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Efficiency of hair waste substrate in constructed wetlands on oil and grease content of kitchen wastewater

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Abstract: Treatment of wastewater from various day-to-day sources, particularly oil spills in water bodies, involves using resources that may adversely affect the environment. This study explores the potential of using human hair as a bio-adsorbent to remove BOD (Biochemical Oxygen Demand) and oil and grease from wastewater. The analysis of the percentage reduction in BOD and oil and grease using human hair was conducted through Response Surface Methodology (RSM) using the Central Composite Design method in Design Expert Software. Two variable parameters were chosen for the experiment: the thickness of the adsorbent (A) and the volume of the input sample (B). Adsorption experiments were carried out in a tray setup across 13 different combinations of these parameters. Human hair, the bio-adsorbent under consideration, proved effective in significantly reducing the BOD of wastewater samples and adsorbing oil and grease. In addition to the two parameters studied, other factors such as pH and contact time could also be considered to estimate the optimal adsorption conditions. The maximum percentage removal achieved in this set of experiments was identified by analyzing the interactive effects of the parameters on BOD reduction. Using human hair for oil adsorption can also result in a significant decrease in BOD, which may be beneficial for further treatment processes.

Introduction

Wastewater from restaurant kitchens and garages is known to cause acute and long-term damage to surrounding ecosystems due to its high content of oil and grease. Additionally, terrestrial oil spills degrade soil quality and persist for long durations (Murray et al., 2019). Several compounds originating from petroleum can remain in the environment even after the visible cleanup of the initial spill, and complete removal may never be achieved (Allan et al., 2012). Due to the toxicological properties of oil constituents, oil spills pose serious health risks not only to marine ecosystems but also to individuals involved in cleanup operations and coastal residents (Laffon et al., 2016). Because of their efficiency, cost-effectiveness, and trace-level adsorption capabilities, biological methods, particularly sorption, are typically chosen over mechanical and chemical methods of oil spill remediation (Jmaa and Kallel, 2019). Natural sorbents offer the advantages of being economical and biodegradable. Some of the sorbents have even demonstrated higher efficiency than commercially used polypropylene materials (Adebajo et al., 2003). These sorbents work by concentrating oil into liquid, semi-solid, or solid phases, enabling easier removal without draining out (Muhammad et al., 2012). One such natural sorbent examined in this study is human hair.

Oil and grease are defined as a group of related materials rather than a specific chemical compound extractable by certain solvents, such as hexane. Under anaerobic conditions, oils and grease hydrolyze to long-chain fatty acids and glycerol. The glycerol further degrades to 1,3-propanediol and subsequently to acetate. The presence of oil and grease in water bodies leads to the formation of an oil layer, which causes significant pollution problems such as the reduction of light penetration and photosynthesis. It further hinders oxygen transfer from the atmosphere to the water medium, leading to a decreased amount of dissolved oxygen at the bottom of the water, adversely affecting the survival of aquatic life in water (Sathinathan and Kiew 2021, Abbas et al., 2011). Oil content is in different forms in the liquid phase and contains single or multiple species in various concentrations. The type and nature of oil in oily wastewater vary from source to source. Oily wastewater includes fats, oils, cutting fluids, lubricants, edible oil, heavy hydrocarbons, including grease, crude oils, diesel oils, and light hydrocarbons, including kerosene, gasoline, and jet fuel. Oil and grease may also endanger human health as some constituents of oily wastewater are very toxic and carcinogenic, affecting the kidney, liver, and blood, and ultimately increasing the risk of cancer (Zhang et al., 2022, Sanghamitra et al., 2021).

Human hair comprises carboxyl, amido, and disulfide groups, which makes it an excellent biosorbent for a variety of pollutants, including oil and grease (Dan et al., 2020, Ogata et al., 2020). Around 65 to 95 percent of human hair is made up of proteins, with other components including water, lipids, polysaccharides, colors, nucleic acids, and trace elements. It is an abundant biomass source with exceptional stability and distinctive chemical properties. Human hair protein is completely hydrolyzed to reveal that it contains 13–18% cysteine, which protects the hair's structure against deterioration and exposure to reducing, oxidizing, and hydrolyzing chemicals. High wet strength, moderate swelling, and insolubility are all attributed to cysteine, a dimer of cysteine that is found in the cuticle layer (Ifelebuegu et al., 2015). The physical adsorption capability of human hair waste is characterized by the surface of human hair having numerous small fissures (heavy metals also adsorb on this site). Long peptide chains with unshared electron pairs, like O and N, are another component of human hair waste. Additionally, it has a lot of S-consisting cysteine, which binds to heavy metals strongly. This finding suggests that there is a strong binding affinity between heavy metals and cysteine amino acids in human hair (Saini and Melo, 2015; Zhang, H. et al., 2018).

Human hair, a naturally occurring biosorbent, is composed of the medulla, cortex, and dead cells made up of the cuticle, water, etc., primarily polymers of amino acids like keratin and cysteine. It is highly hydrophobic and water-repellent because of the cuticle content. Additionally, it has many peptide bonds, CO- and NH- groups that produce hydrogen bonds between nearby molecules on the surface of the hair, as well as a very porous cortex. It can be used repeatedly without significantly losing its sorption properties (Mondal and Basu, 2019). Black human hair is a bioresource that has naturally immobilized keratin and melanin in it. The cortex, which is a mechanical characteristic of the hair fiber, makes up most of the mass of the hair shaft. Melanin granules, which make up around three percent of the weight of hair, are found inside the cortex. It is more appealing for biosorption due to its qualities, including high tensile strength, non-toxicity, and water insolubility. Additionally, it has a high level of resistance to acidic solutions, other harsh chemicals, and environmental deterioration (Syed 2015). Hair is even known for its suitability as a low-cost alternative for the separation of heavy metals like chromium (VI) from aqueous solution (Murthy et al., 2004; Saha et al., 2019; Zhang et al., 2020).

The attraction between the sorbent's outer surface and the sorbate, or the fact that sorbate molecules collect on the sorbent's surface without entering it, is what drives oil adsorption. Oil adsorption occurs in three stages: the diffusion of oil molecules onto the sorbent surface, their capillary action-induced trapping inside the sorbent structure, and the accumulation of oil droplets within the sorbent's porous and uneven structure. Oil builds up on the surface of the adsorbent during physical adsorption because of various forces. Due to the chemical groups on the surface of the adsorbent/adsorbate, chemisorption can also happen with physical adsorption. Chemisorption is the process by which oil and the surface of the adsorbent share electrons to establish a chemical connection (Pagnucco and Philips, 2018). It is more likely that water will be adsorbed in greater amounts when consumed at higher concentrations than oil, however, investigations show that the opposite is true. When examined under an optical microscope, oil is shown to have taken the place of water on the surface of the hair. The adhesive forces between oil and hair are greater than those between water and hair due to selective physical adsorption, even in situations when water is present in higher concentrations than oil. When oil and water are present, hair selectively adsorbs the oil, enabling separation of the two when the hair is passed over a bed of adsorbent (Ukotije-Ikwut et al., 2016). Natural sorbents like hair are observed to possess poor buoyancy, which could be enhanced by replacing the outer material with a more hydrophobic and buoyant one or with the help of flotation devices (Van Den Berg et al., 1967).

The various human hair types that were studied were shown to have better adsorption capacity due to their tight, coarse, and spongy qualities. It could also be because there are more micropores, which aid in capillary action at the hair/oil interface, or because there are rougher cuticles, which are generated by several layers of hair and resemble the scalp. Hair can be recovered and reused using a variety of techniques, including centrifugation, compression, and solvent extraction. Even after being subjected to four cycles of recovery and reuse, the solvent extraction method used in this study demonstrated that there is no substantial decline in adsorption capacity (Badsha et al., 2021). The rate of adsorption decreased at the end of the time duration in longer-term adsorption studies with various ions but still produced a positive slope. This is a sign that the initial diffusion is only present in a specific area of the hair and that a much slower penetration into more central material occurs thereafter (medulla or cortex). The ions appear to penetrate the hair very slowly or not at all in some areas. It is currently unknown whether this component contains any natural ions (Ip et al., 2009).

Human hair is viewed as a waste product, and when it builds up in the waste stream, it can lead to environmental problems like land pollution (Gupta, 2014). Due to its slow decomposition characteristics, this waste eventually takes up a lot of space and forms leachate, which raises the nitrogen level in the water and causes eutrophication (Dan et al., 2020). The present study utilizes this waste of human hair as an adsorbent to remove the oil and grease and BOD in the wastewater from kitchens, restaurants, and garages. Usually, oil and grease traps are used to remove the greasy materials from wastewater. A preliminary treatment procedure is proposed for the removal of oil and grease and BOD at the source to reduce the load on further treatment of wastewater. This constructed wetland uses the appropriate thickness of human hair substrate to effectively treat different loading of BOD, oil and grease.

Materials and Methodology

The sample used in this biosorbent adsorption study is kitchen wastewater, which consists of detergents from utensil washing and other components like proteins, carbohydrates, oil and grease, and other dissolved and suspended compounds. The adsorption of oil and grease content in the sample over the chosen adsorbent occurs in the constructed wetland for a pilot study with various components.

The constructed wetland with a volume of 0.0044 m^3 consists of a layer of coarse aggregates (gravel) of size 10 mm

Fig 1. Constructed wetland for pilot study (An image showing the plastic box in which the layers of sand, adsorbent-hair, and gravel is filled to perform the experiment)

down, fine aggregates (sand) of uniform gradation, and the hair substrate with varying thickness. The tests were run on human hair collected from a local salon from healthy human beings. The hair was washed and cleaned to remove any existing impurities. The bottom-most layer of the tray is comprised of sand, followed by the adsorbent, and some gravel forming the topmost layer. The sand was sieved to a size below 4.75 mm to avoid bigger clumps of impurities present in the sand and was maintained at a height of 6-7 cm for both layers. The constructed wetland with human hair substrate is shown in Figure 1.

The sample solution was analyzed by performing the test of COD, BOD, oil and grease, turbidity, and pH on each different day of the week to establish the range of variation of these parameters. The values are shown in Table 1.

The COD was in the range of 960 mg/L to a maximum of 3360 mg/L, BOD from 6.24 mg/L to 509.09 mg/L, pH ranging from 5.61 to 7.67, turbidity varied from 14 NTU to 307 NTU, and oil and grease varied from 230 to 540 mg/L. The variation of the parameters for all the samples is shown in Table 1. Each day, we observe a large variation in turbidity, BOD, and COD, indicating significant differences in sample characteristics. The treatment to be provided must be efficient enough to handle this large variation. In the case of oil and grease content and pH, the sample shows fewer variations. However, the oil and grease content is not low enough to be disposed of directly into water sources. The wastewater sample is treated using a constructed wetland with human hair substrate, and its effect on reducing BOD and oil and grease content is investigated.

The Response Surface Methodology (RSM) analysis for percentage reduction in BOD and oil and grease of the wastewater using human hair was evaluated using the Central Composite Design method in the Design Expert Software. There were two variable parameters chosen for the experiment: the thickness of the adsorbent (A) and the volume of the input sample (B). The adsorption experiments using hair for the combination of 13 readings with these parameters (as shown in Table 2), which were generated using the Response Surface Methodology model in the Design Expert software, were then carried out in the tray setup.

Table 1. Range of values for different properties of wastewater sample

Sample Number	COD (mg/L)	BOD (mg/L)	pH	Turbidity (NTU)	Oil & Grease (mg/L)
1	960.0	10.18	6.85	29.0	226
2	3120	509.1	7.67	14.0	400
3	1280	168.7	6.59	81.0	100
4	1360	13.44	6.51	83.7	400
5	3360	6.240	5.61	307	10.0
6	1140	11.52	6.72	126	540

Table 2. Combination of the 13 readings of thickness and volume parameters, by using the Response Surface Methodology (RSM) and the Central Composite Design Method in the Design Expert Software.

Thickness (cm)	Volume (L)
1.0	4.0
0.4	3.0
1.0	2.0
2.5	3.0
2.5	1.6
4.6	3.0
4.0	4.0
2.5	4.4
4.0	2.0
2.5	3.0
2.5	3.0
2.5	3.0
2.5	3.0

In Table 2 and Table 3 source means the source of variation in the data. The sum of squares is the sum of squares between the group means and the grand mean. The d_f refers to the degree of freedom, where if there are n total data points collected, then there are n-1 total degrees of freedom. The mean square is the ratio of the sum of squares to the corresponding d_f. The F-value contains the F-statistic, which is the ratio of the mean square to the mean residual square. The p-value in the final column is the probability of rejecting a null hypothesis that is true by mistake. A p-value less than 0.05 is considered statistically significant. The model is a hypothesized relationship between the response variable and the predictor variable. The Residual Error is the amount by which an observed variation differs from the value predicted by the model. While the Lack of Fit is the variation between the actual measurements and the values predicted by the model, pure error meanwhile refers to the variation among any replicates. The Corr Total is the sum of the factors from the model and the residual parts.

Results

Response Surface Methodology (RSM) Analysis for adsorption using Human Hair

RMS analysis results indicate a decrease in BOD values after biosorption using human hair. The model best fits BOD

Table 3. ANOVA table for RSM studies on BOD decrease, for adsorption using Human Hair

Source	Sum of squares	d_f	Mean Square	F-value	p-value	
Model	912.5	5	182.5	5.20	0.026	Significant
A-Thickness	48.27	1	48.27	1.38	0.279	Insignificant
B-Volume	44.36	1	44.36	1.26	0.298	Insignificant
AB	78.32	1	78.32	2.23	0.179	Insignificant
A^2	285.2	1	45.16	8.13	0.025	Significant
B^2	346.0	1	346.0	9.86	0.016	Significant
Residual	245.5	7	35.08			
Lack of fit	245.5	3	81.84			
Pure Error	0	4	0			
Cor Total	1158	12				

Table 4. ANOVA table for RSM studies on Oil & Grease removal, for adsorption using Human Hair

Source	Sum of squares	df	Mean Square	F-value	p-value	
Model	1488	5	297.6	1.62	0.270	Insignificant
A-Volume	37.06	1	27.06	0.20	0.667	
B-Thickness	123.3	1	123.3	0.67	0.439	
AB	15.41	1	15.41	0.08	0.780	
A^2	1329	1	1329	7.25	0.031	
B^2	246.1	1	246.1	1.34	0.285	
Residual	1283	7	183.2			
Lack of fit	1283	3	427.5			
Pure Error	0	4	0			
Cor Total	2771	12				

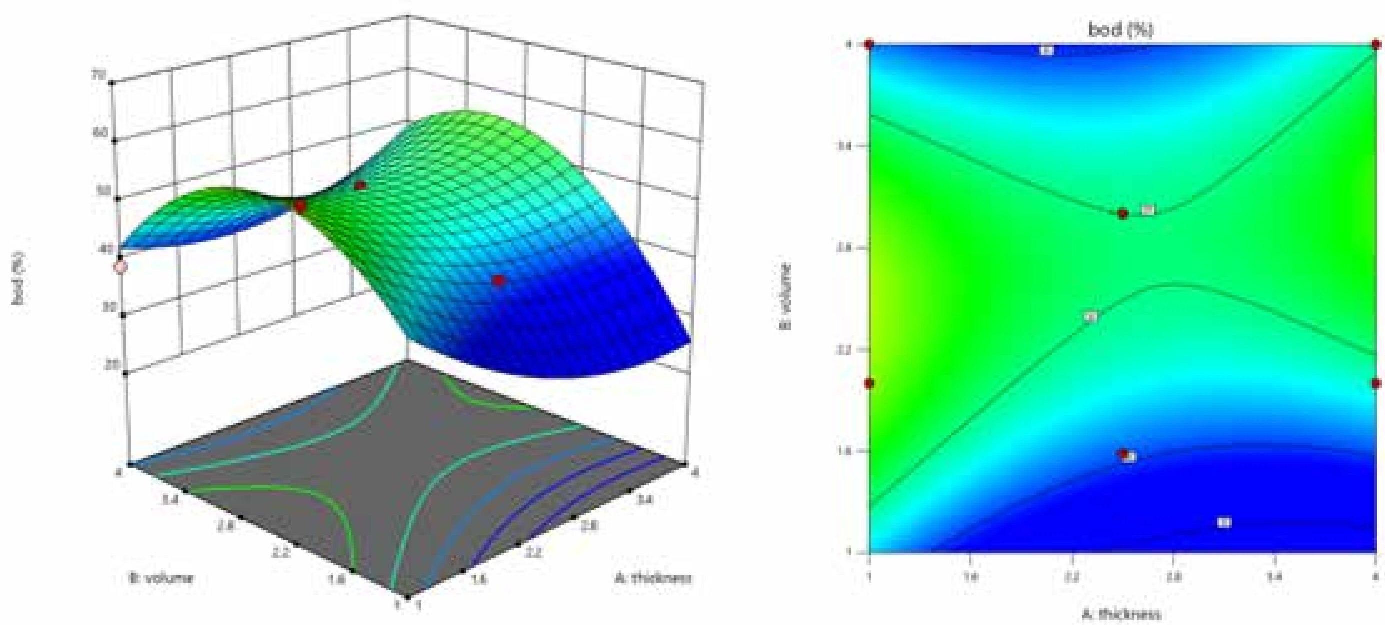

Fig 2. Surface and contour plot for BOD decrease of adsorption using human hair, obtained from RSM studies

experimental results with an R^2 value of 0.788 and an adjusted value of 0.637. With further experiments and significant parameters included, these values could display better results. When measured with adequate precision, the signal-to-noise ratio was found to be greater than 4 as desired. (Burestan N. F. et al., 2021). This ratio compares the range of the predicted values at the design points to the average prediction error. An adequate precision value of 8.23 obtained here indicates that the model can be used to navigate the design space. The RSM studies on the BOD decrease are shown in Table 3.

The model F-value of 5.20 from the above ANOVA Table 3 implies that the model is significant. There is only a 2.60% chance that an F-value this large could occur due to noise. P-values less than 0.05 indicate model terms are significant. In this case, A^2 and B^2 are significant model terms. Values greater than 0.1 indicate that the model terms are not significant.

The final equation of BOD decreases in terms of actual factors obtained from the RSM studies, as shown in the following equation,

$$BOD\ decrease = +38.562 - 23.608 * A + 31.340 * B + 2.95 * A * B + 2.845 * A^2 - 7.052 * B^2$$

The analysis for oil and grease removal suggested that the model best fits the oil and grease experimental results with an R^2 value of 0.862, with an adjusted R^2 value of 0.685. As desired, the signal-to-noise ratio was found to be greater than 4 when measured with adequate precision. This ratio

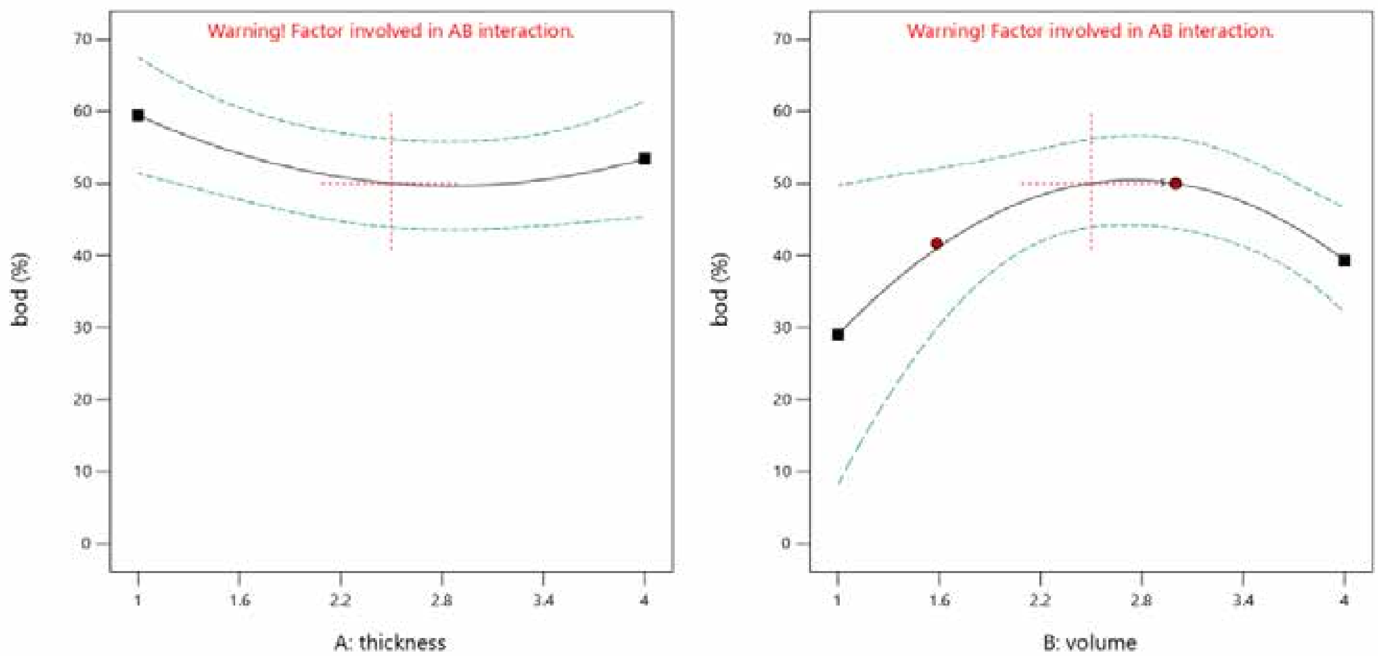

Fig 3. Effect of thickness and volume on BOD decrease by using human hair. Data obtained from RSM studies.

compares the range of the predicted values at the design points to the average prediction error. An adequate precision of 6.39 obtained indicates that the model can be used to navigate the design space. The RSM studies on the percentage decrease of Oil and Grease are shown in Table 4.

The model F-value of 1.62 from the above ANOVA Table 4 implies that the model is insignificant. There is a 26.97% chance that a F-value this large could occur due to noise. P-values less than 0.05 indicate model terms are significant. Values greater than 0.1 indicate that the model terms are not significant.

Interactive effects of various parameters for BOD decrease

The Response Surface Plots help in analyzing the effect of the combination of various parameters on the target parameter. The 3D surface plots are developed with thickness and volume concerning the decrease in BOD.

Figure 2 observes the interaction effect between thickness and volume. The BOD reduction is observed to decrease with increasing thickness. As the volume of the input sample increases, the BOD reduction increases till a certain point and thereafter shows a declining trend. A BOD reduction of 59.36 % was predicted at a thickness of 1 cm and volume of 2.56 liters.

Figure 2 uses the surface and contour plot to show the combined effect of both parameters (thickness and volume) on BOD decrease. Figure 3 shows the effect of both parameters individually on the BOD decrease. In Figure 3, with increasing thickness, a steady decrease in BOD is observed till a certain point, and then we can see a slight increase in its value. This behavior is attributed to a weaker driving force towards adsorption due to the interaction between functional groups of the sample and the adsorbent. Beyond the maximum point, the particles obtain enough energy to attain the driving force needed for adsorption. In terms of volume interaction seen in Figure 3, an initial increase is due to the availability of more adsorption sites, and the decrease is due to saturation of pore sites (Badsha et al., 2021; Ip et al., 2009).

Interactive effects of various parameters for Oil and Grease removal

The Response Surface Plots help in analyzing the effect of the combination of various parameters on the target parameter. The surface plots are developed by varying any two parameters and keeping the other factor constant at its midpoint.

Figure 4 observes the interaction effect between thickness and volume at a constant volume of 7.5 liters. With the combined increase in both the area and the thickness, the oil and grease is initially found to increase and then reduce after a point. A maximum oil and grease reduction of 66.56% was predicted at an area of 42.576% and a thickness of 5 cm.

Figure 4 shows the combined effect of both parameters (thickness and volume) on Oil and Grease reduction, using the surface plot and contour plot. Whereas Figure 5 shows the effect of both parameters individually over the Oil and Grease decrease.

From Figure 5, we obtain the effects of the thickness of the adsorbent and volume of wastewater sample on the Oil and Grease removal percentage. This shows the same trend that is followed above.

As the thickness of the adsorbent layer increases, there is an initial increase in removal due to an increasing concentration gradient between the sample and the adsorbent. There is a decreasing trend observed after the maximum point in the plot, which could be due to low sample seepage through the constructed wetland as a result of sample stagnation over time. In terms of volume's effect on Oil and Grease removal, increasing concentration gradient causes an increase in removal, followed by saturation of pore sites, which initiates a decreasing graph.

Adsorption of Oil and Grease from the wastewater

The adsorption studies of oil and grease on human hair have been examined through various analyses. The percentage adsorption of oil and grease content in the wastewater sample, performed with the adsorbent thickness and sample volume reading combinations as per the BOD studies have been included in Figure 6 given below.

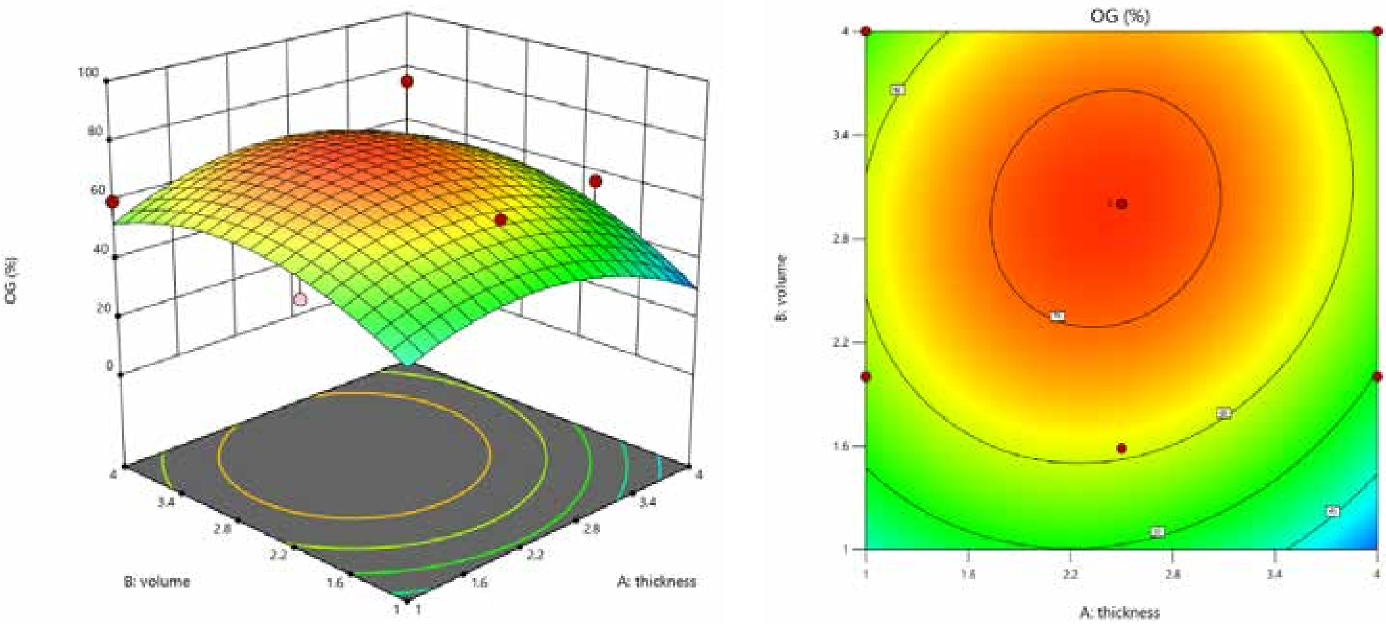

Fig 4. Surface and contour plot for Oil & Grease removal of adsorption using human hair, obtained from RSM studies

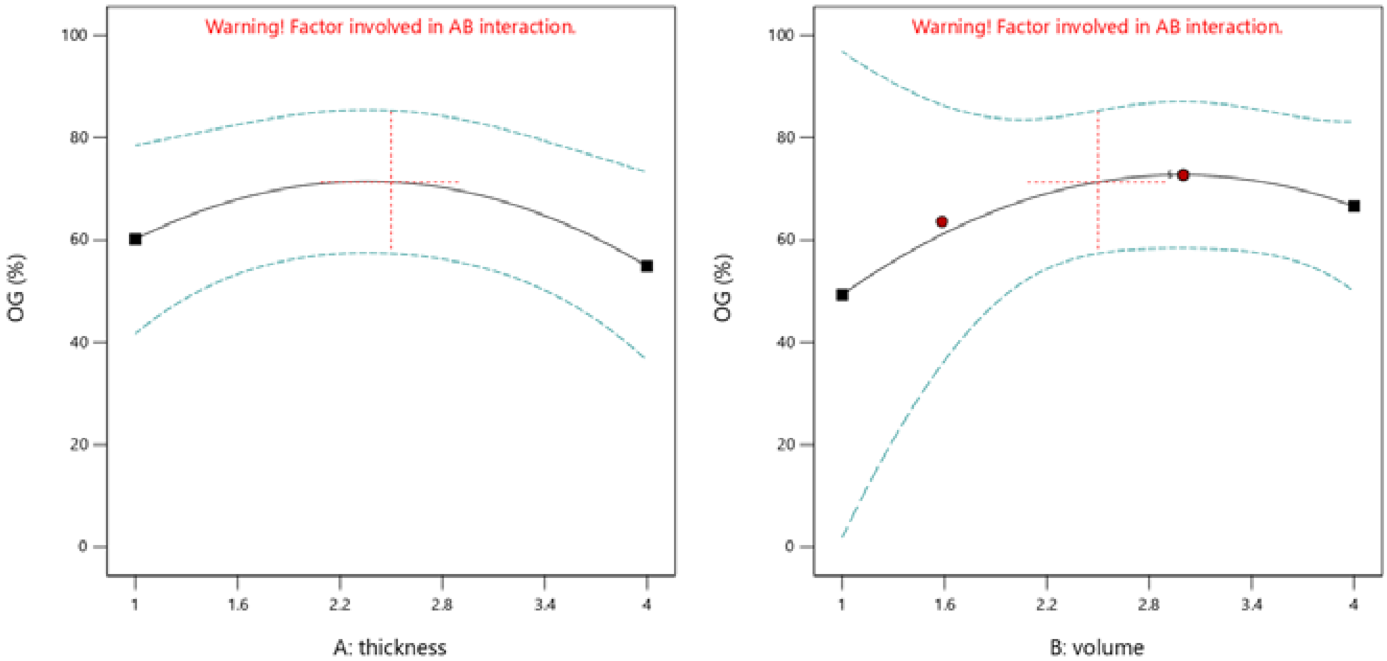

Fig 5. Effect of thickness and volume on Oil & Grease removal by using human hair. Data obtained from RSM studies.

The graph shows that the percentage decrease in oil and grease is considerable in most samples. So, the constructed wetland using human hair substrate is effective in reducing oil and grease content from the kitchen or garage wastewater.

Furthermore, the effectiveness of this is evaluated for BOD reduction in kitchen and garage wastewater. Table 5 shows the BOD of wastewater before and after passing through a human hair substrate. The percentage reduction in BOD is shown in both Table 5 and Figure 7. As can be seen, the reduction in BOD is significant and thus proves that the procedure is effective to be used for preliminary treatment.

Hair is a biopolymer composed largely of cross-linked proteins termed keratins. Human hair can be recovered and reused as an adsorbent, as documented in the following literature. Backwashing of the experimental setup was done, and the adsorbent was recovered and reused. According to the literature, even after 5 times of recovery and reuse, the efficiency of adsorption is not affected. Hence, the same hair can be reutilized as an adsorbent (Ukotije-Ikwut, P. R. et al., 2016; Kumar V. N. et al., 2019).

Conclusion

The bio-adsorbent in consideration, human hair, serves as a good option to considerably reduce the BOD of wastewater samples as well as adsorb the oil and grease in it. The maximum percentage removal in this set of experiments has been mentioned by studying the interactive effects of the parameters over the BOD decrease. While implementing hair for oil adsorption in various applications, its BOD can also be decreased by a significant amount, which could be useful in further treatment of this wastewater.

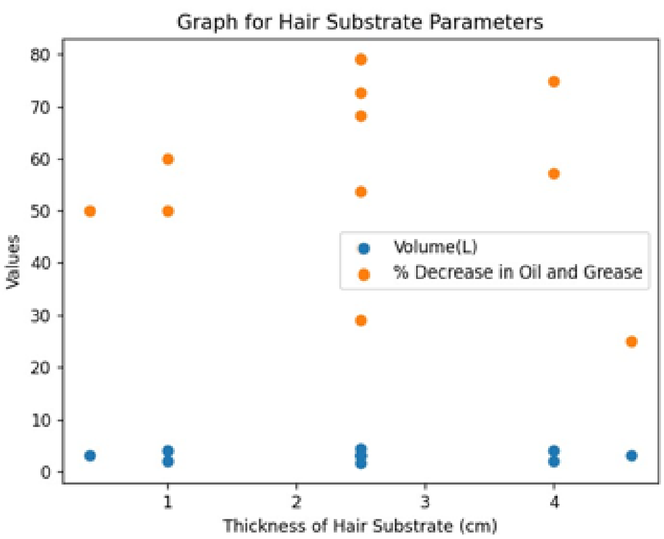

Fig 6. Reduction of oil and grease for different thicknesses of hair substrate and volume of sample in a constructed wetland.

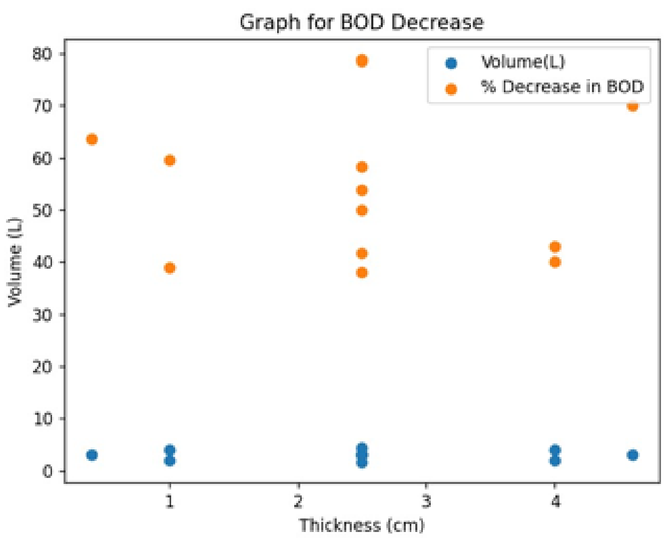

Fig 7. Reduction of BOD for different thicknesses of hair substrate and volume of sample in constructed wetland.

Table 5. Percentage reduction of BOD

Thickness(cm)	Volume(L)	BOD[i] -mg/L	BOD[f] -mg/ L	% BOD dec
1.0	4.0	25.92	15.84	38.89
0.4	3.0	590.4	215.0	63.58
1.0	2.0	590.4	238.6	59.59
2.5	3.0	5.760	2.880	50.00
2.5	1.6	5.760	3.360	41.67
4.6	3.0	28.80	8.640	70.00
4.0	4.0	28.80	17.28	40.00
2.5	4.4	175.2	108.5	38.08
4.0	2.0	175.2	99.84	43.01
2.5	3.0	62.40	28.80	53.85
2.5	3.0	86.40	18.24	78.89
2.5	3.0	62.40	13.44	78.46
2.5	3.0	86.40	36.00	58.33

The use of a human hair substrate for oil separation from water has also been studied by various other studies and found to be effective at the laboratory level. It is observed that the oil and grease are removed up to 79% from the wastewater, effectively for a 2.5 cm thick hair substrate layer and 3-liter wastewater volume. Similarly, BOD was reduced by 78.8% for a 2.5 cm thick hair substrate layer and a 3-liter wastewater volume. Even though the process does not minimize the concentration to micro levels, it still reduces the cost considerably by lowering the amount of demulsifying agents needed. Among other developments to be investigated, multistage arrangements or reverse flow can also be considered for better results.

A major advantage of utilizing human hair as an adsorbent in this regard is its reusability, where with minimal backwashing procedures, the adsorbent can be salvaged without significant loss in its adsorption efficiency. With further studies in this regard, hair as an adsorbent can be extended for oil adsorption beyond its current applications, especially in heavy metal adsorption.

Declaration

– The authors do not have any conflicts of interest to declare.
– The manuscript corresponding to the information mentioned above has been edited by Manipal Universal Press (MUP) for language, readability, grammar and consistency.

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