studies on the removal of chromium, copper and zinc from aqueous solution. The effects of various influencing parameters such as initial pH, contact time, dose of adsorbent and initial concentration of adsorbate are studied.

## Materials and Methods

### *Preparation of Adsorbent (FLY ASH)*

The pulverized fly ash used in this study was collected from the National Thermal Power Corporation (NTPC), Ramagundam, Andhra Pradesh. The raw PFA was initially washed thoroughly with distilled water to remove soluble impurities. It was then activated by treating with concentrated sulphuric acid (98% w/w) in a 1:1 ratio. The acid treated PFA was placed in a hot air oven maintained at 150°C for 12 hours. After activation the PFA was again and again washed with distilled water until all traces of free acid were removed, followed by drying at 110°C for 5 hours.

### *Preparation of Adsorbent-Triticum Spelta (ATS)*

Triticum spelta was collected from areas in and around Dindigul district, Tamil Nadu. The activation process was carried out in two stages.

1. ***Carbonization***

   The raw material was submitted to carbonization process that is converting it into wood charcoal. This process was carried out in mud-pits, brick kilns or portable metallic kilns.

2. ***Chemical Activation***

   The obtained wood charcoal was treated with concentrated sulphuric acid and kept in an oven at 150°C for 12 hours. This activation step is used to enhance the internal surface area of the adsorbent by creating more active sites for adsorption.

### Characterization of Adsorbents

The physicochemical properties such as moisture content, particle density, ash content, and water-soluble components of activated pulverized fly ash (PFA) and activated Triticum spelta (ATS) were determined. The results are presented in below Table.

| ***PARAMETERS*** | ***PFA*** | ***ATS*** |
|---|---|---|
| pH | 6.89 | 5.76 |
| Bulk density(g/cc) | 0.99 | 0.97 |
| Particle density(g/cc) | 1.98 | 1.45 |
| Moisture (%) | 27.90 | 7.25 |
| Ash (%) | 68.90 | 22.99 |

The bulk density and particle density of the adsorbents significantly influence the adsorption capacity for metal ions. A decrease in bulk density generally enhances adsorption efficiency. Adsorbents with bulk density values below 1.25 g/cc are classified as fine, those values between 1.25-2.00 g/cc as medium and values above 2.00 g/cc as coarse. Similarly, particle density values below 2.25 g/cc indicates fine materials, 2.25-4.00 g/cc indicates medium and values above 4.00 g/cc indicates coarse adsorbents.

In the present study, the bulk density and particle density values obtained are closer to fine in nature.

- Both PFA (0.99 g/cc) and ATS (0.97 g/cc) fall below 1.25 g/cc classified them as fine adsorbents with high surface area.
- Particle densities (1.98 g/cc and 1.45 g/cc) confirm their fine porous structure.
- Moisture was higher in PFA (27.90%) compared to ATS (7.25%) is potentially reducing active adsorption sites.
- Ash content was significantly higher in PFA (68.90%) than in ATS (22.99%), suggesting that ATS has a superior adsorption efficiency.

***Other Chemicals***

All chemicals used were in this method is analytical reagent (AR) grade. Standard buffer solutions of pH 4, 7, and 9 were used for calibration of the pH meter. The following solutions were prepared as per standard procedures:

- Individual standard solutions of Cr(II), Cu(II), and Zn(II) ions.
- Sodium hydroxide (NaOH) pellets.
- Sodium nitrate ($NaNO_3$).
- 0.5 N Hydrochloric acid (HCl).
- Concentrated nitric acid ($HNO_3$).

***Heavy metal analysis***
***Analysis of heavy metals using atomic adsorption spectro photometer (AAS)***

The determination of heavy metal ions was carried out using the Atomic Absorption Spectrophotometer (AAS). AAS was chosen in this study because it provides reliable results even at concentrations lower than 6 mg/L. This technique is recognized for its sensitivity, selectivity, and reproducibility in trace metal analysis which is particularly suitable for analyzing industrial effluents. The extent of absorption is proportional to the concentration of the element in the sample. The advantage of this method is that each element absorbs light at a unique wavelength for reducing spectral interference. Furthermore, Flame AAS provides higher sensitivity for transition metals compared to flame emission spectroscopy. The Direct Air Acetylene Flame Method was used as it ensures efficient atomization of transition metals.

- Wavelength for Chromium (II) is absorbed at 282.3 nm.
- Wavelength for Copper (II) is absorbed at 222.8 nm.
- Wavelength for Zinc (II) is absorbed at 213.9 nm.

***Batch Adsorption Experiments***

Batch adsorption studies were conducted to evaluate the removal efficiency of Pulverized Fly Ash (PFA), Activated Triticum spelta (ATS), and a 50:50 mixture (PFA:ATS) for the presences of chromium, copper, and zinc ions.

Batch adsorption studies were carried out in 250 ml Borosil shaker bottles containing 100 ml of aqueous solution and a pre-weighed quantity of adsorbent. Three different adsorbent systems were employed. One is pure form of PFA, another one is pure ATS and finally a 50:50 mixture of PFA and ATS. The bottles were shaken at a controlled temperature of 30-35°C for predetermined contact times as it was determined from preliminary kinetic studies. The initial concentration of adsorbate was maintained at 10 g/L. To study the influence of adsorbent dosage, the amount of adsorbent was varied within the range of 10-50 g/L while the adsorbate concentration was adjusted between 5 and 25 mg/L. The role of pH on adsorption was evaluated by adjusting the solution pH from strongly acidic (pH 2) to strongly alkaline (pH 12). Furthermore, the effect of contact time was examined by varying the duration of shaking between 30 and 180 minutes.

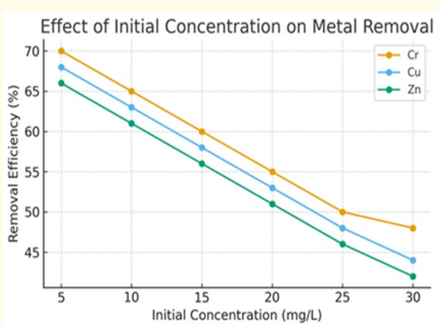
The adsorption efficiency was evaluated by varying the following parameters:

1. Initial concentration of adsorbate.
2. Adsorbent dosage.
3. Effect of pH.
4. Contact time.

***Study of Varying Concentrations***
***Effect of Initial Concentration***

The effect of variation in initial concentrations of Cr(II), Cu(II), and Zn(II) ions was studied at constant adsorbent dosage (10 g/L) and contact time (60 minutes). Results are indicated that the amount of metal ions adsorbed increased with increasing initial concentration, while the percentage removal decreased. This is allotted to the limited availability of active adsorption sites on the adsorbent surface at higher solute concentrations. The readings are observed that the optimum concentration range for efficient removal of heavy metals lies between 15-20 mg/L. The comparative adsorption efficiency of different adsorbents is summarized below:

| *Adsorbent* | *Percentage Removal (Identical Conditions %)* | *Percentage Removal (Optimum %)* |
|---|---|---|
| PFA | 48 | 60-70 |
| ATS | 58 | 70-75 |
| 50:50 (PFA: ATS) | 52 | 64-70 |

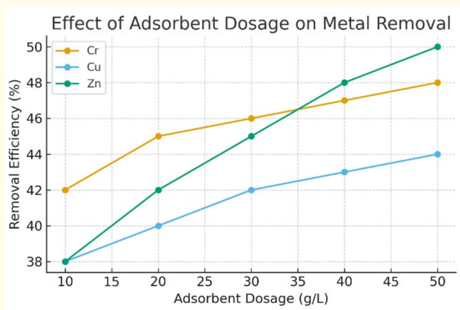


### *Effect of Adsorbent Dosage*

The impact of adsorbent dosage (10-50 g/L) was examined while keeping other parameters as a constant. The results are clearly explained that increase in the percentage removal of heavy metals with increasing adsorbent dosage, due to the availability of a larger number of active binding sites.

The adsorption efficiency of the three adsorbents was examined under optimum experimental conditions. Activated Triticum spelta (ATS) exhibited the highest removal efficiency while achieving 46-54% for chromium, 40-60% for copper, and 42-58% for zinc. In Pulverized Fly Ash (PFA) showed slightly lower performance with removal efficiencies of 42-48% for chromium, 38-44% for copper, and 38-50% for zinc. The 50:50 mixture of PFA and ATS demonstrated intermediate efficiency of removing 45-55% of chromium, 40-50% of copper, and 42-50% of zinc. These results clearly indicate that ATS is the most effective adsorbent among the three adsorbents.

| *Heavy Metal* | *PFA (%)* | *ATS (%)* | *50:50 Mixture (%)* |
|---|---|---|---|
| Chromium | 42-48 | 45-68 | 45-55 |
| Copper | 38-44 | 40-60 | 40-50 |
| Zinc | 38-50 | 42-64 | 42-50 |

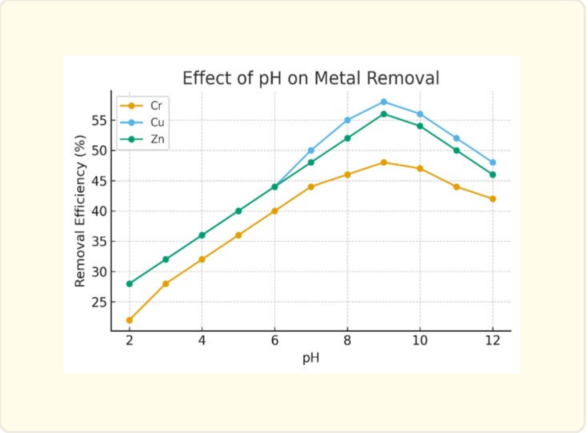



### *Effect of pH*

The adsorption of heavy metals is highly pH-dependent. Experiments were conducted at pH values ranging from 2 to 12 under optimum temperature (296 K) and contact time (60 min).The observed removal efficiencies are summarized below:

| *Heavy Metal* | *PFA (%)* | *ATS (%)* | *50:50 Mixture (%)* |
|---|---|---|---|
| Chromium | 22-48 | 22-66 | 9.2 |
| Copper | 28-44 | 20-58 | 8.4 |
| Zinc | 28-46 | 22-62 | 8.3 |

The results indicate that adsorption efficiency increases with pH. It reaches the maximum removal in slightly alkaline conditions before declining at higher pH levels. The optimum pH values were found for Chromium at 9.2, Copper at 8.4 and Zinc at 8.3. The slight decrease in solution pH (ΔpH = 0.2-0.5 units) after adsorption suggested that proton exchange occurred between metal ions and functional groups (such as phenolic, carboxylic, and enolic groups) present on the adsorbent surfaces. This confirms that ion-exchange and surface complexation were the dominant mechanisms of metal removal.




***Effect of Contact Time***

The influence of contact time on the removal of heavy metal ions was studied by varying the interaction period between the adsorbent and adsorbate from 30 minutes to 180 minutes under experimental conditions (temperature 296 K, adsorbent dose 10 g/L, and initial concentration 15-20 mg/L). The results showed that the adsorption efficiency increased rapidly during the initial stages of contact time due to the availability of a large number of vacant active sites on the adsorbent surface. The equilibrium contact time for effective removal of chromium, copper, and zinc ions was found to be around 90-120 minutes, after which the removal efficiency remained nearly constant.

| ***Adsorbent*** | ***Contact Time (min)*** | ***30*** | ***60*** | ***90*** | ***120*** | ***150*** | ***180*** |
|---|---|---|---|---|---|---|---|
| PFA | Chromium Removal (%) | 28 | 36 | 42 | 46 | 47 | 48 |
| | Copper Removal (%) | 25 | 32 | 38 | 42 | 43 | 44 |
| | Zinc Removal (%) | 26 | 34 | 40 | 45 | 47 | 48 |

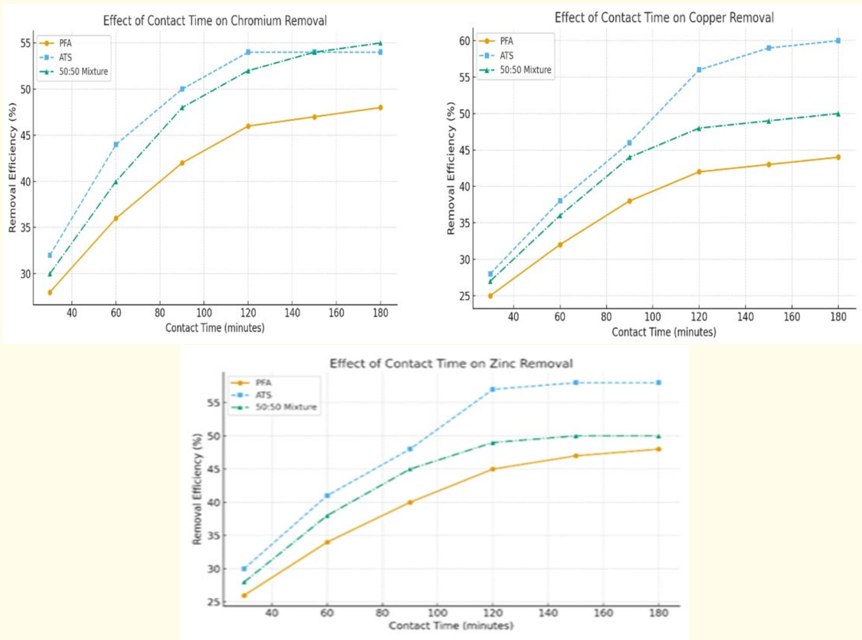
| ***Adsorbent*** | ***Contact Time (min)*** | ***30*** | ***60*** | ***90*** | ***120*** | ***150*** | ***180*** |
|---|---|---|---|---|---|---|---|
| ATS | Chromium Removal (%) | 32 | 44 | 50 | 54 | 54 | 54 |
| | Copper Removal (%) | 28 | 38 | 46 | 56 | 59 | 60 |
| | Zinc Removal (%) | 30 | 41 | 48 | 57 | 58 | 58 |

| ***Adsorbent*** | ***Contact Time (min)*** | ***30*** | ***60*** | ***90*** | ***120*** | ***150*** | ***180*** |
|---|---|---|---|---|---|---|---|
| 50:50 (PFA:ATS) | Chromium Removal (%) | 30 | 40 | 48 | 52 | 54 | 55 |
| | Copper Removal (%) | 27 | 36 | 44 | 48 | 49 | 50 |
| | Zinc Removal (%) | 28 | 38 | 45 | 49 | 50 | 50 |

***Table***: Effect of Contact Time on Removal of Heavy Metals.

From the tabulation the followings are findings

- Adsorption of all three heavy metals increases with contact time.
- A rapid uptake occurs within the first 60-90 minutes due to the availability of a large number of vacant active sites.
- Beyond 120 minutes, the adsorption rate slows and approaches equilibrium.
- The optimum contact time for effective adsorption is found to be around 120 minutes, after which the system reaches equilibrium.




## Conclusion

The present investigation explained about the utilization of Pulverized Fly Ash (PFA), Activated *Triticum spelta* (ATS), and their 50:50 mixture as low-cost adsorbents for the removal of heavy metals such as Chromium ($Cr^{2+}$), Copper ($Cu^{2+}$), and Zinc ($Zn^{2+}$) from aqueous solutions. The adsorbents were prepared, activated, and characterized in terms of their physicochemical properties, including pH, bulk density, particle density, moisture content and ash content. Batch adsorption experiments were systematically carried out by varying key parameters such as initial adsorbate concentration, adsorbent dosage, pH of the solution, and contact time.

The findings of the study can be summarized as follows:

Adsorption increased with rising metal concentration but the percentage of removal decreased due to the limited availability of active adsorption sites. Optimum concentration for effective removal was found to be in the range of 15-20 ppm. Maximum removal was achieved at a dosage of 50 g/L, where the removal efficiencies ranged from 48% (PFA) to 60% (ATS). Adsorption was strongly pH-dependent, with maximum removal observed in the slightly alkaline range (pH 8-9). The optimum pH values for removal were 9.2 for Chromium, 8.4 for Copper, and 8.3 for Zinc. Overall, the study demonstrated that ATS exhibited superior adsorption capacity compared to PFA, while the 50:50 mixture provided balanced and synergistic performance. The low cost, high availability, and eco-friendly nature of PFA and ATS make them promising alternatives for the treatment of heavy metal-contaminated wastewater.

This research highlights the potential of locally available, low-cost materials such as PFA and ATS as effective alternatives for wastewater treatment. With further refinement, large-scale application, and integration into treatment plants, these adsorbents could significantly contribute to sustainable and eco-friendly heavy metal remediation technologies.

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