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Research Article

Effect of Ambient Conditions on Kinetic Behavior in Color Removal with Modified Adsorbent

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Abstract

In this study, the adsorption isotherms for removal of Maxilon Golden Yellow GL EC 400% (MGY) dye, which is widely used in industrial facilities from aqueous solutions by using modified clay were investigated. Clay (K_0), obtained from the Avanos District of Nevşehir Province, was used as the adsorbent material, modified with ultrasonic sound waves (K_1). According to the results obtained, as the contact time increased in the adsorption experiments performed with both clays, the adsorption efficiency also increased. When analyses were made according to the Langmuir and Freundlich Isotherms, it was determined that the adsorption experiments were more suitable for the Langmuir Isotherm. It has been determined that no external energy is required for adsorption. Highest color removal for modified clay (K_1); Obtained at pH 4 (99.01%) in 300 minutes and 25 ^oC. It has been determined that clay, which is abundant in nature, can be used as an alternative important method in the removal of MGY dye with this new modification method.

Keywords
Ultrasonic sound
Adsorpsiyon
Dye
Modified clay

Modifiye Adsorbent ile Renk Gideriminde Ortam Koşullarının Kinetik Davranışa Etkisi

Özet

Bu çalışmada; endüstriyel tesislerde yaygın olarak kullanılan Maxillon Golden Yellow GL EC 400% (MGY) boyasının, sulu çözeltiden modifiyeli kil ile gideriminde adsorpsiyon izotermleri incelenmiştir. Adsorplayıcı madde olarak Nevşehir İli'nin Avanos İlçesi'nden temin edilen doğal kil (K_0), ultrasonik ses dalgaları ile modifiye edilerek (K_1) kullanılmıştır. Elde edilen sonuçlara göre, her iki kil ile gerçekleştirilen adsorpsiyon deneylerinde temas süresi arttıkça adsorpsiyon verimliliği de artmıştır. Langmuir ve Freundlich İzotermlerine göre analizler yapıldığında ise adsorpsiyon deneylerinin Langmuir İzotermine daha uygun olduğu belirlenmiştir. Adsorpsiyon için dışarıdan enerjiye ihtiyaç duyulmadığı tespit edilmiştir. Modifiye kil (K_1) için en yüksek renk giderimi; pH 4'de (%99.01) 300 dakika ve 25 ^oC'de elde edilmiştir. Doğada bol miktarda bulunan kilin bu yeni modifikasyon metodu ile MGY boyasının gideriminde alternatif önemli bir metot olarak kullanılabilirliği belirlenmiştir.

Anahtar Kelimeler
Ultrasonik ses
Adsorpsiyon
Boya
Modifiye kil

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1.INTRODUCTION

54% of the existing dye waste seen worldwide originates from textile industries, and 46% from dyeing industries, paper industries, tanneries, and dye production industries [1]. Dyed wastewater resulting from these sectors carries toxic risks for human and environmental health as it has mutagenic and carcinogenic properties. Discharging this dyed wastewater into the environment causes the death of aquatic species such as fish, plants, and mammals, eutrophication, odor, and aesthetic problems caused by color change. In addition, such colored wastewater prevents photosynthetic activity by reducing the penetration of sunlight into benthic organisms in the aquatic areas where it is discharged, as well as causing visual pollution [2-5].

There are many studies on the treatment of dyed wastewater, the main ones being adsorption [6], biosorption [7], membrane filtration [8], ozonation [9], biological treatment [10], electrocoagulation [11], Fenton modifications (foto, sono etc.) [12-16] and electrooxidation [17]. Among existing technologies, the most suitable method for the removal of dyestuffs from wastewater is adsorption [18], since other techniques used have some disadvantages due to application cost, removal efficiency, and toxic sludges produced [19, 20].

Ultrasound is a rapidly growing field in purification engineering and is an effective tool for breaking down different organic pollutants, including dyes. The effectiveness of ultrasound is due to the radical production that occurs with the cavitation bubble formed in water and the particle surface is more activated by the effect of mechanical force, thus obtaining a larger surface area. Diffusion of adsorbed molecules through the pore is achieved faster due to the mechanical force effect caused by ultrasound. These forces increase and enlarge the pores on the surface of the adsorbent material, thus extending the surface for adsorption. Thus, they increase the adsorption capacity of the material [20, 21, 22].

In this study, environmental conditions were examined in the removal of MGY dye found in industrial wastewater from aqueous solution by using natural clay modified with ultrasonic sound waves. The usability of modified natural clay, which is abundant in nature and does not contain toxic substances, as an alternative treatment material in the removal of MGY dye from an aqueous solution, was investigated. Studies on the change in adsorption efficiency and determination of the appropriate isotherm were carried out at different temperatures and under optimum experimental conditions. Additionally, it was tried to reveal how the ambient temperature and pH balance affect the adsorption mechanism.

2.MATERIAL AND METHOD

In the study, MGY dyestuff was used because it is a dyestuff used extensively in industrial facilities and has not been studied much in the literature. Commercially available MGY dye was used to prepare an aqueous solution by preparing a stock solution of 100 mg/L without purification. The chemical structure of MGY dye is given in Figure 1.

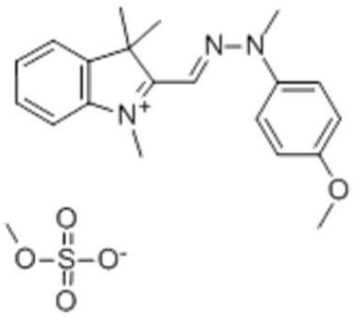

Figure 1. Chemical structure of MGY dye [23].

Natural clay used as adsorbent in the study was obtained from the stream bed of Cin stream, which is approximately 5 km away from Avanos District of Nevşehir province. After washing with 1 N H_2SO_4, natural clay (K_0) was brought to a neutral pH level by washing with distilled water (Minipure, Destup). Natural clay, whose pH was brought to neutral, was precipitated by centrifugation (NÜVE, NF 200) at 5000 rpm for 10 minutes. It was dried in an

oven (JSR, JSOF-100) for 48 hours at 105 ^{o}C. Since the particle size of the material is important, dried natural clays (K_0) were sieved in 150-200 µm sieves. Half of the natural clay (K_0) was kept in a desiccator to be used in batch adsorption experiments. The other half was used to add 50 g of clay to beakers containing 500 mL of pure water and undergo modification on an ultrasound device (Bandelin, HD 2200) at 100% power level (200Watt) for 30 minutes. After the modification process, the modified clay (K_1) was dried again under the same conditions and was kept in the desiccator until used in the batch adsorption experiments.

All MGY solutions were prepared with ultrapure water (MP Minipure Destup). As a result of examining the studies conducted by different researchers [24, 25], it was decided that the amount of adsorbent would provide maximum removal with 2.0 g. Therefore, to determine the appropriate conditions for the removal of MGY dye from clay aqueous solution, 2.0 g was weighed and added to 100 mL of MGY solution prepared at 100 mg/L. The experimental study was carried out at different pHs (2-12), different contact times (5-300 min) and different temperatures (25-45^{o}C). The pH of the solution was adjusted with 0.1 N Hydrochloric acid and 0.1 N Sodium hydroxide using a multi-parameter meter (Hach Lange, HD30d). To determine the amount of adsorbed MGY dye, solids were precipitated from the solutions by centrifugation (NÜVE, NF 200) for 5 minutes at 2400 rpm after the adsorption process was completed. After centrifugation, the sample was separated from the adsorbent using 0.45 µm membrane filters (Millipore Corp., Bedford, Mass.). Thermo Scientific Aqua Mate Plus UV-VIS model spectrophotometer was used in the measurements.

The amount of MGY adsorbed at equilibrium; q_e (mg g^{-1}) was determined by the equation given below:

$$q_e = V(C_0 - C_e)/W \quad (1)$$

In the formula, the adsorption capacity in mg/g is expressed as qe; initial and equilibrium concentrations in mg/L are expressed as C_0 *and* C_e; the volume of the dyestuff solution in mL is expressed as *V*; the mass of the adsorbent used in mg is expressed as *W* [26].

Freundlich adsorption isotherm is expressed by the following equations, which are obtained assuming that multilayer adsorption occurs on a heterogeneous adsorbent surface [27].

$$\log q_e = \log K_f + (1/n) \log C_e \quad (2)$$

In the formula, the Freundlich constant is shown as K_f (mg/g) and the Freundlich coefficient is shown as n. The parameter n describes the active sites on the adsorbent surface of natural adsorbents with low energy heterogeneity. This parameter is also an indicator of the adsorption intensity, and a value between 1 and 10 indicates a good adsorption process. If n is less than 1, it indicates that the adsorption is chemical, and if it is greater than 1, it indicates that it is physical [28].

The Langmuir isotherm is expressed by the following equation.

$$\frac{C_e}{q_e} = \frac{1}{Q_0 \, b} + \frac{1}{Q_0} C_e \quad (3)$$

Q_0 (mg g^{-1}) and b (L mg^{-1}) are Langmuir constants related to the adsorption capacity and adsorption rate, qe is the amount of MGY dye adsorbed at equilibrium (mg g^{-1}), and C_e is the liquid phase equilibrium concentration (mg L^{-1}) [29].

3.RESULT AND DISCUSSION

3.1. Adsorption isotherms for 25 ^{o}C temperature

Adsorption isotherms obtained for K_0 and K_1 clays at all initial pH values at 25 ^{o}C are given in Figure 2- Figure 7. When the adsorbed amounts of adsorbent are examined, it is seen that more adsorption is achieved with K_1 clay at all pH values at 25 ^{o}C adsorption temperature.

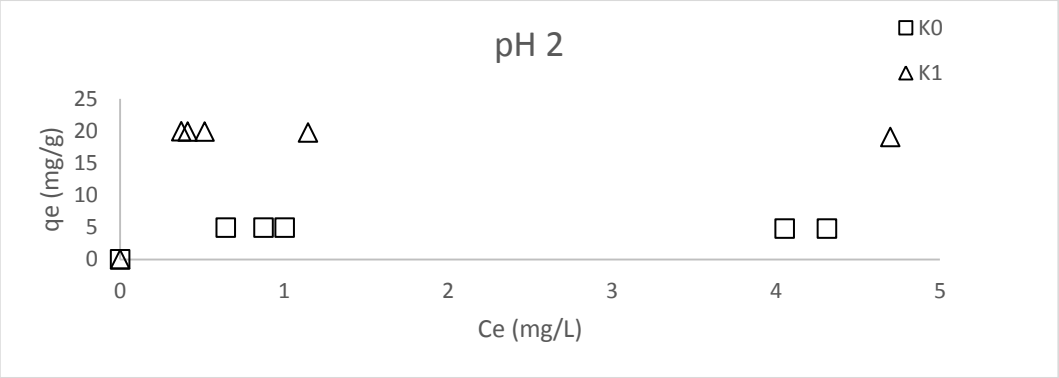

Figure 2. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 2

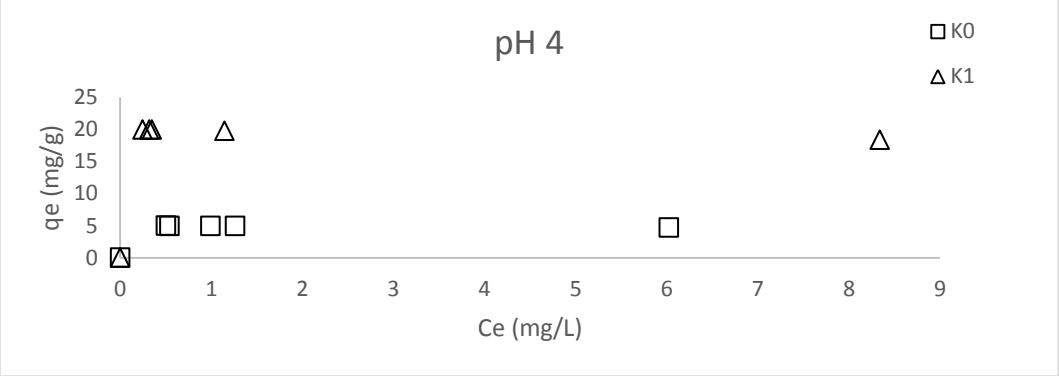

Figure 3. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 4

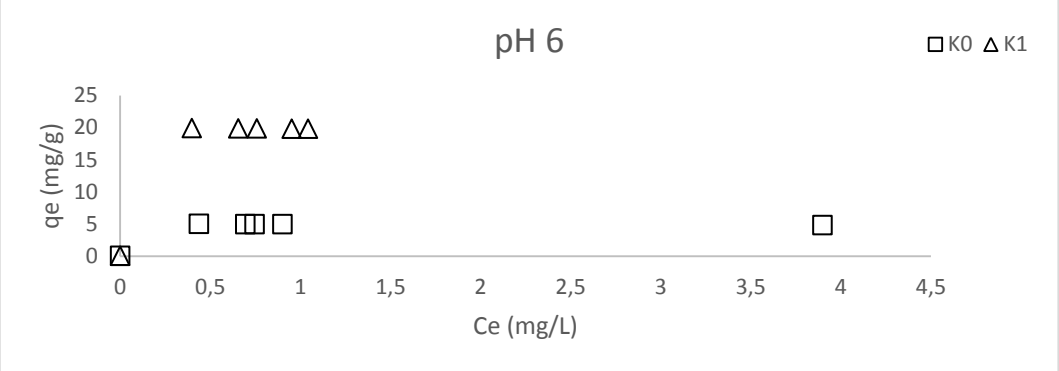

Figure 4. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 6

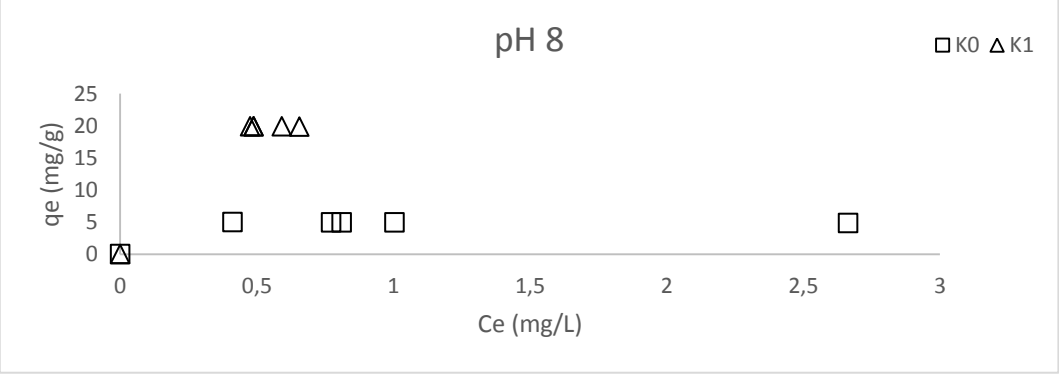

Figure 5. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 8

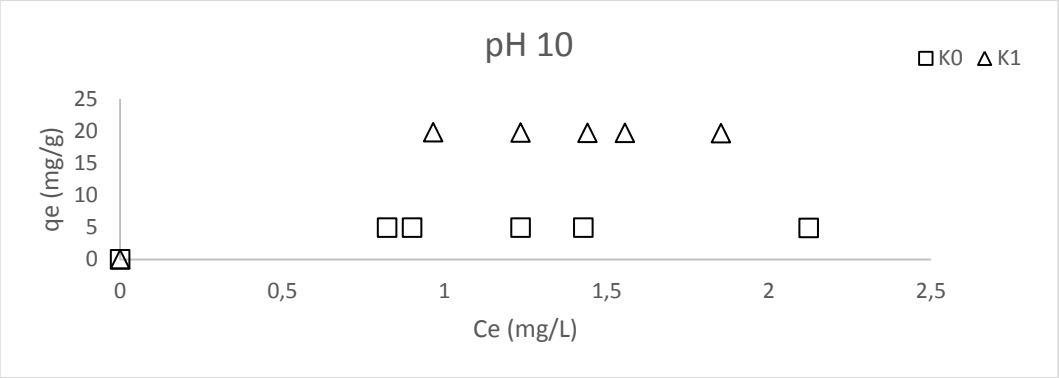

Figure 6. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 10

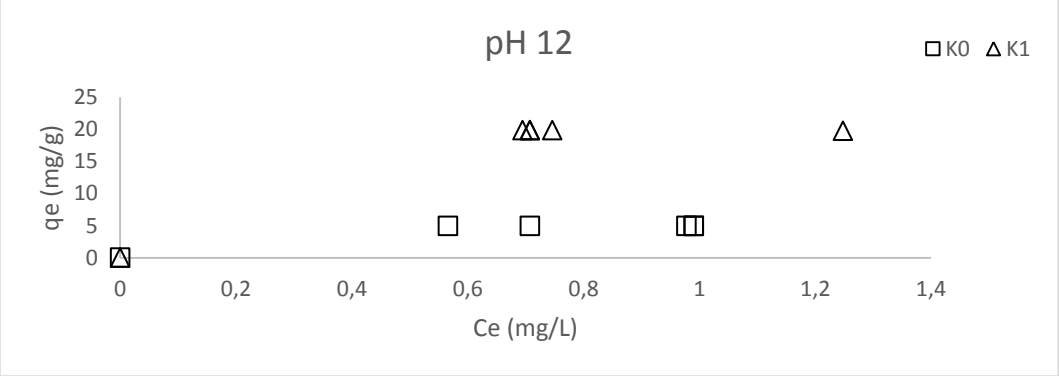

Figure 7. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 12

The Langmuir equation states that adsorption occurs on very strong homogeneous surfaces. However, this situation loses its validity because there is chemical heterogeneity in the structure of natural adsorbents.

The isotherm and constants given for K_0 clay were taken from our previous research study [30]. When the isotherms and constants given for K_0 and K_1 clays in Table 1 are examined, the highest R^2 value is at pH 12 (R^2=0.98) for K_0 clay and at pH 8 and pH 12 (R^2=0.99) for K_1 clay was obtained. Therefore, it is possible to say that the Langmuir Isotherm is more applicable for K_0 and K_1 clays under basic conditions. When the b constant, that is, the adsorption energy values between adsorbent and adsorbate, is examined, it is clear that this value is higher in the analyses performed with K_0 clay. It is seen that the b value varies between 2.43×10-4 – 13.4×10-4 L/mg, and the energy between the adsorbent and adsorbate decreases as pH increases. When Q_{max} adsorption capacities are examined, it is possible to say that the adsorption capacity of K_1 clay is up to 17 times higher than that of K_0 clay. No significant

relationship was found between pH change and adsorption capacity. R_L value is an important characteristic of Langmuir Isotherm and is used as a separation factor. The R_L value is desired to be between $0<R_L<1$ for good adsorption [31]. It is seen that the R_L values for K_0 and K_1 clay vary between 0.82-0.95, that is, between 0 and 1, under all pH conditions. Therefore, no external energy is needed for adsorption. Accordingly, it was determined that the analyses were in compliance with the Langmuir Isotherm.

Table 1. Langmuir Isotherms for 25 ^oC temperature

	K_0 Clay				**K_1 Clay**			
25 ^0C	**Langmuir Isotherms**	**Langmuir Isotherms Constants**			**Langmuir Isotherms**	**Langmuir Isotherms Constants**		
	R^2	$b\times10^{-4}$ (L/mg)	Q_{max} (mg/g)	R_L	R^2	$b\times10^{-4}$ (L/mg)	Q_{max} (mg/g)	R_L
pH 2	0.91	13.4	156	0.82	0.71	0.41	1250	0.95
pH 4	0.65	11.3	185	0.82	0.59	0.47	1111	0.94
pH 6	0.72	7.25	285	0.82	0.92	0.10	5000	0.95
pH 8	0.67	4.09	500	0.83	0.99	0.10	5000	0.95
pH 10	0.91	6.99	294	0.83	0.95	0.46	1111	0.95
pH 12	0.98	2.43	833	0.83	0.99	0.25	2000	0.95

When Table 2 is examined, according to the Freundlich Isotherms for K_0 and K_1 clays at 25 ^oC, the highest R^2 value for K_0 clay is at pH 12 (R^2=0.98), and for K_1 clay it is at pH 8 and pH 12 (R^2=0.99) was obtained. In other words, it has been determined that basic conditions are more suitable for Freundlich Isotherms, as in Langmuir Isotherms. K_f Freundlich constants vary between 4.92-4.95 mg/g for K_0 clay and 19.44-19.82 mg/g for K_1 clay. The constant "n" gives information about the adsorption density. The fact that the value of n is greater than 1 is evidence that the adsorption is physical. However, since the value of n is not between 0 and 1, it can be said that adsorption does not comply with the Freundlich Isotherm.

Table 2. Freundlich Isotherms for 25 ^oC temperature

	K_0 Clay			**K_1 Clay**		
25 ^0C	**Freundlich Isotherms**	**Freundlich Isotherms Constants**		**Freundlich Isotherms**	**Freundlich Isotherms Constants**	
	R^2	K_f (mg/g)	n	R^2	K_f (mg/g)	n

pH 2	0.98	4.93	48.07	0.93	19.65	57.47
pH 4	0.90	4.92	42.91	0.92	19.44	41.84
pH 6	0.95	4.92	57.47	0.97	19.80	149.25
pH 8	0.92	4.93	78.12	0.99	19.82	178.57
pH 10	0.97	4.94	72.46	0.98	19.80	72.99
pH 12	0.99	4.95	128.20	0.99	19.79	104.16

3.2. Adsorption isotherms for 35 ^{o}C temperature

Figure 8- Figure 13 shows the adsorption isotherms obtained for K_0 and K_1 clays at all initial pH values at 35 ^{o}C. It was determined that K_1 values were higher and K_0 values were lower.

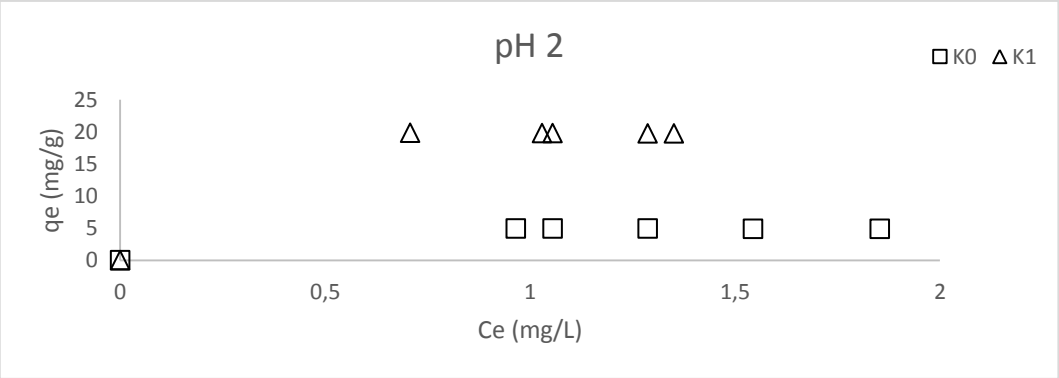

Figure 8. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 2

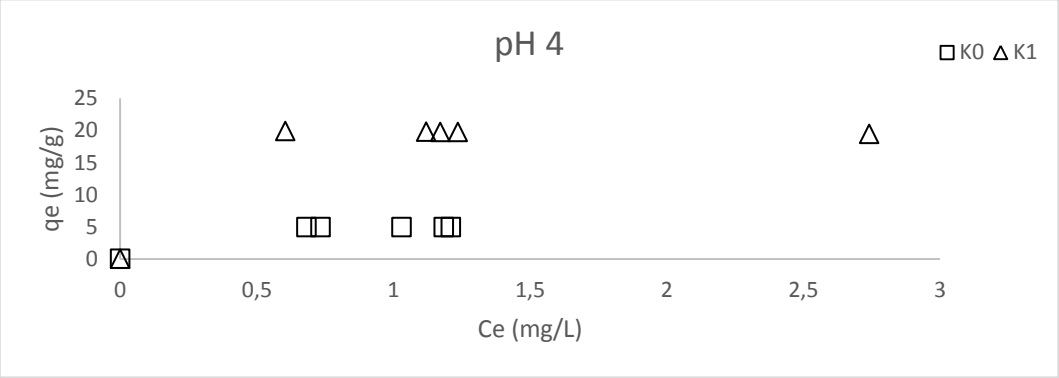

Figure 9. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 4

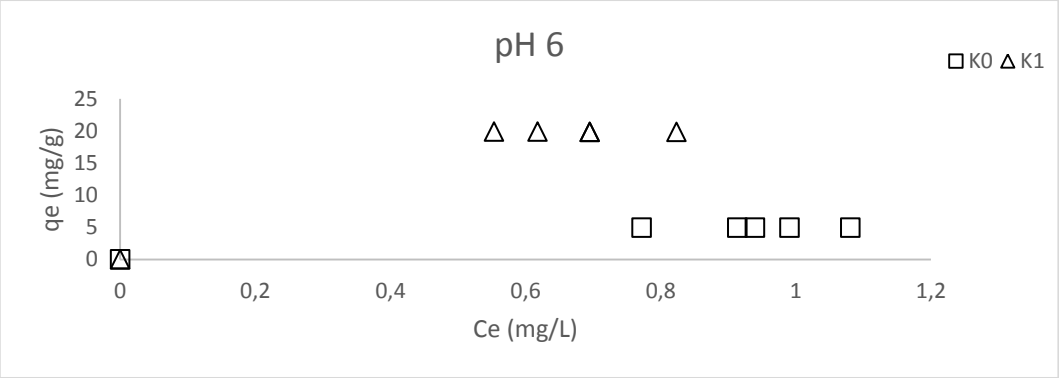

Figure 10. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 6

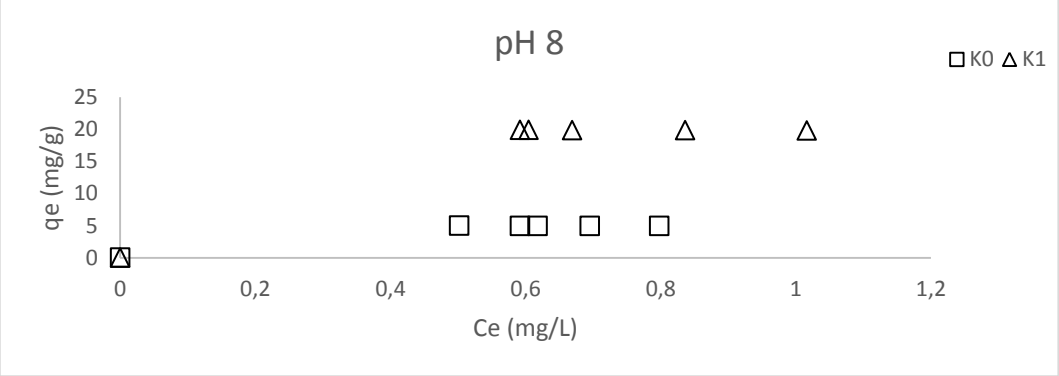

Figure 11. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 8

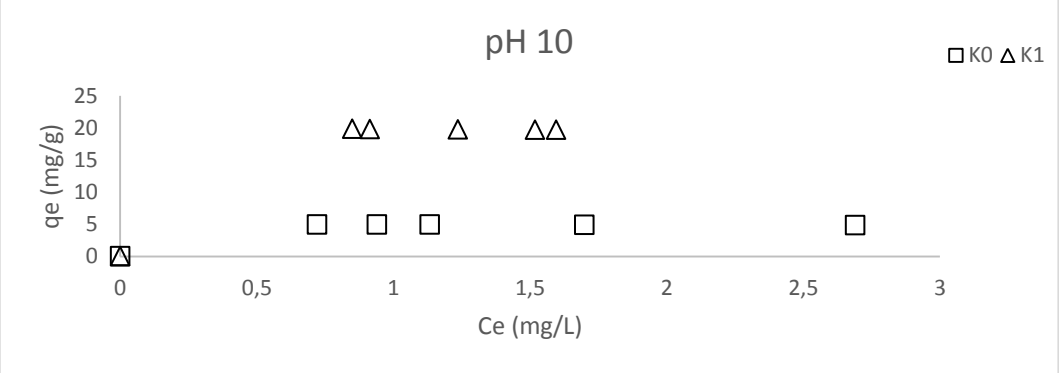

Figure 12. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 10

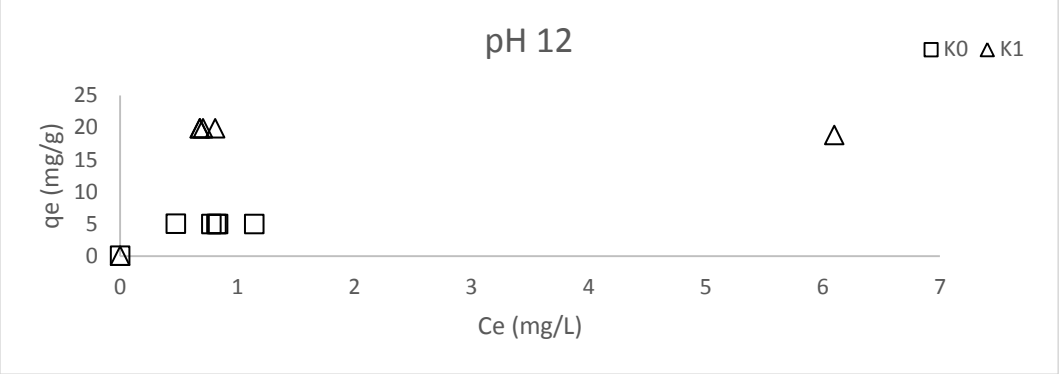

Figure 13. Change in the amount of substance adsorbed by K_0 and K_1 clays at pH 12

When the isotherms and constants given for K_0 and K_1 clays in Table 3 are examined, the highest R^2 value is at pH 4 and 6 (R^2=0.98) for K_0 clay and at pH 6, 8 and 10 for K_1 clay (R^2= 0.98) was obtained. Therefore, it is possible to say that the Langmuir Isotherm is more applicable for K_0 clay in acidic conditions. When the constant b, that is, the adsorption energy values between adsorbent and adsorbate, is examined, it is seen that this value is higher in the analyses performed with K_0 clay and the b value varies between 1.62×10-4 – 7.42×10-4 L/mg. When Q_{max} adsorption capacities are examined, it is possible to say that the adsorption capacity of K_1 clay is generally higher than that of K_0 clay. No significant relationship was found between pH change and adsorption capacity. It is seen that the R_L values for K_0 and K_1 clay vary between 0.82-0.95, that is, between 0 and 1, under all pH conditions. Therefore, no external energy is needed for adsorption. Accordingly, it was determined that the analyses were in compliance with the Langmuir Isotherm.

Table 3. Langmuir Isotherms for 35 ^oC temperature

	K_0 Clay				**K_1 Clay**			
35 ^0C	**Langmuir Isotherms**	**Langmuir Isotherms Constants**			**Langmuir Isotherms**	**Langmuir Isotherms Constants**		
	R^2	b×10^{-4} (L/mg)	Q_{max} (mg/g)	R_L	R^2	b× 10^{-4} (L/mg)	Q_{max} (mg/g)	R_L
pH 2	0.96	7.42	277	0.82	0.96	5.00	2000	0.95
pH 4	0.98	3.46	588	0.83	0.73	0.41	1250	0.95
pH 6	0.98	3.46	588	0.83	0.98	0.10	5000	0.95
pH 8	0.97	1.62	1250	0.83	0.98	0.15	3333	0.95
pH 10	0.86	7.84	263	0.83	0.98	0.35	1428	0.95
pH 12	0.90	2.03	1000	0.83	0.97	1.23	434	0.95

When Table 4 is examined, according to the Freundlich Isotherms for K_0 and K_1 clays at 35 ^oC, the highest R^2 value for K_0 clay is at pH 2, 4, 6, and 8 (R^2 = 0.99), while for K_1 clay it is at pH 6, 8, 10 and 12 (R^2=0.99). Therefore, it is possible to say that acidic conditions are suitable for K_0 clay and basic conditions are suitable for K_1 clay. K_f Freundlich constants vary between 4.94-4.95 mg/g for K_0 clay and 19.78-19.81 mg/g for K_1 clay. Since the value of n is not between 0 and 1, it can be said that adsorption does not comply with the Freundlich Isotherm.

Table 4. Freundlich Isotherms for 35 ^{o}C temperature

	K_0 Clay			K_1 Clay		
35 ^{0}C	**Freundlich Isotherms**	**Freundlich Isotherms Constants**		**Freundlich Isotherms**	**Freundlich Isotherms Constants**	
	R^2	K_f (mg/g)	n	R^2	K_f (mg/g)	n
pH 2	0.99	4.95	72.99	0.98	19.79	100
pH 4	0.99	4.94	107.52	0.93	19.78	68
pH 6	0.99	4.95	108.69	0.99	19.81	147.05
pH 8	0.99	4.95	156.25	0.99	19.80	128.20
pH 10	0.96	4.94	65.78	0.99	19.79	84.03
pH 12	0.97	4.95	133.33	0.99	19.68	38.61

CONCLUSION

For the removal of Maxillon Golden Yellow GL EC 400% dye from an aqueous solution, the adsorption isotherms of non-toxic natural clay (K_0) and clay modified with ultrasonic sound waves (K_1) were examined. The study results are summarized as follows:

-Experimental studies showed that natural clay (K_0) can be used as an alternative adsorbent for the removal of MGY dye from aqueous solutions.

-When adsorption isotherms were examined, it was determined that K_1 clay had a higher adsorption capacity under all conditions compared to K_0 clay. It has been determined that Ultrasonic sound waves increase the adsorption capacity of the material due to the surface expansion for adsorption by increasing the pores on the surface of the clay.

-When modification is applied with the ultrasonic sound method, the amount of adsorbent required to be applied for adsorption will decrease, and thus the cost of the adsorption analysis will decrease. This can be used as an advantage in the treatment of large volumes of colored wastewater. It was determined that the most efficient adsorption for K_0 and K_1 clays was obtained at the 300th minute under all conditions and as the contact time increased, the adsorption capacity increased.

-It was determined that the increase in adsorption temperature had a negative effect at pH 2 and pH 4. Therefore, it is possible to say that the interaction between dyestuff and clay was weak when pH was 2 and pH 4. The reason for this can be explained as under low pH conditions, H^+ ions cover the gaps in the surface area and hinder the capture capacity of MGY 400. It has been determined that the most suitable temperature for color removal with K_0 clay and K_1 clay is 25^{o}C for pH values 2, 4, 6 and 8, and 35 ^{o}C for pH values 10 and 12.

-When the suitability analyses for Langmuir and Freundlich isotherms were made, it was determined that adsorption studies were more suitable for Langmuir Isotherms. It has been determined that as pH increases, the energy between the adsorbent and adsorbate decreases, the energy between the adsorbent and adsorbate is higher at 25 ^{o}C and 35 ^{o}C temperatures, the adsorption falls into the physical adsorption class, and there is no need for external energy for adsorption to occur for both clay types.

-It is possible and feasible for the ecosystem to provide low-cost treatment of color-containing wastewater and discharge it into the natural environment by increasing the contact time of clay obtained from Avanos District of Nevşehir Province and modified with ultrasonic sound wave, with a continuous system at natural ambient temperatures.

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