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Equilibrium Isotherms of Formaldehyde Elimination from the Aqueous Solutions Containing Natural Adsorbents of Rice Bran and the Resulting Ashes

Marzieh Bagheri [a,*] | Mohammad Nasiri [b] | Amirreza Talaiekhozani [c] | Iman Abedi [a]

[a] *Department of Chemical Engineering, Jami Institute of Technology, Isfahan, Iran.*
[b] *Department of Chemistry, Malekashtar University of Technology, Isfahan, Iran.*
[c] *Department of Civil Engineering, Jami Institute of Technology, Isfahan, Iran.*

***Corresponding author:** Marzieh Bagheri
Department of Chemical Engineering, Jami Institute of Technology, Isfahan, Iran, 7134567532. Tell: +98-9125420218.
E-mail address: miss.bagheri40@yahoo.com

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ABSTRACT

Background: In this pioneering work, the comparisons conducted on the elimination efficiency of formaldehyde by rice bran and the resulting ashes utilized in the adsorption process.
Methods: In this study the optimal hydraulic retention time, temperature, pH value and adsorbents dosage for the elimination of formaldehyde using three adsorbents including rice bran, ashes of rice bran produced in 300°C (Carbon-300) and in 500°C (Carbon-500) were determined. The method of one factor at the time was used to optimize the above mentioned factors.
Results: The best adsorption conditions for 1000 mg/L of formaldehyde and 1 g of adsorbent in acidic environment (pH = 4) is reported at 80°C (with elimination percentage of 70%w/w for rice bran, 83%w/w for carbon-300 and 90%w/w for carbon-500). Also it was revealed that the adsorption of formaldehyde by rice bran adsorbent and Carbon-300 is a function of Langmuir adsorption isotherm while the resulting carbon in 500°C is a function of Freundlich adsorption isotherm.
Conclusion: In all experiments, the rice bran ashes showed much greater capacity for formaldehyde removal than one for rice bran. Rice bran is an ideal option in terms availability and the resulting waste could be eliminated through incineration.

1. Introduction

Organic pollutants are among the most frequent environmental contaminants. Formaldehyde is an organic pollutant with toxicity and mutagenicity, which could kill microorganisms [1].

As such, it is difficult to remove formaldehyde from wastewater using biological methods. Moreover, formaldehyde is suspected to have carcinogenic effects on humans.

Formaldehyde has wide applications in various industries, including the production of polymers, adhesives, sponges, explosives materials, textiles, leather, and cosmetics.

Formaldehyde is also used in agriculture, woodworks, and construction [2].

This compound is found in several industrial wastewaters due to its common application.

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Several methods have been proposed for the elimination of formaldehyde, such as advanced chemical oxidation [3-5], electrochemical degradation [6], dielectric plasma discharge [7], biodegradation [8], and adsorption. Adsorption is an effective technique to remove formaldehyde from wastewater. Several studies have denoted that adsorption processes are cost-effective methods to eliminate organic compounds (e.g., formaldehyde) from industrial wastewater [9-11].

Numerous adsorbents have been introduced for the removal of formaldehyde from wastewater, such as active carbon, aluminum oxide [12], human hair [11], sheep wool [9], ceramic materials soaked in potassium permanganate [12], active carbon soaked in manganese dioxide [13], zeolite [14], silted graphite [15], carbonic fibers [16], active carbon modified with hexamethylenediamine [17], carbon activated with amine groups [18], carbon modified by para-benzoic acid [19], and bones [20]. To date, attempts have been made to employ inexpensive natural adsorbents [21, 22]; such examples are bagasse, wood, polyurethane foam, pistachio shells, olives, rice bran, and wheat bran, which are cost-efficient, organic materials.

Low cost, availability, and proper adsorption are among the foremost properties of suitable adsorbents [23]. Recently, agricultural wastes (e.g., rice bran, wheat, barley, rice stalks, wood wastes, and sugarcane bagasse) have been widely recognized as effective natural adsorbents. Rice bran is an agricultural waste, which is a byproduct of the rice mill industry. Approximately 100 million tons of rice bran is produced worldwide every year. It is estimated that roughly 90% of the global production of rice bran is in developing countries. Using rice bran to produce adsorbents could be a proper solution to the disposal problems of rice bran in these countries [23].

Although rice bran has been applied to remove several organic compounds, it has not yet been used for formaldehyde removal from wastewater. The present study aimed to evaluate the effects of pH, temperature, adsorbent dosage, and hydraulic retention time (HRT) on formaldehyde removal from synthetic wastewater using the powder of crude rice bran and its ashes, investigate the effects of the temperature of 300-500°C on the production of rice bran ashes, and assess the adsorption isotherms for crude rice bran and its ashes.

2. Materials and Methods

2.1. Preparation of Adsorbents

Initially, rice bran was meticulously rinsed to wash off the possible contaminants. This was accomplished by washing the rice bran with nitric acid 1 M, followed by rinsing with distilled water repeatedly in order to clear the impurities.

The rinsed bran was preserved at the temperature of 105°C for 24 hours to eliminate its moisture. Afterwards, it was ground and sieved so as to obtain the uniform materials. In the next step, the rice bran was heated for two hours in an electric furnace at the temperature of 300°C and 500°C under conditions with limited oxygen. The resulting ashes were powdered in order to maximize the adsorption capability and stored in a desiccators.

2.2. Investigation of Effective Parameters for Formaldehyde Adsorption

The optimal HRT, temperature, pH, and adsorbent dosages for the elimination of formaldehyde were determined using three adsorbents, including crude rice bran and the ashes of the rice bran produced at the temperature of 300°C (carbon-300) and 500°C (carbon-500).

In the present study, temperature was evaluated within the range of 40-80°C, and the pH values of 3-14 were assessed to find the optimum pH. Moreover, 500 mL of synthetic wastewater contaminated with 1,000 mg/L of formaldehyde was prepared. HRT was investigated for 1-15 hours, and the adsorbent dosages were examined within the range of 0.1-2 g/L. One-factor-at-a-time method was applied to optimize the mentioned factors.

2.3. Analytical Methods

Formalin solution (37% w/w of formaldehyde), sulfuric acid, nitric acid, and sodium hydroxide were procured from Merck Company, Germany. A digital pH-meter (3020, Jenway, UK) was used for pH measurements. A spectrophotometer (DR-5000, Hach, Canada) was used for absorbance measurements of samples. The COD Reactor (16500, Hach, Canada) dry-batch incubator that provided the temperature required in Hatch's test system for COD determination. In order to mix the solutions was used as with a magnetic stirrer (MR3001, Heidolph, Germany). The digital scale by 0.0001 accuracy (AEL-200, Shimadzu, Japan) was used to measure weight. The heater (C-MagHP10, IKA, Germany) with adjustable temperature.

In the present study, formaldehyde was measured based on the chemical oxygen demand (COD) [24]. The concentration of COD was converted into the formaldehyde concentration using Equation 1, as follows:

$$C_{COD} = 1.0667 \times C_F \tag{1}$$

Where C_{COD} is the COD concentration (mg/L), and C_F represents the concentration of formaldehyde (mg/L).

3. Results and Discussion

3.1. Effect of Retention Time on Adsorption Efficiency

The optimal HRT for achieving the maximum pollutant removal efficiency is an important factor in the adsorption process. In order to determine the optimal HRT, the COD of the solution was measured at the HRTs of 1, 2, 5, 10, and 15 hours, and the elimination performance was investigated as well.

COD concentration in the solution containing formaldehyde (1,000 ppm) was 540 mg/L (witness solution).

In order to measure the COD in the formaldehyde solution (1,000 ppm), two milligrams of the solution was transferred to an appropriate vial after it was heated for two hours and cooled prior to measuring the COD using the spectrometer, and the obtained value was 540 mg/L. In the second stage of the experiment, one gram of the rice bran powder and the resulting ashes were separately added to 100 mL of the formaldehyde solution at the concentration of 1,000 mg/L in a volumetric flask (100 mL). The mixture was preserved in a shaker for five minutes at 190 rounds per minute prior to transferring to a beaker.

After two hours, the formaldehyde solution (2 mL) was transferred to a vial. Following that, the vial was kept in a COD digestion reactor and heated at the temperature of 150°C for two hours. The same process was repeated simultaneously with a blank sample with distilled water, in which distilled water (2 mL) was added to a vial and preserved in a COD digestion reactor at the temperature of 150°C for two hours. At the end of the mentioned procedures, the vial was cooled. Afterwards, the sample vial was kept in a cell holder, and the COD of the sample vial was reported in mg/L.

Figure 1 shows that formaldehyde elimination within the range of 10-15 hours was relatively stable, and the rate of adsorption had no change after this period. This could be due to the saturation of the structure of rice bran by formaldehyde. It is recommended that the optimal retention time for the industrial use of formaldehyde be maintained at less than 10 hours since the efficiency of the adsorption cannot be altered beyond this time scale. Figure 1 also depicts that the elimination capacity of the rice bran ashes was higher than rice bran.

3.2. Effect of Temperature on Formaldehyde Adsorption

Temperature is another important influential factor in the efficiency of formaldehyde adsorption. The adsorption process is handled either chemically or physically. High temperature increases the adsorption ability for the chemical adsorption process with an endothermic reaction. The results of this experiment are presented in Figure 2, which indicates that higher temperatures increase the formaldehyde removal capability. Furthermore, the results of temperature effects on the adsorption indicated that adsorption using rice bran and its ashes is of the chemical adsorption process type. However, by increasing the temperature from 40°C to 80°C, there is only 5% increase in the removal capability. Therefore, it could be concluded that temperature was not a paramount parameter in formaldehyde removal in this experiment.

According to the present study, the effects of temperature on the prepared rice bran samples at the temperature of 300°C was higher compared to the other samples. Similarly, the removal efficiency at carbon-300 was higher than rice bran. Moreover, our findings demonstrated that the carbon powders prepared at the temperature of 500°C had higher capability in the removal of formaldehyde from aquatic environments compared to activated carbon-300.

At the temperature of 550ºC, organic compounds are converted into carbon dioxide and water. Therefore, the temperature used for activated carbon production (approximately 550ºC) increases porosity. Higher porosity is associated with higher adsorption sites, which reveals the reason for the higher efficacy of activated carbon-500 in formaldehyde removal.

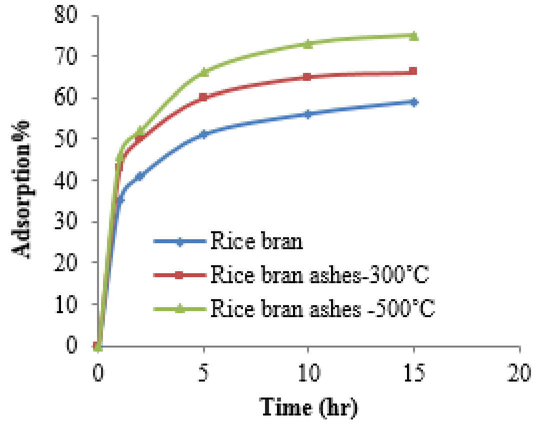


Figure 1: Effect of hydraulic retention time on formaldehyde adsorption by three produced adsorbents (pH of 4; temperature of 40°C and the amount of adsorbent 1 g/L)

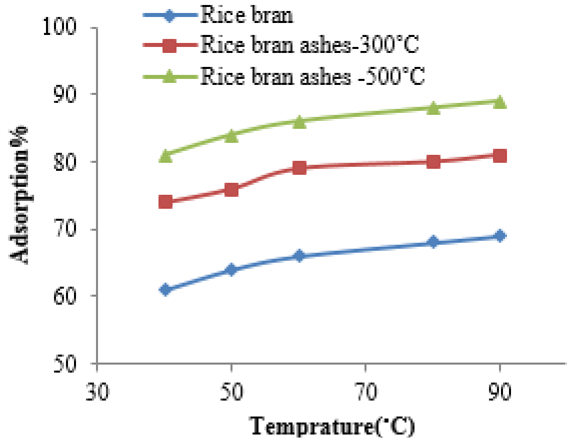
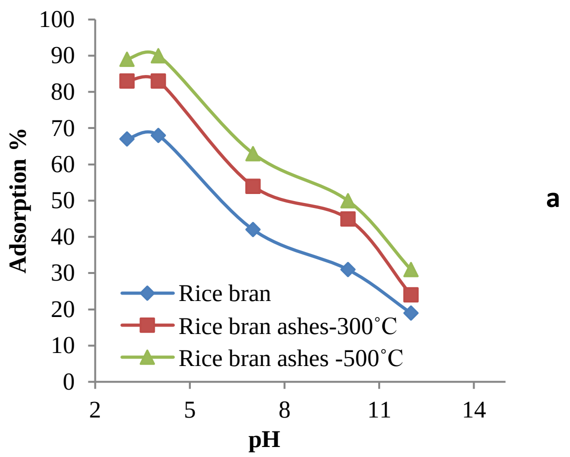


Figure 2: Comparison of temperature effect on the formaldehyde adsorption by three produced adsorbents (pH of 4; the amount of adsorbent 1 g/L and HRT of 10 hr)

3.3. Effect of pH on Formaldehyde Adsorption by Rice Bran and its Ashes

The pH of the applied solution is another important factor to determine the removal efficiency of formaldehyde using rice bran and its ashes. In each experiment, a sulfuric acid solution (0.5 molar) and NaOH (1 molar) was applied in order to adjust the appropriate pH. The pH range shifted between 3, 5, 7, 9, 12, and 14. As can be seen in Figure 3a, the formaldehyde removal rate was higher in acidic environments (pH = 4) for rice bran and its ashes. This could be due to the special structure of carbonyl in formaldehyde, in which the dual connection between carbon and oxygen in the carbonyl group is easily broken in acidic environments and in presence of H^+ ions; as a result, these ions are easily adsorbed by oxygen to form a new C-OH compound. In other words, acidic environments increase the reactivity of the substance, causing it to be easily adsorbed and increase the removal rate.

Hydroxide ions may increase in alkaline environments, and due to the electrophilic properties of the carbonyl group, hydroxide ions possess no capacity to be adsorbed into the carbonyl group. Therefore, the higher rates of formaldehyde could remain in the environment, and the removal efficiency decreases.



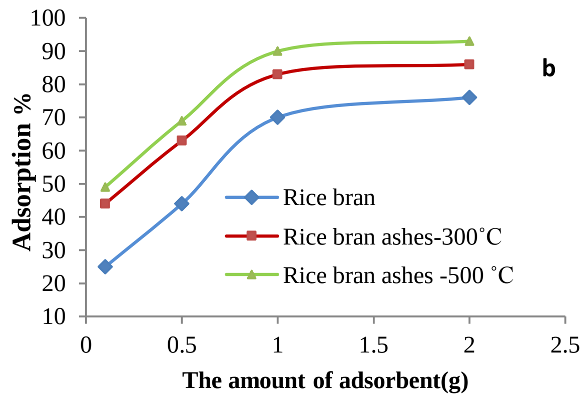


Figure 3: (a) The comparison of pH effects on formaldehyde removal efficiency in the three produced adsorbents (the amount of adsorbent 1 g/L, HRT of 10 hr and temperature 80°C) and (b) The comparison of dosages of three produced adsorbents on the removal efficiency (HRT of 10 hr, temperature 80°C and pH of 4)

3.4. Effect of Adsorbent Dosage on Formaldehyde Adsorption

The adsorbent dosage is considered to be another influential factor in formaldehyde removal from wastewater. At this stage of the study, the application effect of various dosages of rice bran, rice bran ashes (300°C), and rice bran (500°C) was investigated within the range of 0.1-2 g/L. The obtained results indicated that the increased adsorbent dosage from 0.1 to 1 g/L could increase the formaldehyde removal performance, while the increased adsorbent dosage within the range of 1-2 g/L had no significant effect on formaldehyde removal from wastewater (Figure 3b).

According to the results of the present study, the highest and lowest concentrations of formaldehyde were removed by rice bran ashes (500°C) and raw rice bran, respectively. It could be due to the fact that the rice bran ashes (500°C) had a higher specific surface compared to the other adsorbents used in the study. Higher specific surface results in higher formaldehyde adsorption from wastewater. In another research, Talaiekhozani et al. (2011) reported that 16 grams of formaldehyde could be adsorbed on one gram of human hair [11], while in the present study, only 0.9 gram of formaldehyde was adsorbed in the rice bran ashes (500°C).

In the mentioned study, the adsorption of formaldehyde in human hair was a chemical process; therefore, human hair cannot be recovered, while the rice bran ashes (500°C) in our research could be easily regenerated by heating at the temperature of 500°C repeatedly.

3.5. Adsorption Isotherms

In the current research, several isotherm models were used to analyze the experimental data and describe the adsorption balance, including the Freundlich and Langmuir adsorption isotherms. These models were applied to provide perspectives on the adsorption mechanisms, surface properties, and adsorbent affinity and explaining the experimental data. In the Langmuir isotherms, adsorption occurs on one layer and is positional, resulting in an optimal adsorption reaction. The reaction heat is independent of the deductible coverage and equal across the adsorption surface.

The adsorbent surface possesses certain amounts of adsorption sites. The linear type of the Langmuir equation is derived from Equation 2, as follows:

$$C_e/q_e = [1/bq_{max} + C_e/q_{max}] \quad (2)$$

Where q_e is the equilibrium adsorption capacity, C_e is the equilibrium concentration, q_{max} represents the maximum adsorption capacity, and b is the correlation-coefficient.

In case the Freundlich isotherm was achieved by the assumption of a heterogeneous surface with an uneven distribution of adsorption heat, we used equations 3 and 4, as follows:

$$q_e = K_f\, C_e^{1/n} \quad (3)$$

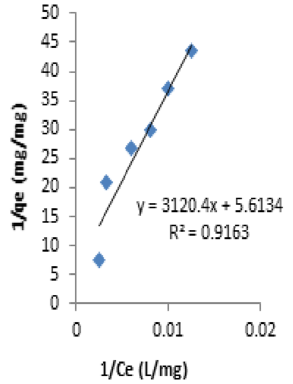
$$Logq_e = LogK_f + 1/n\ \log C_e \quad (4)$$

Where K_f and n are the Freundlich constants, which are dependent on the adsorption capacity and the adsorption intensity, respectively [25].

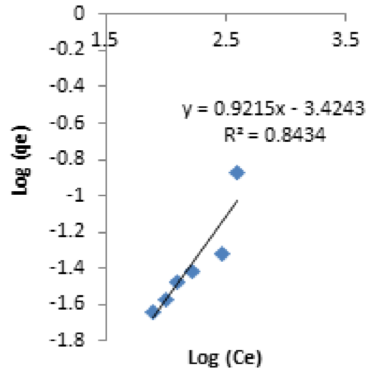
At this stage, the adsorption isotherms were investigated in association with the three adsorbents in order to identify the type of the isotherm each adsorbent followed.

Figures 4a and 4b depict the Langmuir and Fruendlich isotherms for the rice bran, respectively.

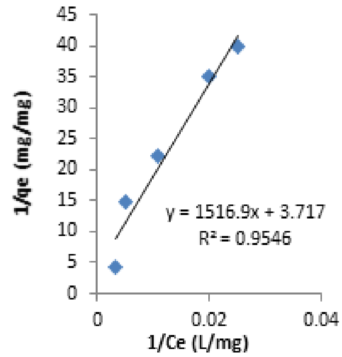
Figures 4c and 5d illustrate the Langmuir and Fruendlich isotherms for carbon-300, respectively. Figures 4e and 4f show the Langmuir and Fruendlich isotherms for carbon-500, respectively. The Langmuir constants (q_m and b), and Freundlich constants (K_f and n) were calculated for the mentioned adsorbents, and the obtained values are presented in Table 1. The respective correlation-coefficients (R^2) of both the isotherm models are also presented in Table 1. The aforementioned experiments were carried out at the pH of 4, HRT of 10 hours, and adsorbent dosage of one gram.



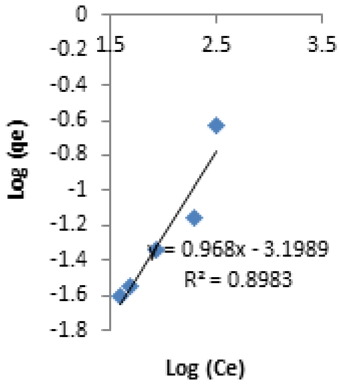
(a) The Langmuir isotherm graph relating to rice bran adsorbent



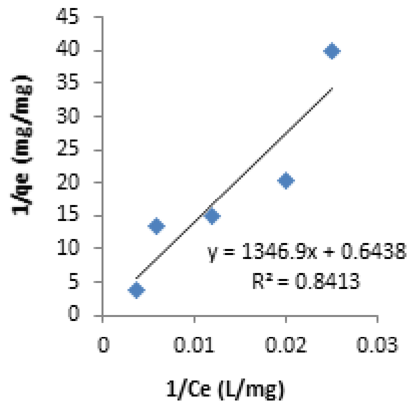
(b) The fruendlich isotherm graph` relating to rice bran adsorbent



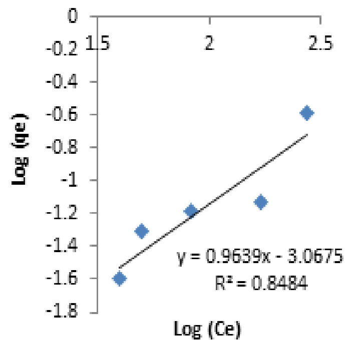
(c) The isotherm graph of langmuir for carbon-300



(d) The isotherm graph of fruendlich for carbon-300



(e) The isotherm graph of langmuir for carbon-500



(f) The isotherm graph of fruendlich for carbon-500

Figure 4: The isotherm graphs of Langmuir and Fruendlich for different produced activated carbons

Based on the calculated correlation-coefficients, the Freundlich isotherm was more suitable for the estimation of formaldehyde removal from wastewater using rice bran and carbon-500 compared to the Langmuir isotherm (Table 1). However, the Langmuir isotherm was more suitable for the estimation of formaldehyde removal from wastewater using carbon-300. Since the amount of formaldehyde removal for all the produced adsorbents was observed to increase at higher temperatures, the removal of formaldehyde is speculated to be a chemical adsorption. Therefore, the three adsorbents could be considered as new and efficient agents due to their high elimination percentage of formaldehyde. Therefore, the three adsorbents could be considered as new and efficient agents due to their high elimination percentage of formaldehyde.

We attempted to present a comparison between the other studies regarding formaldehyde removal using other adsorbents with our findings. According to the information in Table 2, the adsorbent utilized in the present study possessed a high capacity in the removal of formaldehyde.

Table1: Constants parameters and correlation coefficients (R^2) calculated for synthesized adsorbents

	Langmuire			Freundlich		
Adsorbent	**q_m**	**b**	**R^2**	**K_f**	**n**	**R^2**
Rice Bran	0.178	0.00179	0.9163	0.000376	1.085	0.8434
Carbon-300	0.269	0.00245	0.9456	0.000633	1.033	0.8983
Carbon-500	1.55	0.00478	0.8413	0.000856	1.037	0.8484

Table 2: Comparison of the present work with other works

No	Adsorbent	Formaldehyde (mg/L)	The amount of adsorbent dosage (g)	Adsorption (%)	Ref
1	Granular activated carbon loaded with MnO_2	1.8	1.0	72.1	[13]
2	Natural zeolite	2.0	2.0	2.0	[14]
3	Graphite oxide silylated by 3 Aminopropylmethyldiethoxysilan	2.14	1.5	78	[15]
4	Bone char (BC) modified with aceticacid	200	100	98	[20]
5	Silver nano-particles attached to granular activated carbon	19.0	8.0	70	[26]
6	Human hair	24000	12.5	67	[11]
7	Sheep wool	27000	2.0	98.4	[9]
This work	Charcoal-500 derived from rice bran	1000	1.0	90	-
This work	Charcoal-300 derived from rice bran	1000	1.0	83	-
This work	Rice bran	1000	1.0	70	-

4. Conclusion

According to the experimental data, the formaldehyde adsorption by rice bran and carbon-300 followed the Langmuir isotherms, whereas the adsorption of white powder-500 followed the Fruendlich isotherms. The optimal conditions for the elimination of formaldehyde were identified as acidic environment (pH = 4) and temperature of 80°C. Therefore, rice bran and its ashes could be used as an efficient adsorbent for the elimination of formaldehyde from industrial wastewater. Furthermore, increased temperature from 300°C to 500°C could produce carbon with better activation and higher formaldehyde removal efficiency. In all the experiments, the rice bran ashes showed greater capacity for formaldehyde removal compared to rice bran. As such, it could be concluded that rice bran is an inexpensive adsorbent compared to the adsorbents that are currently utilized.

Author contribution

In this study the experimental design, data analysis and manuscript writing were conducted by M. B., M. N., and A.R. T. Also, the experiments were carried out by I. A. All authors revised and approved the final manuscript.

Conflict of Interest

Authors are aware of and comply with the best practice in publication ethics, specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of the figures, competing interests, and compliance with the policies on research ethics.

Additionally, the authors declare no conflicts of interest that would prejudice the impartiality of this scientific work.

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