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Adsorption-Desorption, Thermogravimetric Analysis and Antimicrobial Activity of Purple *Brassica oleracea* var. capitata *F. rubra* (Purple Cabbage) as Sustainable Natural Wool Dye

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This study focussed on the dyeing wool fabrics with colourant purple *Brassica oleracea* var. capitata *F. rubra*. The process of dyeing was taken place at 60 ºC and pH 5. Adsorption and desorption isotherms experiments investigated by Langmuir and Frendulich models, Langmuir was fitted with the experimental data dyeing adsorption than Freundlich model. The adsorption kinetic of dyeing experiments was studied by using pseudo-first and pseudo-second models and pseudo-second model was more suitable for wool dyeing experiment. The thermal stability of the dye (purple *Brassica oleracea* var. capitata *F. rubra*) was studied using thermogravimetric and differential thermal (TGA/DTG) analysis. The antimicrobial activity of purple cabbage methanolic extract recorded of high exhibited *Staphylococus aureus* (17-18 mm), weak against fungi (11-12 mm), moderate against *Bacillus subtilis* (14-15 mm) and no results against *Escherichia coli*. The results obtained from aqueous and petroleum were negative.

Keywords: Purple cabbage, Antimicrobial activity, Adsorption, Dyeing.

INTRODUCTION

Dyeing textile by using sustainable resources and eco-friendly became necessary in our life. Extract colours from sustainable natural sources like red, pink flowers vegetables and fruits are rich source to dye wool, because of pigmentation process is the most important step in the production of textiles [1]. The environment polluted by chemicals used in industries for dye purposes, are also known by high toxicity and remove out the effluent during the dyeing process to the environment [2]. The textile industry consumes large quantities of water in dying processes in many steps; when fixing dyes on the textile and finishing steps, so that causes the stream of waste waters containing artificial hazardous colours for human and aquatic health which are produced from the textile industry, in a large amounts [3]. Thus, the treatment of toxic effluents and removal of the colours from wastewater is considered as a big environmental and economic problem [4]. Toxic wastewater affect negatively on ground water because we consume water. In addition to that there are many issues faces human health during the contact of dyes with skin and eye to cause pain and irritation with redness [5,6].

Globally, the interest for using natural dyes extracted from the coloured plants for process of dye was taken place in industries and started to limit the use of artificial and chemical pigments since years [7]. Natural dyes are a sustainable eco-friendly, renewable, cheap and available they are also known as low or non-toxicity, easily biodegradability, such pigments are appropriate environmentally. For mankind use, it doesn't have allergic effect for skin, which means skin friendly and may also provide a benefit to health for whom wearing clothes dyed by natural pigments leading a satisfaction for consumer [8]. Natural dyes for the reasons of their originality as well as the low-cost natural resources of usage is also consi-dered to be easily treatable [9,10].

Flavoinds are natural the source of pigments [11] and purple cabbage is one of the sustainable natural dye source.

The red or purple cabbage (*Brassica oleracea* L. var. capitata *F. rubra*), is known as a plant to some regions of Europe known Mediterranean regions, in these days cabbage is planting every where in the world and utilized for nutrition an important vegetable [12]. Purple cabbage is known is a favourable origin of anthocyanin for wool dyeing, pigments can be used in a very broad pH range and characterized by a high stability to heat and light in comparing to anthocyanins extracted from other natural sources [13]. Red cabbage is of the another natural pigments that supply red colour, to natural textiles such as wool [14]. Physico-chemical interaction which is occurring during the adsorption and diffusion of the dying solution onto fiber of the textiles for example wool taking place to give best fixation of dyes [15]. In this article, purple of *Brassica oleracea* var. capitata *F. rubra* (purple cabbage) vegetable were used for its possibility to dye and textile colouration. The present studies also investigate the biological effect of anthocyanins and thermograms analysis for the thermodynamic properties of the natural dye.

EXPERIMENTAL

The reagents used were for pH adjustment hydrochloric acid was used. Potassium alum ($Al_2K_2(SO_4)_4$) was used as mordant solutions of 5 g/L, NaCl used were of analytical grade corresponding to a purity of 99%.

Thermal analysis which were taken place by Shimadzu thermogravimetry TGA-50H instruments for the TGA and DTG analysis at a rate of heating of 10 ºC/min in nitrogen at shaking bath mosphere (10 mL/min). Spectrophotometer (GENESYS 10S UV-VIS spectrophotometer), microbalance (with 0.1 mg readability) was used for weighing samples. For shaking and adsorption experiments, a magnetic hotplate stirrer and mechanically shaker of type Spin Magnetic instrument model: MS7-H550-Pro were used (± 1 rpm speed resolution, control accuracy of work plate ± 1 ºC, stoppered pyrex glass flask used for adsorption experiments. filter paper which is type Whatman of (0.45 µm pore size) was used for filtration dye.

Adsorbate: The purple cabbage was the source of natural dye, it was collected from the domestic market in Saudi Arabia. It was washed by distilled water, cut to a very small pieces, then dryness of the sample was by air-dried for few days, the dried sample was milled with high speed blender to obtain fine powder, then passed through a mesh sieve size of 1-2 mm. The pure powder was kept in test tube until used. Purple *Brassica oleracea* var. capitata *F. rubra* (purple cabbage was abbreviated as PBO).

General procedure

Preparation of adsorbent wool scouring : Wool fabric washed with 1 g/L of sodium carbonate solution and 2 g/L of detergent solution characterized by non-ionic nature at 35 ºC for 60 min then washed wool scoured with distilled water and then with deionized water, after that the material was dried at room temperature.

Mordanting: The scoured materials were mordanted by using an aqueous solution of the mordent as a liquor with ratio 1:100 at 80 ºC for 35 min, then they were washed, squeezed and dried at room temperature.

Extraction of anthocyanin from purple cabbage for dyeing and the testing of antimicrobial activity: Dry and pre-aqueous extract of PBO purple cabbage (100 g) was soaked in 1000 mL of 80% methanol. The same sample size was also treated using 1000 mL petroleum ether for 24 h at room temperature. The extract was filtered, using filter paper No. 4, after which the solvent was evaporated under reduced pressure using rotary evaporator apparatus.

Process of dyeing: The first step for wool dyeing was the pH adjustment of solution by using HCl, then wool samples were dyed for 1 h with a liquor ratio of 1:100 samples. After dyeing, the samples were washed to remove the excess dye, which was unfixed during the dyeing process, using 1 g/mL non-ionic detergent at 35 ºC for 5 min, rinsed in cold water and dried at room temperature. Determination of the adsorption by the amount of dye adsorbed per gram of fabric (q) (mg/g fabric) was determined through a mass balance and adsorption kinetics of dyeing for wool samples were done by immersing wool with dye by a liquor ratio 1:100. The wool fabrics were removed from dyeing equipment at different dyeing times.

Adsorption and desorption process: The adsorbed quantity of dye (g) of wool (q) (mg/g wool) was determined through a mass balance, means determined by the difference in weight of wool before and after adsorption. The amount of adsorption at equilibrium, q_e (mg/g), was calculated by using the following equation:

$$q_e = \frac{(C_o - C_e)}{W} \times V \quad (1)$$

where (dye-wool) concentrations expressed by C_o and C_e (mg/L) are the dye initially and equilibrium, respectively; V is the volume of solution (L), W is the weight of dry adsorbent (g).

The adsorption isotherm models for dye process used were Langmuir adsorption isotherm [16-18]. Freundlich model was the another adsorption model used in this study, Freundlich model fit with a surface contains different sites of different energies that are not equally available, so the type of adsorption in this case is a heterogeneous adsorption, which predicts the formation of multilayers [19]. If the Langmuir and Freundlich models do not give reasonable data of results for the experiment, then Langmuir-Freundlich model can used to explain both Langmuir type as the Freundlich adsorptive behaviour, which better fits in heterogeneous surface of adsorbent as in case of wool [20], it is expressed as follows:

$$q_e = \frac{q_m (K_{LF} C_e)^n}{1 + q_m (K_{LF} C_e)^n} \quad (2)$$

where C_e is the dye concentration at equilibrium (mg/L), q_e is the amount of dye adsorbed at equilibrium (mg/g), q_m is the maximum quantity adsorbed in the monolayer (mg/g), n is a constant of heterogeneity and K_{LF} is the Langmuir-Freundlich adsorption constant (L/mg) [21].

Desorption isotherms: Desorption was carried out by using 50 mL of 0.01 M of NaCl added to 50 mL of sample with 50 g fully loaded wool fabric containing initial concen-

tration of 0.06 M of dye. In order to study the effect, sodium electrolyte solution was prepared and added to the suspension and stirred until equilibrium with molecular ratio of salt and dye attained. In separate glass, 5 mL suspension sample was shaken for 1 h, the supernatant of 5 mL from mixture was taken from the batch at predetermined time intervals and centrifuged for 20 min at 2500 rpm. Desorbed dye concentrations were determinated from their absorbance characteristics in the UV-visible range. The efficiency of desorption is known as dye desorbed amount per gram of adsorbent at equilibrium, hence the quantity of desorbed dye was calculated by the following equation:

$$Q_i = \frac{(C_o - C_t)V}{m} \quad (3)$$

where Q_i is the quantity of dye in mg per gram of adsorbent, C_o and C_t are respectively the initial concentrations and time t of the dye (mg/L), V: volume of solution (L), m: mass of adsorbent used (g). The percentage of dye removal was calculated from the relationship:

$$\text{Dye removal (\%)} = \frac{(C_o - C_t)}{C_o} \quad (4)$$

C_o and C_t are respectively the initial concentrations and at the time t of the dye (mg/L).

Kinetic studies: The kinetic batch experimental data were collected and interpreted by using two different kinetic models the pseudo-first order model and the pseudo-second order kinetic model.

Pseudo-first-order: Kinetic model the rate constant of adsorption was determined from the pseudo-first-order equation given by Langergren & Svenska [22] as:

$$\ln (q_e - q_t) = \ln q_e - k_1 t \quad (5)$$

where q_e and q_t are the amounts of PBO adsorbed (mg/g) at equilibrium and at time t (min), respectively and k_1 is the rate constant adsorption (min^{-1}).

Pseudo-second-order: This kinetic model is used to describe the chemisorption, which contains the valence forces by the sharing electrons or exchange between the dye and the fabric as covalent forces [23]. The pseudo-second-order equation [22] based on equilibrium adsorption is expressed as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_2 e} + \frac{1}{q_e} \cdot t \quad (6)$$

Antibacterial and fungi activities: The Petri dish agar diffusion method [24] was adopted to evaluate the antibacterial and antifungi activity. A 2 mL of stock suspension 10^8–10^9 CFU/mL bacteria and fungi mixed with 200 mL of molten sterile nutrient agar, respectively. Maintained at 45 ºC, 20 mL aliquots of the incubated nutrient agar were distributed into sterile Petri-dishes. The agar was left to set and in each of these plates (10 mm in diameter) were cut using a sterile cork borer (No. 4) and agar discs were removed. Alternate cups were filled with 0.1 mL samples of each of the extracts using automatic microliter pipette and allowed to diffuse at room temperature for 2 h. The plates were then incubated in the upright position at 37 ºC for 8 h. Two replicates were carried out for each extract against each of the test organisms. Simultaneously positive controls involving the addition of methanol as instead of the extracts were carried out separately. After incubation the diameters of the resultant growth inhibition zones were measured, averaged and the mean values were tabulated.

Preparation of bacterial suspensions: Aliquots (1 mL) of a 24 h broth culture of *Bacillus subtilis* (NCTC 8236), *Staphylococcus aureus* (ATCC 25923), *Eschericha coli* (ATCC 25923) and *Pseudomonus aeruginosa* (ATCC 9763) was aseptically distributed onto nutrient agar slope and incubated at 37 ºC for 24 h. The bacterial growth was harvested and washed off with sterile normal saline and finally suspended in 100 mL of normal saline to produce a suspension containing about (10^8-10^9) colony forming units per mL. The suspension was stored in the refrigerator at 4 ºC until used.

Statistical analysis: A mean value of data was analyzed by the one-way ANOVA. The efficacies were obtained by calculating the differences between the edema size in the treated and the control and the values were transformed into percentage using mean index.

RESULTS AND DISCUSSION

Effect of dye concentration on the adsorption of dye on wool fiber: Table-1 shows the amount of adsorbed dye was high on the surface of the wool fabrics, since the structure of wool contains a high amount of functional groups [25]. In addition, it was found that the capacity of the adsorption depends on the concentration, therefore at the concentration of dye 140 mg/mL, a high amount of dye adsorption on wool fabrics was observed. The result is attributed to an increase in the initial dye concentration, which increase the driving forces of the concentration gradient [18]. The molecules of the wool contain amino acid in acidic pH, in which proteins were formed. Free amino and carboxyl groups, which are, formed from proteins gives an amphoteric behaviour for wool [26]. During the dyeing of wool, a hydrogen bond occurs between the dyestuff and the amino groups of the wool [18,25]. Moreover, the amino groups (-NH_2) of protein present in the wool fiber are protonated, which help to attract anionic dyestuffs [27]. During the dyeing processes, it was observed that all samples of wool showed intense colouration ranging from faint to red colour when the concentration increasing from 60 to 140 mg/L at 60 ºC. The higher intense red colour were shown for two samples of initial concentrations (140 mg/mL) and (120 mg/mL) and faint red colour for the rest three samples with concentrations (60 mg/mL), (80 mg/L) and (100 mg/mL). The variable colours of dye/g onto wool are shown in Table-1.

Adsorption isotherms: Fig. 1 shows the equilibrium adsorption isotherm of dye (PBO) onto the surface of wool fabric. At the first stage, a short plateau was observed in the isotherms, in this stage the dye have similar affinity for all dye molecules on to the initial active unsaturated sites of the wool, indicating the multilayer chemisorption process was formed, due to the strong electrostatic forces of the multilayer after the monolayer formation involving in the wool fabrics. The second plateau arise due to the formation of second monolayer as a new surface

TABLE-1
VARIABLE COLOURS FOR THE ADSORPTION OF DYE ONTO WOOL FABRIC IN DYEING PROCESS WITH EXTRACT OF (PBO) PURPLE CABBAGE

Concentration (mg/mL)	q (mg/g wool)	Intensity of colour
60	3.15	Faint red
80	3.866	Faint red
100	5.07	Medium red
120	5.37	Red
140	6.07	Red

in which chemisorption can be formed. As a result, the second plateau also indicates for the saturation state of a new surface. The slope of the adsorption isotherms known as type S2 [28]. The lower results obtained for desorption indicated the efficiency properties of this adsorbents in the process of adsorption.

Langmuir-Freundlich model: Langmuir-Freundlich model adsorption and desorption isotherms are shown in Figs. 2a-b and 3a-b for both Langmuir and Freundlich models. The q_{max} values are close to the experimental value q_{max}, the n values denotes the heterogeneity, which is higher than one, indicating the formation of a homogeneous monolayer then followed by multilayer formation and thus confirmed the strong adsorption of PBO on wool fiber, that means the predominant of chemical adsorption in the dyeing process. The correlation coefficients R^2 of Langmuir-Freundlich isotherms values are shown in Table-2, indicates a monolayer adsorption. The equation of

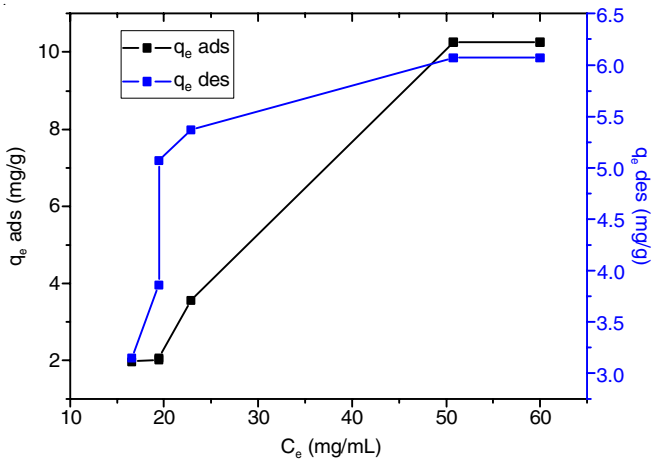

Fig. 1. Non-linear adsorption/desorption isotherms of Langmuir-Freundlich model of (PBO) dye onto wool fabrics

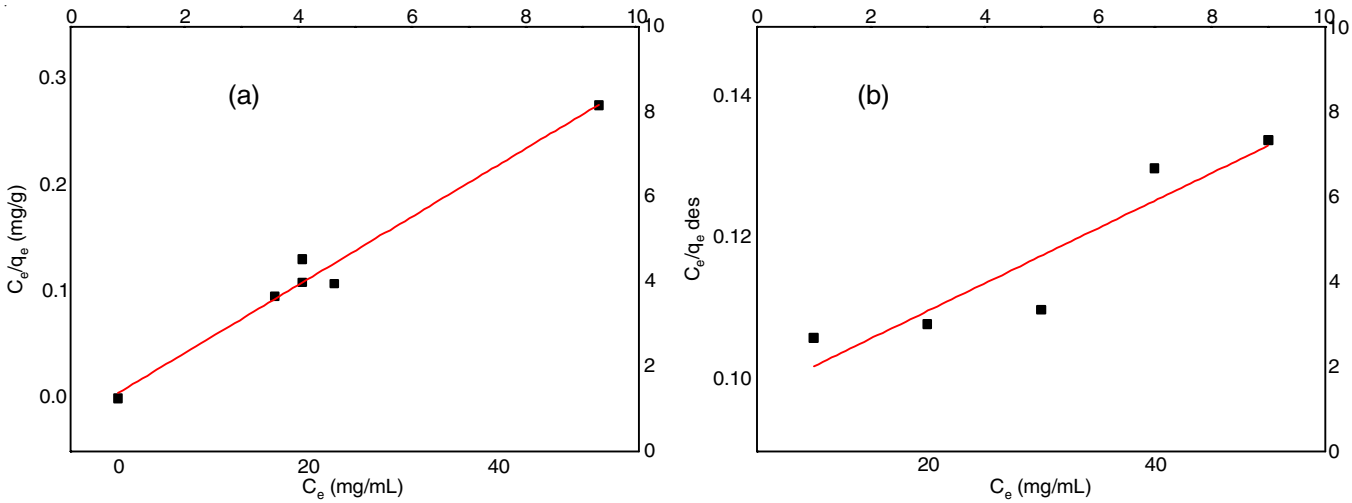

Fig. 2. Langmuir linear isotherm for (a) adsorption purple cabbage onto wool fabrics (b) desorption of purple cabbage onto wool fabrics

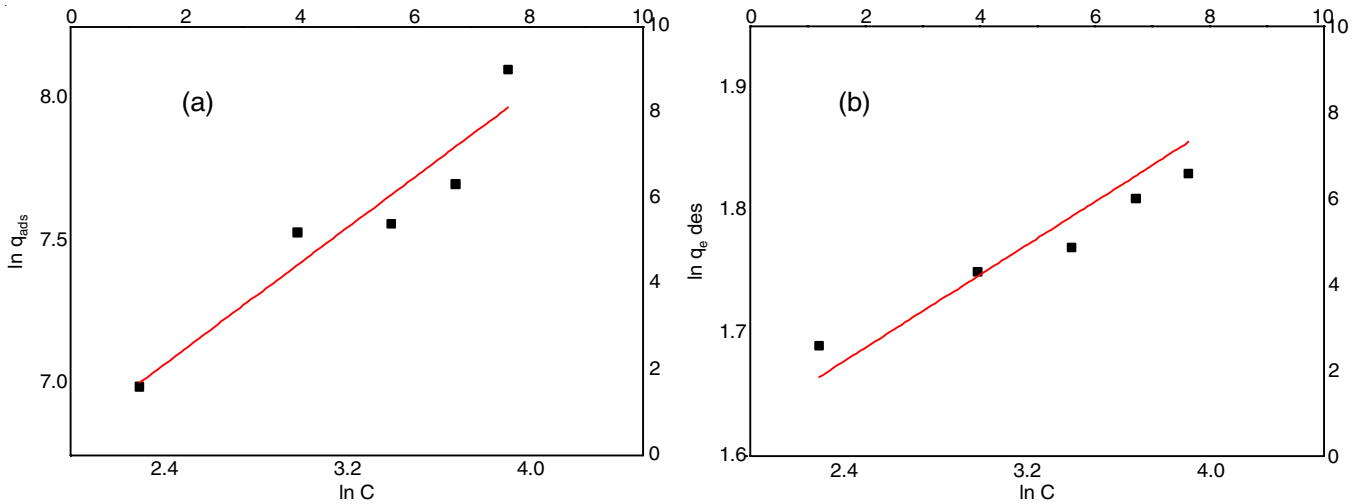

Fig. 3. Freundlich linear isotherm for (a) adsorption purple cabbage onto wool fabrics (b) desorption of purple cabbage onto wool fabrics

TABLE-2
ADSORPTION/DESORPTION ISOTHERMS MODEL PARAMETERS OF LANGMUIR AND FREUNDLICH ISOTHERMS FOR PURPLE CABBAGE (PBO) ONTO WOOL FABRIC

Isotherms	R^2	q_{max}(mg/g)	K	b (L/mg)	1/n	n
Langmuir adsorption	0.97	188.67	–	0.0053	–	–
Langmuir desorption	0.81	10.65	–	–	–	–
Frendulich adsorption	0.87	–	0.74	–	0.59	1.69
Frendulich desorption	0.92	–	0.14	–	0.11	9.09

dimensionless constant called or separation factor (R_L) also called equilibrium parameter is used to identify of adsorption process (eqn. 7):

$$R_L = \frac{1}{1 + bC_i} \quad (7)$$

where C_i is the initial concentration of PBO. If the value of R_L lies between 0 and 1, the adsorption process is favourable; and if R_L is greater than 1, then the adsorption process is unfavourable [17].

The R_L results at different initial concentrations (C_i) are shown in Fig. 4, where the value of R_L in the range 0–1 confirms the adsorption process is favourable. R_L values at higher initial C_i concentration are more favourable. The favourable behaviour was explained due to relation to the irreversibility of the system, which depends on the qualitative estimation of C_i (PBO dye) interactions.

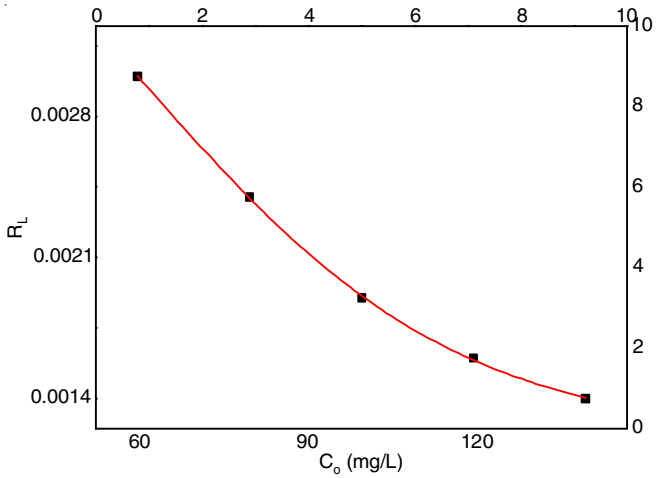

Fig. 4. Separation factor *versus* initial purple cabbage dye

Kinetic of adsorption: Pseudo-first order constants were calculated, as rate constant (k_1) correlation coefficient R_2 and q were found from the slope and intercept (Table-3), which resulted by plotting ln ($q_e - q_t$) *versus* t (Fig. 5). The observation of the adsorption was fast with contact time at early steps and gradually decreased with time until it became constant. The same behaviour was observed in the work of Chairat *et al.* [18].

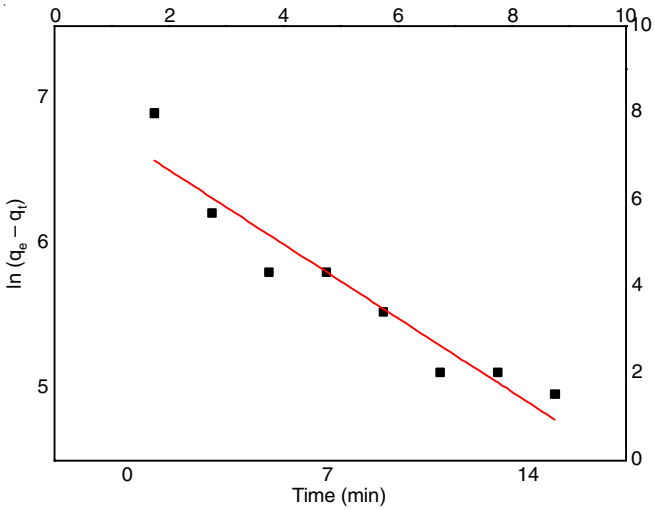

Fig. 5. Pseudo-first-order kinetics for adsorption of purple cabbage onto wool fabric at 35 ºC

The results indicate that the pseudo second-order model is more suitable than the pseudo-first order because it describes well the adsorption of purple cabbage dye on wool (Fig. 6), as a linear fit of pseudo-second-order model. On the other hand, the results of pseudo-first order model is small in the correlation factor R^2 and also not close to the value of 1.

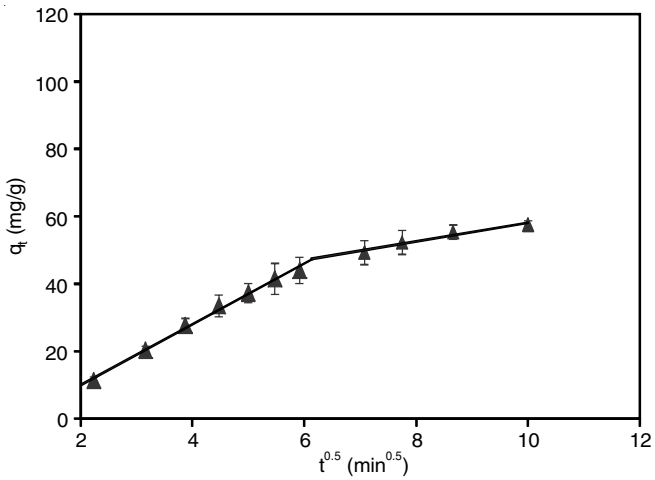

Fig. 6. Pseudo-second-order kinetics for adsorption of purple cabbage onto wool fabric at 35 ºC

TABLE-3
PSEUDO-FIRST-ORDER AND PSEUDO-SECOND-ORDER KINETIC MODEL PARAMETERS FOR DIFFERENT INITIAL CONCENTRATIONS OF PURPLE CABBAGE (PBO) ONTO WOOL FABRIC

Pseudo-first-order kinetic models			Pseudo-second-order kinetic model		
q_e (mg/g)	k_1 (min^{-1})	R^2	q_e (mg/g)	k_2 (g wool mg dye^{-1} min^{-1})	R^2
6.889	0.127	0.89	162	1.32×10^{-5}	0.995

Thermal analyses: TG/DTG was carried out to evaluate the thermal stability of anthocyanins extracted from the purple cabbage (PBO) in nitrogen atmosphere are shown in Fig. 7. The curves show the thermal decomposition of PBO and the results are summarized in Table-4. TG/DTG shows the loss in weight of about 10% up to 210 ºC. This could be due to the evolution of the solvent molecules from the PBO [28]. Between 210 ºC and 320 ºC, dye lost 42.7% of its original weight, above 450 and 650 ºC, a gradual loss of 42% of its original weight was observed. This could be attributed to the decomposition and volatilization of the low molecular weight fractions [29]. From DTA curves, it is observed that PBO thermally degraded over the temperature range of 150-996.92 ºC. The weight loss in the temperature range 150-350 ºC shows the loss of water of hydration also a narrow broad peak of endothermic decomposition followed by a sharp exothermic reaction from 150-650 ºC was observed [30].

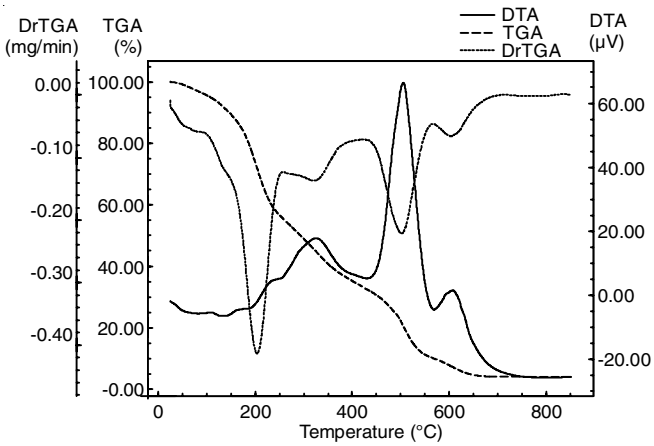

Fig. 7. Thermal gravemetric analysis (TGA) and differential of thermal analysis (DTA) data of PBO purple cabbage

TABLE-4
THERMOANALYTICAL RESULTS (TG) AND DIFFERENTIAL OF THERMAL ANALYSIS (DTA) DATA OF (PBO) PURPLE CABBAGE

Anthocyanins from cabbage	Δm (%)	Ti (°C)	T_f (°C)	T_{max}
Stage 1	4	70	200	75
Stage 2	15	210	320	510
Stage 3	50	450	650	610
Differential thermal analysis (DTA) data of (PBO) purple cabbage				
DTA peaks (T_m, °C)			Nature	
650			Exo	
499			Exo	
350			Exo	
150			Endo	

Antimicrobial activity of methanolic, petroleum ether and aqueous extract of purple cabbages: The antimicrobial activities of aqueous, methanolic and petroleum ether extracts of purple cabbage (PBO) were also evaluated. The results data recorded in Table-5 against the standard organism show that methanolic extract weak against fungi (11-12 mm) but exhibited high activity against *Staphylococus aureus* (17-18 mm) and moderate against *Bacillus subtilis* (14-15 mm). The results obtained from aqueous and petroleum extracts were negative.

Conclusion

This study confirmed that the purple cabbage methanolic extract recorded of high exhibited *Staphylococus aureus* (17-18 mm), weak against fungi (11-12 mm), moderate against *Bacillus subtilis* (14-15 mm) and no results against *Escherichia coli*. The results obtained from aqueous and petroleum extracts were negative.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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TABLE-5
ANTIMICROBIAL ACTIVITY METHANOLIC, PETROLEUM ETHER AND AQUEOUS OF PURPLE CABBAGE

Solvent	Standard organism used MDIZ (mm)			
	Bacteria			Fungi
	Bacillus subtilis	*Staphylococus aureus*	*Escherichia coli*	*Candida albicans*
Methanol (70%)	15	18	–	–
Petroleum ether (3%)	–	–	–	–
Aqueous	–	–	–	–

MDIZ: Mean diameter of growth inhibition zone in (mm). Average of 2 replicates; MIZD (mm): > 18 mm: Sensitive (14-18) mm: Intermediate, < 14 mm: Resistant; (-) No inhibition zone.

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