Kinetics and Thermodynamics of Adsorption of Basic Blue 41 on Used Black Tea Leaves

Mohammad Abul Hossain*, Md Mohibullah

Abstract— This study presents the feasibility of used black tea leaves (UBTL) as a low cost adsorbent for the adsorptive removal of Basic Blue 41 (BB41), a common pollutant in the textile waste water. In batch process, the effects of concentration and temperature on the removal process were investigated. Experimental results were evaluated using different kinetic model equations. At an optimum pH of 2.0, the adsorption followed pseudo second order rate equation for different initial concentrations. The equilibrium amount adsorbed was recorded as 23.5 $mg \cdot g^{-1}$ at 30 ^{o}C which increased with increasing temperature. Calculated activation energy and the thermodynamic parameters suggested that the adsorption of BB41 is endothermic, spontaneous and physical in nature. The study revealed that the UBTL as a potential adsorbent for effective removal of BB41 from aqueous solution.

Key words— Basic Blue 41, adsorption, used black tea leaves, pseudo second order kinetics and endothermic process.

1 Introduction

Environmental pollution, especially in water bodies, has become a matter of growing concern now-a-days. Industrial waste water containing deleterious constituents are very often directly discharged into the water bodies without proper processing. Such practices are widespread mainly due to the huge costs involved in the waste treatment. Rapid growth in industrial sector is playing a vital role in the economical development of Bangladesh and many other developing countries. Bangladesh is the second largest ready-made garments exporter in the world. To support this sector, a large number of textile industries have grown up in Dhaka city and nearby areas. Manufacturers from these industries are dumping untreated waste waters into local streams and rivers, thereby causing detrimental effects to the environment and public health. A pollution assessment carried out by IWM found that industrial sources, notably the textile industry, tanneries, and the pharmaceutical industry, were the largest contributors to pollution in the Dhaka watershed [1]. It is estimated that about 280,000 tons of dyes are discharged annually in effluents from textile dyeing industries [2]. Wastes from different chemical processes are being discarded by most convenient routes, which usually stack down the drain or onto the ground. These practices are resulting in carcinogenesis [3], bio-toxicity and bioaccumulation [4], eventually causing loss of soil productivity, interfering in self purification process of water, killing fishes, declining bird populations, deforming the shape of animals.

Azo dyes like Basic Blue 41 are largely used in textile industries because of their low cost, stability and variety of colors [5]. However, their low degree of fixation on the fiber results in the release of substantial amounts of the dye in the wastewater [6]. Moreover, they are biologically nondegradable because of their aromatic structure. The treatment of highly colored waste water containing hazardous industrial chemical effluent is therefore one of the growing needs of the present time. Chemists have developed means of directing chemical science towards the removal of pollution and thereby improving the environment.

There are many technological methods to remove deleterious dyes from industrial wastewater before discharging them into water bodies. The most important physical, chemical and biological techniques includes adsorption [7, 8], membrane filtration [9], photo-degradation [10], advanced oxidation [11], coagulation [12], ion-exchange [13] and biodegradation [14]. However, most of the techniques mentioned above are not eco-friendly and economically feasible due to the huge costs involved in their operation [15]. Among the available options, adsorption is widely regarded as the best process to remove dyes from aquatic system when low cost adsorbent is used [16, 17]. Apart from the economic point of view, this technique edges others in term of simple designs and land required [18].

Again, many adsorbents, being costly, have limited application especially in developing countries. Research works are therefore going on to find out effective alternatives. In present study, used black tea leaves (UBTL) were considered as low cost adsorbents. With an annual black tea leaves production of about 3.5 million tons and consumption of about 3.25 million tons globally [19]. The UBTL is not only cheap and readily available but also environmentally friendly as it recycles waste products [20]. Moreover, high adsorption capacity of used black tea leaves as a low cost adsorbent to Cr (VI) was report-

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ed [21]. The effectiveness of UBTL was found in its unique properties including the high content of carbon, high porosity, and reactive functional sites [22].

Basic Blue 41 (BB41), a water soluble cationic dye, is one of the most prospective dyes used in textile industries [23]. Here the adsorption kinetics and thermodynamic studies were carried out to investigate the efficiency of the adsorptive removal of BB41 from aqueous system by UBTL under different experimental conditions such as the effect of concentration and temperature in batch process.

2 Experimental

2.1 Chemicals

All chemicals used in the study were analytical grade. Basic Blue 41 (BB-41) of analytical grade was purchased from Sigma-Aldrich and was used without further purification. It is a deep blue colored basic dye which appears as dry powder. It's IUPAC name is 2-[N-ethyl-4-[(6-methoxy-3-methyl-1,3-benzoth-iazol-3-ium-2-yl)-diazenyl]-anilino]-ethanol; methyl sulfate. Its molecular formula is $C_{20}H_{26}N_4O_6S_2$ and formula weight is 482.57 g·mol^{-1}. The dye is soluble in water. The structural formula of Basic Blue 41 is given in Fig. 1.

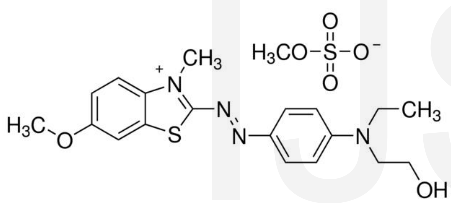

Fig. 1. Structural formula of Basic Blue 41.

2.2 Adsorbent

Fresh black tea leaves were collected from Dhaka City in Bangladesh. About 500 g fresh black tea leaves were added to about 700 mL of distilled water and were boiled for about 6 hours. Boiled tea leaves were washed in several times by hot distilled water followed by cold distilled water until the tea color was completely removed. The colorless solid was dried in an oven at 105 °C for 10 hours. The tea leaves were then sieved through the metallic sieve of mesh size (212 - 300 μm) and screened out. Prepared used black tea leaves (UBTL) was kept in an air tight bottle. The nature of prepared UBTL surface was investigated by Scanning Electron Microscope (JSM-6490LA, JEOL, Japan). Heterogeneous surface of the prepared UBTL is presented in Fig. 2.

2.3 Analysis of Adsorbate

A stock solution of 1000 mg·L^{-1} was prepared by dissolving required amount of BB41 in distilled water. A number of standard solutions were prepared from the stock solution at optimum pH 6.0 and their absorbance values were recorded using UV-visible spectrophotometer (UV-1650A, Shimadzu, Japan) at a pre-determined λ_{max} of 608 nm. The absorbance were then plotted against the corresponding concentrations to construct a calibration curve from which absorption coefficient was found as 0.160 mg·L^{-1}·cm^{-1} and the range of concentration was indicated as 0.1 to 30 mg·L^{-1}.

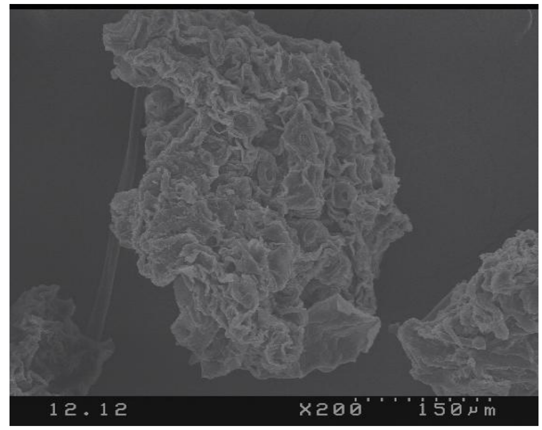

Fig. 2. SEM microgram of prepared UBTL (×200).

2.4 Adsorption Experiments

In case of adsorption at solid-liquid interface, pH of solution is very important factor. Preliminary experiments were performed to investigate the variation of solution pH with initial pH of solution during the adsorption of BB41 on UBTL and the optimum initial pH of solution for adsorption of BB41 on UBTL was selected at about 2.0 where the minimum change of solution pH (± 0.05) was recorded during adsorption.

Effect of concentration: Effect of adsorbat concentration on adsorption kinetics was performed in batch process. A fixed concentration of BB41 solution was adjusted at pH 2.0. Each of 25 mL solution was taken in 5 different bottles containing 0.0025 g of UBTL and all the bottles were shaken in a thermostatic mechanical shaker at 30±0.2 °C. After different time of intervals, solution of each bottle was separated. After proper dilution and adjusted pH at 6.0, their absorbance was measured at λ_{max} = 608 nm using UV-visible spectrophotometer (UV-1650A, Shimadzu, Japan). The values were converted into concentrations using the calibration curve. The initial concentration of the solution was determined the same analytical procedure. Similar kinetic experiments were performed for different concentrations of BB41 solution using same pH 2.0 and temperature at 30±0.2 °C.

Effect of temperature: To determine the effect of temperature on the adsorption kinetics, kinetic experiments were carried out at three different temperatures using initial concentration of about 100 mg·L^{-1} of BB41 solution at pH 2.0, keeping other parameters constant. The change of concentrations of BB41 with time for different temperatures was estimated using same analytical method descrived previously.

3 Results and Discussions

3.1 Effect of Concentration

Adsorption kinetics of BB41 on UBTL was studied by investigating the change of concentration of BB41 with time for different initial concentrations where other conditions were fixed. The amount adsorbed of BB41 ($q_t = x/m$) on UBTL at different contact times for different initial concentration was calculated from the following equation:

$$q_t = (C_o - C_t) \times \frac{V}{m} \tag{1}$$

where, C_o is the initial concentration of BB41 mg·L^{-1}, C_t is the concentration of BB41 at time t (mg·L^{-1}), q_t is the amount adsorbed at time t (mg·g^{-1}), V is the volume of solution in liter and m is the mass of adsorbent in g.

The change of concentrations of BB41 solutions with contact time for different initial concentrations is presented in Figure 3 (a & b) which shows that the concentration of BB41 at various initial concentrations gradually decreased with time. As the time passed, BB41 accumulated on the surfaces of UBTL, so the concentration of BB41 in solution decreased.

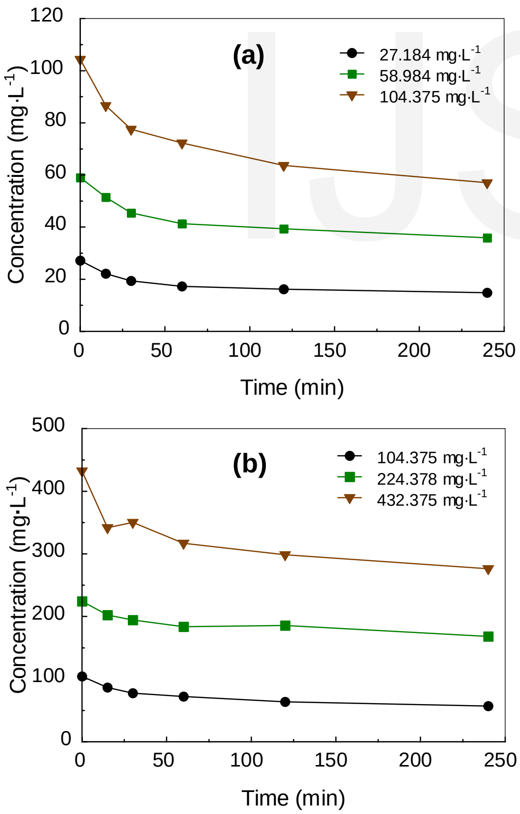

Fig. 3(a & b). Variation of the concentration of BB41 with time for different initial concentrations of BB41 at pH 2.0 and at 30 ± 0.2 °C for different initial concentrations.

3.2 Analysis of Adsorption Kinetics

The kinetics of the adsorption was analyzed using different kinetic equations as follows:

Simple first order kinetic equation: The first order kinetic equation [24, 25] is expressed as follows:

$$\ln C_t = -k_1 t + \ln C_o \tag{2}$$

where, C_o and C_t are the concentrations (mg·L^{-1}) of BB41 at times t_0 and t respectively, k_1 is the first order rate constant. The verification of the equation (2) for the adsorption of BB41 on UBTL was done by plotting lnC_t vs. time as presented in Fig. 4 (a & b). The figure shows that the adsorption kinetics of BB41 on UBTL doesn't follow the simple first order kinetics satisfactorily at any concentration.

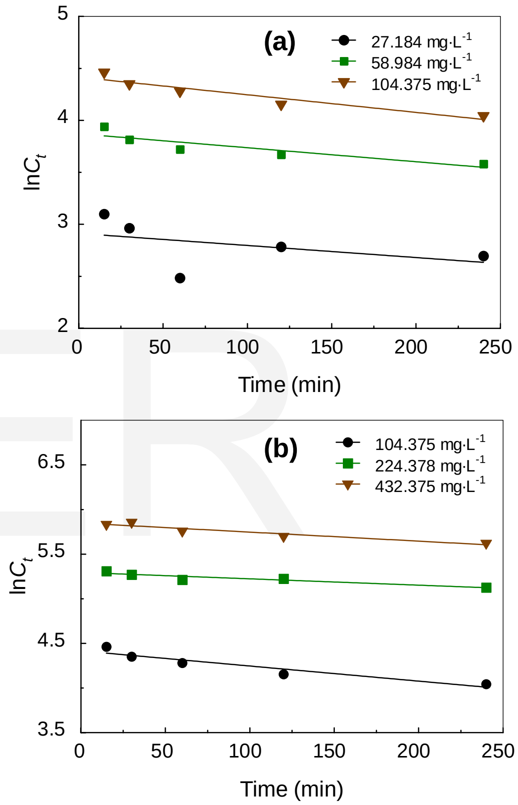

Fig. 4(a & b). Application of the first order kinetic equation for the adsorption of BB41 on UBTL at pH 2.0 and at temperature 30 ± 0.2 °C for different initial concentrations.

Simple second order kinetic equation: The second order kinetic equation [24, 25] is expressed as follows:

$$\frac{1}{C_t} = k_2 t + \frac{1}{C_o} \tag{3}$$

where, C_o and C_t are the concentrations (mg·L^{-1}) of BB41 at times t_o and t respectively, k_2 is the second order rate constant. Plots of 1/C_t vs. t are presented in Fig. 5 (a & b) which indicates that adsorption of BB41 on UBTL doesn't follow second order kinetics satisfactorily at lower concentration but comparatively better fitness is observed at higher concentration.

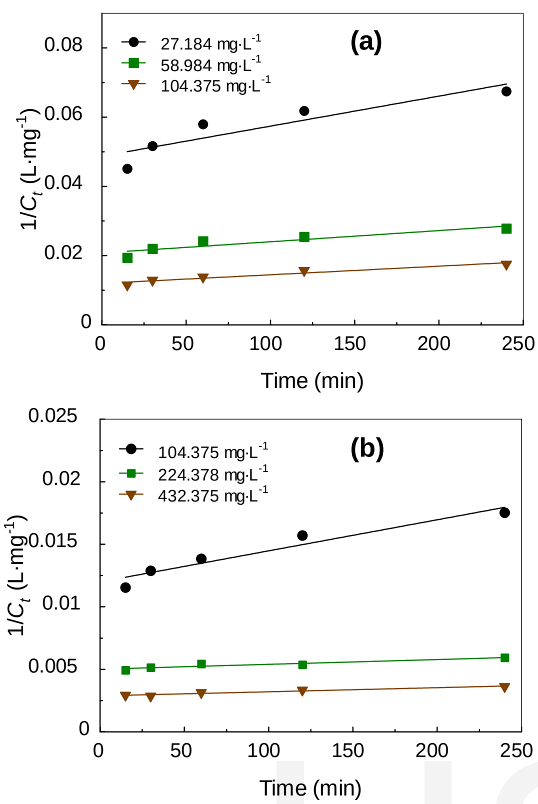

Fig. 5(a & b). Application of the second order kinetic equation for the adsorption of BB41 on UBTL at pH 2.0 and at temperature 30 ± 0.2 °C for different initial concentrations.

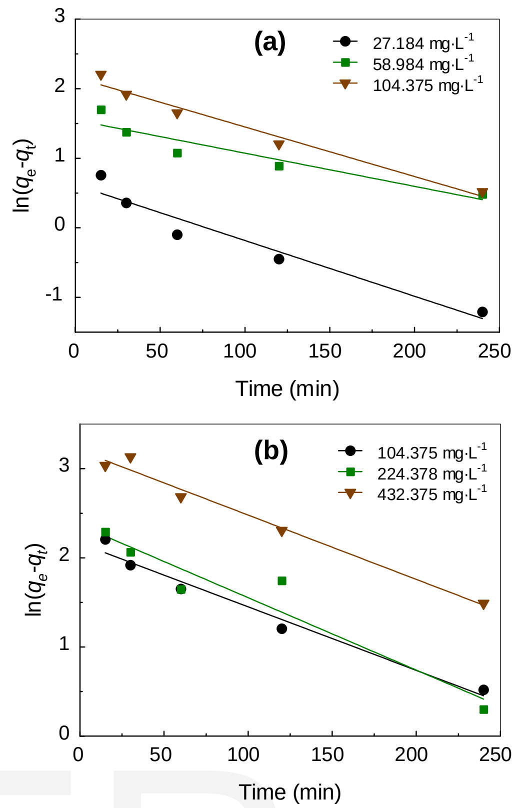

Fig. 6(a & b). Application of the pseudo first order kinetic equation for the adsorption of BB41 on UBTL at pH 2.0 and at temperature 30 ± 0.2 °C for different initial concentrations.

Lagergren pseudo first-order kinetic equation: Integrated form of the pseudo first-order equation [26] is expressed as follows:

$$\ln(q_e - q_t) = \ln q_e - k_{1p} t \tag{4}$$

where, q_e and q_t are the adsorption capacity at equilibrium and at time *t*, respectively (mg·g^{-1}), k_{1p} is the rate constant of pseudo first order equation (L·min^{-1}). Pseudo first order equation (4) was verified by plotting ln(q_e-q_t) vs. *t* as presented in Fig. 6(a & b). The experimental data of the adsorption follow pseudo first order kinetics at higher concentration but at lower concentration it shows irregularity.

Pseudo second order kinetic equation: Integrated form of the pseudo second order rate equation [27] can be represented as follows:

$$\frac{t}{q_t} = \frac{1}{k_{2p} q_e^2} + \frac{1}{q_e} t \tag{5}$$

where, q_t and q_e (mg·g^{-1}) are the amount adsorbed at time, *t* and equilibrium amount adsorbed respectively. k_{2p} is pseudo second order rate constant (g·mg^{-1}·min^{-1}). The equation was also applied to the present system by plotting t/q_t vs. *t* as shown in Fig. 7(a & b). This figure shows that each plot gives straight line with the whole range of concentration indicating the best fitness of pseudo second order rate equation for the adsorption of BB41 on UBTL in both at low and high concentration of BB41.

Intra-particle diffusion model: Intra-particle diffusion model is very important because the internal diffusion determines the adsorption rate in most of the liquid systems. Equation (6) is a general representation of the kinetics, where the intercept is related to the mass transfer across the boundary layer and the expected value of the exponent is 0.5 (for Fickian diffusion and plate geometry).

$$q = k_{in} t^{n} + c \tag{6}$$

Weber and Morris [28] proposed an intra-particle diffusion model (Eq. 7) which describes the time evolution of the concentration in adsorbed state, where the rate constant (k_{in}) is obtained from the slope of the plot of q_t versus $t^{0.5}$ and is related to the respective intra-particle diffusion coefficient (*D*) according to the equation (8).

$$q_t = k_{in} t^{0.5} \tag{7}$$

$$k_{in} = 6 \frac{q_o}{R} \sqrt{\frac{D}{\pi}} \tag{8}$$

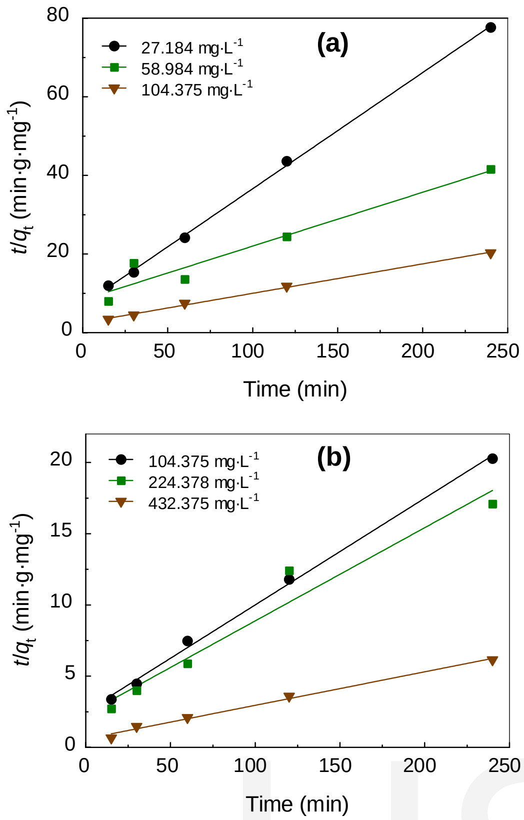

Fig. 7(a & b). Application of the pseudo second order kinetic equation for the adsorption of BB41 on UBTL at pH 2.0 and at temperature 30 ± 0.2 °C for different initial concentrations.

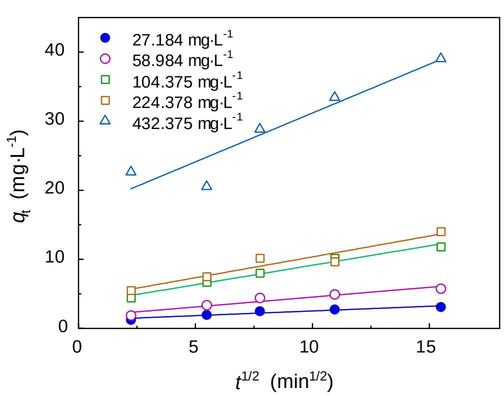

Fig. 8. Application of the intra-particle diffusion model for the adsorption of BB41 on UBTL at pH 2.0 and at temperature 30 ± 0.2 °C for different initial concentrations.

Figure 8 shows the linear plots of the q_t vs. $t^{1/2}$ for each initial concentration of BB41 for its adsorption on UBTL with less values of co-relation of coefficient (R^2), presented in Table 1 to compare with other kinetic equations.

Thus the experimental data are verified to first order, second order, pseudo first order, pseudo second order kinetic equations and intra-particle diffusion model but the data fitness based on the value of correlation coefficient, shows that the pseudo second order rate equation is well fitted over other equations. Well expression of intra-particle diffusion model for the adsorption of Malachite Green (MG) on UBTL has been reported elsewhere [29].

TABLE 1

A COMPARISON OF THE DATA FITNESS TO THE FIRST ORDER, SECOND ORDER, PSEUDO FIRST ORDER, PSEUDO SECOND ORDER KINETICS EQUATIONS AND INTRA-PARTICLE DIFFUSION MODEL

Initial conc. C_0 (mg·L^{-1})	First order R^2	Second order R^2	Pseudo first order R^2	Pseudo second order R^2	Intra-particle diffusion R^2
27.184	0.499	0.828	0.937	0.999	0.923
58.984	0.796	0.838	0.885	0.936	0.932
104.375	0.896	0.930	0.974	0.997	0.983
224.378	0.874	0.877	0.918	0.951	0.922
432.375	0.910	0.929	0.981	0.991	0.890

3.3 Effect of Temperature on Adsorption Kinetics

To investigate the effect of temperature on the adsorption kinetics of BB41 on UBTL, batch kinetics experiments were carried out at different temperatures using same concentration of BB41 solution at pH 2.0. Figure 9 shows the variation of amount adsorbed with contact time for different temperatures. The figure shows that, equilibrium amount adsorbed increases with increase of temperature suggesting the endothermic nature of adsorption. This can be explained by the fact that as the temperature increases the number of molecule acquired activation is also increases [21] due to the increased velocity of the BB41 molecules.

Pseudo second order kinetics equation was verified by plotting t/q_t vs. t at different temperatures as shown in Fig. 10. The equilibrium amount adsorbed, calculated from pseudo second order kinetic plot, linearly increased with the increase of temperature as shown in Fig. 11, which indicated endothermic nature of the adsorption. Figure 12 shows that the

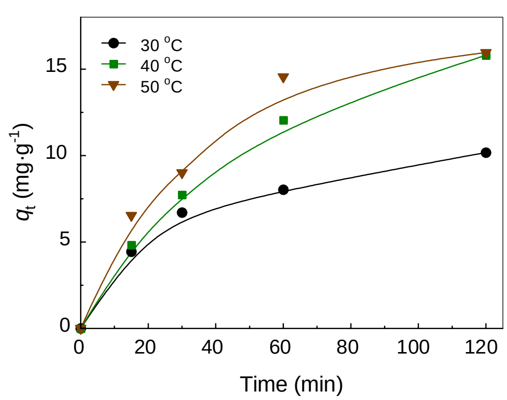

Fig. 9. Change of the amount adsorbed of BB41 with time for a fixed initial concentration at pH 2.0 for different temperatures.

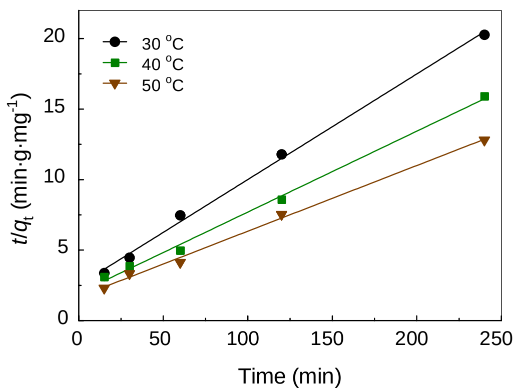

Fig. 10. Application of pseudo second order kinetics equation for the adsorption of BB41 with time for a fixed initial concentration at pH 2.0 for different temperatures.

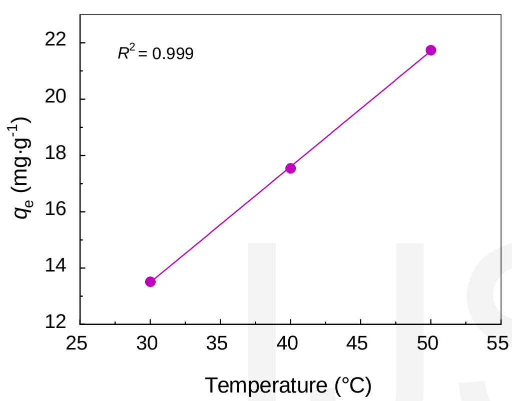

Fig. 11. Variation of equilibrium amount adsorbed with temperature for adsorption of BB41 on UBTL at pH 2.0.

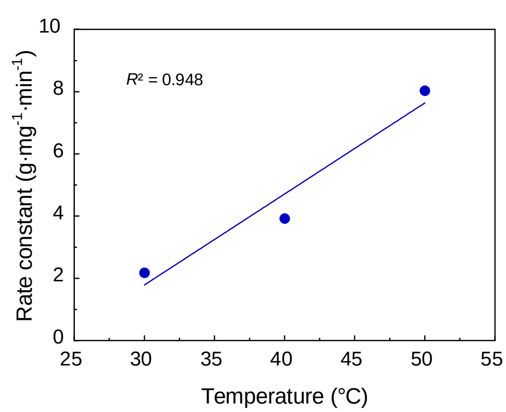

Fig. 12. Variation of pseudo order rate constant, *k* with temperature for adsorption of BB41 on UBTL at pH 2.0.

pseudo order rate constant, *k* increased with increase of temperature. Apparent activation energy was calculated from the plot of ln*k* versus 1/*T* as shown in Fig. 13.

3.4 Adsorption Thermodynamics

Thermodynamic parameters such as standard Gibbs free energy ΔG^o, enthalpy ΔH^o and entropy ΔS^o were calculated by

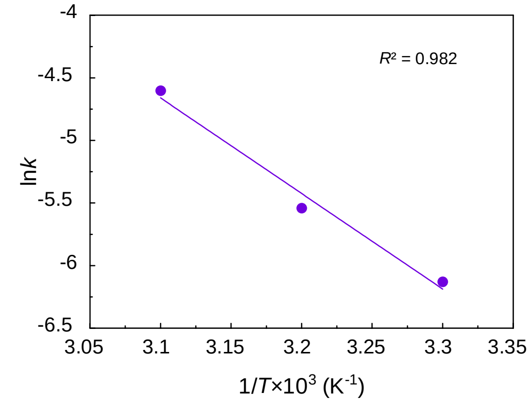

Fig. 13. A plot of ln*k* versus 1/*T* for apparent activation energy.

applying the following equations [30],

$$K_c = \frac{C_{ae}}{C_e} \quad (1.6)$$

$$\Delta G^o = -RT \ln K_c \quad (1.7)$$

$$\log K_c = \frac{\Delta S^o}{2.303R} - \frac{\Delta H^o}{2.303RT} \quad (1.8)$$

where, K_c = the equilibrium constant, C_e = the equilibrium concentration of BB41 in solution (mg·L^{-1}), C_{Ae} = the solid-phase concentration of BB41 at equilibrium (mg·L^{-1}), *R* = the molar gas constant (8.314 J·mol^{-1}·K^{-1}) and *T* = temperature (K). The values of ΔH^o and ΔS^o were calculated from the slope and intercept of the plot of logK_c versus 1/*T* as shown in Fig. 14. The calculated values of enthalpy (ΔH^o) and entropy (ΔS^o) changes are 64.2 kJ·mol^{-1} and 0.25 kJ·mol^{-1}·K^{-1}, respectively, presented in Table 2. The positive value of enthalpy change suggested that the adsorption is endothermic in nature. Again the negative values of ΔG^o from kinetic experiment indicate that the system is spontaneous. The shaking process and the normal condition of temperature are enough to supply the energy required for the adsorption process.

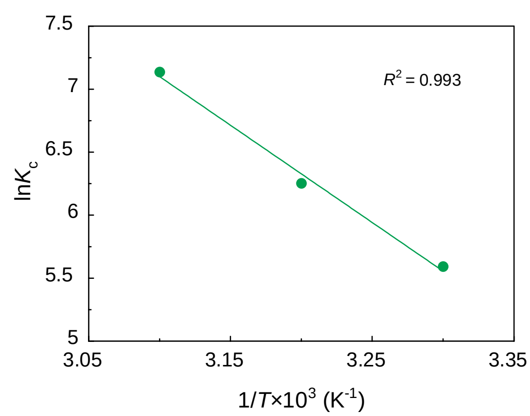

Fig. 14. A plot of lnK_c versus 1/*T* for determination of the change of enthalpy and entropy of the adsorption.

TABLE 2
THERMODYNAMIC PARAMETERS OF BB41 ADSORPTION ON UBTL

Temperature (K)	K_c	ΔG^o ($kJ\cdot mol^{-1}$)	ΔH^o ($kJ\cdot mol^{-1}$)	ΔS^o ($kJ\cdot K^{-1}.mol^{-1}$)
303	1.074	- 0.178		
313	2.078	- 1.902	64.2	0.25
323	5.028	- 4.337		

4 CONCLUSION

Heterogeneous surface of the used black tea leaves, UBTL contains large number of cavities which promotes the adsorption in a suitable manner. The pH of Basic Blue 41 solution plays an important role in the adsorption process and pH 2.0 is suitable for this adsorption. Adsorption kinetics of BB41 on UBTL follows the pseudo second order kinetic equation. The equilibrium amount adsorbed increasing with increase of temperature suggesting the endothermic nature of adsorption. The negative values of ΔG^o from kinetic experiment indicated that the adsorption is spontaneous.

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REFERENCES

[1] The World Bank Report, "The Bangladesh Responsible Sourcing Initiative: A New Model for Green Growth," vol. 14, 2014.

[2] J. X. Liu, G. X, ZH, T. WY, "Decolourisation of a Dye Industry Effluent by Aspergillus Fumigatus XC6," *Appl. Microbiol. Biotechnol*., vol. 74, no. 1, pp. 239–243, 2007.

[3] H. M. Pinheiro, E. Touraud, O. Thomas, "Aromatic Amines from Azo Dye Reduction: Status Review with Emphasis on Direct UV Spectrophotometric Detection in Ttextile Industry Wastewaters," *Dyes and Pigments*, vol. 61. no. 2, pp. 121-139, 2004.

[4] H. Moawad, W. M. Abdel-Rahim, M. Khalafallah, "Evaluation of Bio-Toxicity of Textile Dyes Using Two Bioassays," *J. Basic Microbiol*, vol. 43, pp. 218-229, 2003.

[5] J. Griffiths, "Developments in the Light Adsorption Properties of Dyes–Color and Photochemical Reactions. Development In the Chemistry and Technology of Organic Dyes, Society of Chemistry Industry, Oxford, pp. 1-30, 1984.

[6] M. Jonstrup, N. Kumar, M. Murto, B. Mattiasson, "Sequential Anaerobic–aerobic Treatment of Azo Dyes: Decolourisation and Amine Degradability," *Desalination*, vol. 280, pp. 339–346, 2011.

[7] M. A. Hossain, Z. Hasan, T. S. A. Islam, "Kinetic Evaluation on the Adsorption of Reactive Black 5 on Used Black Tea Leaves, *Dhaka Univ. J. Sci.*, vol. 59, no. 2, pp. 193–197, 2011.

[8] M. A. Hossain, M. T. Hassan, "Kinetic and Thermodynamic Study of the Adsorption of Crystal Violet on Used Black Tea Leaves," *Orbital Elec. J. Chem.*, vol. 5, no. 3, pp. 148-156, 2013.

[9] M. F. Abid, M. A. Zablouk, A. M. A. Alameer, "Experimental Study of Dye Removal from Industrial Wastewater by Membrane Technologies of Reverse Osmosis and Nanofiltration," *Iranian J Environ Health Sci Eng*., vol. 9, no. 1, pp. 17-27, 2012.

[10] R. Jain, M. Mathur, S. Sikarwar, A. Mittal, "Removal of the Hazardous Dye Rhodamine B through Photocatalytic and Adsorptive Treatments," *J. of Env. Management*, vol. 85, no. 4, pp. 956-964, 2007.

[11] A. Al-Kedasi, A. Idris, K. Saed, C. T. Guan, "Treatment of Textile Wastewater by Advanced Oxidation Process – a Review," *Global Nest: The Int. J.*, vol. 6, no. 3, 2004.

[12] M. Malakootian, A. Fatehizadeh, "Color Removal from Water by Coagulation/caustic Soda and Lime," *Iran. J. Environ. Health. Sci. Eng.*, vol. 7, no. 3, pp. 267-272, 2010.

[13] D. Kavitha, C. Namasivayam, "Recyclying Coir Pith, an Agricultural Solid Waste, for the Removal of Procion Orange from Waste Water." *Dyes Pigments*., vol. 74, pp. 237-248, 2007.

[14] F. Gholami-Borujeni, A. H. Mahvi, S. Nasseri, M. A. Faramarzi, R. Nabizadeh, M. Alimohammadi, "Application of Immobilized Horseradish Peroxidase for Removal and Detoxification of Azo Dye from Aqueous Solution," *Res. J. Chem. and Environ*., vol. 15, no. 2, pp. 217-222, 2011.

[15] Z. R. Holan, B. Volesky, "Biosorption of Lead and Nickel by Biomass of Marine Algae," *Biotechnology and Bioengineering*, vol. 43, no. 11, pp. 1001–1009, 1994.

[16] J. Allen, S. B. Koumanova, "Decolourization of Water/wastewater using Adsorption (Review)," *J. Univ. Chem. Technol. Metal*., vol. 40, pp. 175-192, 2005.

[17] Rafatullah, M., Sulaiman, O., Hashim, R., Ahmad, A.; Adsorption of methylene blue on low-cost adsorbents: a review. J. Hazard. Mater., 177, 70-80, 2010

[18] M. N. Rashed, "Adsorption Technique for the Removal of Organic Pollutants from Water and Wastewater. Organic Pollutants- Monitoring, Risk and Treatment, Chap. 7.

[19] AO (Food and Agricultural organization of the United Nations); World Tea Production and Trade: Current and Future Development, 2015.

[20] A. O. Ifelebuegu, J. E. Ukpebor, C. C. Obidiegwu, B. C. Kwofi, "Comparative Potential of Black Tea Leaves Waste to Granular Activated Carbon in Adsorption of Endocrine Disrupting Compounds from Aqueous Solution," *Global J. Environ. Sci. Manage*., vol. 1, no. 3, pp. 205-214, 2015.

[21] M. A. Hossain, M. Kumita, Y. Michigami, S. Mori, "Kinetics of Cr (VI) Adsorption on Used Black Tea Leaves," *J. Chem. Eng. Jpn*., vol. 38, pp. 402-406, 2005.

[22] A. Malakahmad, S. Tan, S. Yavari, "Valorizaion of Wasted Black Tea as a Low-Cost Adsorbant for Nickel and Zinc Removal from Aqueous Solution," J. of Chem., pp.1-8, 2016.

[23] M. Z. Mehrizi, A. Badiei, "Highly Efficient Removal of Basic Blue 41 with Nanoporous Silica," *Water Resource and Industry*, vol. 5, pp. 49-57, 2014.

[24] M. A. Hossain, M. S. Alam, "Kinetic Adsorption of Rhodamine B on Used Black Tea Leaves," *Iran. J. Environ. Health. Sci. Eng*., vl. 9, pp. 2-15, 2012.

[25] M. A. Hossain, Z. Hasan, T. S. A. Islam, "Kinetic Evaluation on the Adsorption of Reactive Black 5 on Used Black Tea Leaves," *Dhaka Univ. J. Sci.,* vol. 59, no. 2, pp. 193–197, 2011.

[26] S. Lagregren, "About the Theory of so-called Adsorption of Soluble Substances," *K. Sven. Vetenskapsakad. Handl*, vol. 24, no. 4, pp. 1–39, 1898.

[27] Y. S. Ho, G. McKay, "The Kinetics of Sorption of Divalent Metal Ions onto Sphagnum Moss Peat," *Water Res.*, vol. 34, pp. 735–742, 2000.

[28] W. J. Weber, J. C. Morris, "Kinetics of Adsorption Carbon from Solutions," *Journal Sanitary Engeering Division Proceedings, American Society of Civil Engineers*, vol. 89, pp. 31-60, 1963.

[29] M. A. Hossain, M. L. Hossain, "Dyanamic Modeling of the Transport Mechanism of Malachite Green to Adsorb on Used Black Tea Leaves," *Int. J. Recent Scientific Res.,* vol. 4, no. 10, pp. 1575-1579, 2013.

[30] P. S. Kumar, R. Gayathri, "Adsorption of Pb^{2+} Ions from Aqueous Solutions onto Bael Tree Leaf Powder," *J. of Eng. Sci. and Tech*., vol. 4, no. 4, 381-399, 2009.