Original Research

Removal of Crystal Violet Dye from Wastewater by Solidified Landfilled Sludge and its Modified Products

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Abstract

This study addresses the removal of crystal violet (CV) from aqueous solutions using solidified land-filled sewage sludge and its modified products as adsorbents. After the sludge was characterized using instrumental techniques (SEM, FTIR, and TG-DTA), adsorption studies were performed in a batch system, and the effects of various experimental parameters were evaluated upon CV adsorption. The results revealed that more irregular pores, higher surface roughness, and a greater content of oxygen functional groups formed in adsorbents derived from direct incineration (SC). Batch experiments revealed that stirring intension had the least effect on CV adsorption. With increasing adsorbent dosage, the CV removal efficiency increased, but the opposite result was observed for the effect of particle size. CV adsorption onto the three adsorbents was a rapid adsorption process and that onto SC was the most rapid, with the first-order kinetic model best describing the adsorption. The equilibrium data fit the Landgmiur isotherm models best, whereas much higher CV adsorption capacity and better adsorption strength onto SC were found. Desorption studies showed that acid solution was beneficial to the desorption process and the desorption rate of acetic acid reached up to about 55%. Those results proved that the solidified landfilled sewage sludge previously modified by incineration treatment was an effective adsorbent for CV removal from aqueous solution.

Keywords: solidfied landfilled sludge, adsorption, desorption, crystal violet

Introduction

As a result of rapid industrialization, especially the printing and dyeing industries. The treatment of wastewater containing some toxic organic compounds has been an urgent matter in China [1]. Crystal violet (CV) is a well-known toxic cationic dye and belongs to the class of triarylmethane dyes widely used for textile dying, food additives, pharmaceutical industries, cosmetic, plastic, and paper printing [2]. The presence of dyes even in a very low concentration in water (less than 1 ppm for some) is highly

visible and undesirable [3]. In extreme cases, it may cause vomiting, shock, jaundice, and tissue necrosis and may lead to respiratory and kidney failure [4]. Therefore, it is necessary to eliminate CV from wastewater in order to at protect both water resources and human health.

The various treatment methods include adsorption [5], electrochemical oxidation [6], and biological treatment [7] which, are available for the removal of dye from wastewater. Among these methods, the adsorption process is widely used in wastewater treatments with the advantages of simplicity in operation and good removal efficiency. However, the relatively expensive cost has hindered the development of activated carbon for wastewater treatment [8]. Exploitation of affordable and efficient adsorbent

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рН	9.18	Al ₂ O ₃ (%)	6.58
EC (ms/cm)	1.79	SiO ₂ (%)	22.3
Organic matters (%)	19.2	CaO (%)	26.9
Total nitrogen (%)	1.10	TiO ₂ (%)	0.36
Total phosphorus (%)	0.71	MnO (%)	0.04
Total potassium (%)	1.17	Fe ₂ O ₃ (%)	2.89
Cation exchange capacity (cmol/Kg)	0.17	SrO (%)	0.03
MgO (%)	17.7		

Table 1. The basic physical and chemical properties of natural solidifying landfilled sludge.

has gradually entered the people's vision and become a research focus. Currently, many investigators have focus on the feasibility of the low-cost adsorbents prepared from cheap materials such as waste apricot [9], tomato paste waste [10], red mud [11], etc. Solidified landfilled sewage sludge is produced from landfilling [12], resulting from being solidified before landfill due to its high moisture content and poor mechanics [13]. Since solidified landfilled sludges are abundant, low-cost and occupying spaces [14], recent reports have concentrated on utilization of solidified landfilled sewage sludge as adsorbents for wastewater treatment [15] and for oil production by pyrolysis [16]. The main objective of this investigation was to explore the potential of solidified landfilled sludge as a lowcost adsorbent for the treatment of dye wastewater. The effects of modification method, adsorbent dosage, stirring intension, and particle size on CV adsorption rate has been investigated. Adsorption kinetics, isotherms, and potential desorption efficiency also were evaluated.

Materials and Methods

Preparation and Characterization of Adsorbent Material

The solidified landfilled sewage sludge (SA) was obtained from the Laogang landfill in Shanghai, China, a

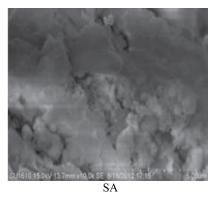
sample first collected as original sludge from Bailonggang wastewater plant in Shanghai, China, solidified by $\rm M_{\rm l}$ solidifier (Shanghai Tongji Construction Co., Ltd and Tongji University), and then processed through the landfill. It was sampled from 20-30 cm deep under the mud surface one year after the landfill was closed, taken back to the lab in a plastic bag, and finally dried, crushed, and sieved to obtain particles size of less than 2 mm. The basic physical and chemical properties of SA are as seen in Table 1.

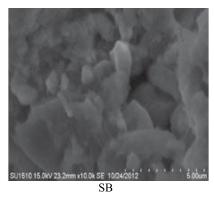
Modified adsorbents of solidified landfilled sewage sludge include previously ZnCl₂- and H₂SO₄-activated adsorbents (SB), and directly incinerated adsorbent (SC) SB was produced by mixing SA with activation solution containing 5 mol/L H₂SO₄ and 5 mol/L ZnCl₂ with volume ratio being 2:1, and the impregnation ratio of solidified landfilled sewage sludge and the activation solution being 1:2.5, then stood quiescently for 24 hr, dried in a drying oven at 100°C for 24 hr, and pyrolyzed in the muffle furnace at 550°C for 1 hr. SC was made from direct pyrolysis of SA in a muffle furnace at 550°C for 1 hr. The production rate of SB and SC was similar, being about 75%. All the prepared modified adsorbents were cooled, ground, and sieved in order to obtain particle size of ≤0.15 mm, ≤0.25 mm, ≤1 mm, ≤2 mm, and the pH value of the prepared SB and SC was 5.94 and 11.76, respectively.

Scanning electron microscopy (SEM) (SU1210; Hitachi Limited, Japan) analysis was carried out to disclose the surface texture and morphology of SA, SB, and SC. Fourier transform infrared spectroscopy (FTIR) spectra SA, SB, and SC were obtained to determine the surface functional groups by using Nicolet Magna-380 FIIR spectrophotometer (Thermo Nicolet, USA) in the range of 3200-400cm⁻¹. Thermogravimetric analysis at 900°C was achieved in a thermogravimetric equipment DTG-60 thermal analyzer (Shimadzu, Japan) under nitrogen atmosphere. 10 mg samples were heated at 10°C/min up to 900°C using a nitrogen flow rate of 50 mL/min. Two replicates of thermogravimetric analysis were performed.

Batch Adsorption Studies

Crystal violet (CV, analytically pure) with relative molecular mass being 407.99 was obtained from Hunan Chemical Reagent Factory, China. The stock solutions





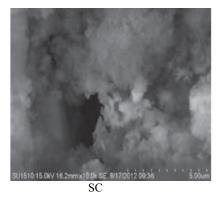


Fig. 1. Micromorphological characteristics of natural solidifying landfilled sludge (SA) and its modified contents (SB, SC).

of CV was prepared by appropriate dilution with deionized water to a final concentration of 2000 mg/L, and the desired experimental concentrations were prepared by diluting the stock solution. The concentration of CV was measured using UV spectrophotometry (UV-1100; Mapada, China) at 586 nm wavelength [1], the change in the concentration of the dye before and after adsorption gave the dye uptake. Each experiment was duplicated under identical conditions.

Effect of Adsorbent Dosage

In order to evaluate the effect of adsorbent dosage on CV uptake, this parameter was performed at 0.5, 1, 2, 3, 4 g/L. Adsorbent was mixed with 1 L CV solution (200 mg/L) in a 1 L beaker and was stirred at 100 r/min for 1 hr.

Effect of Stirring Intension

To check the effect of stirring intension on CV adsorption, 1 L samples consisting of a portion (2 g) of adsorbents and initial CV concentration of 200 mg/L were poured into 1 L beaker stirred at various stirring rates of 50, 100, 150, 200, 250 r/min respectively for 1 hr.

Effect of Particle Size

Aimed at determining the effect of particle size, 2 g of adsorbent dosage at various particle sizes 0.15, 0.25, 1.0, and 2.0 mm were poured into 1 L CV solution (200 mg/L), and stirred at 100 r/min for 1 hr.

Equilibrium and Kinetics Studies

The equilibrium adsorption isotherm was determined using batch studies. 1000 mL samples consisting of a portion (1.0 g) of the adsorbent and various initial dye concentrations 10-480 mg/L were poured into the 1L reaction beakers and stirred at 100 r/min for 1 hr. Kinetics

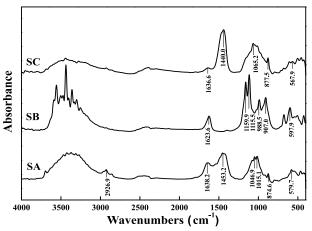


Fig. 2. The FIRT spectra of experimental sludges (SA, SB, and SC).

measurements were conducted by analyzing the different CV concentrations to determine the CV uptake at various time intervals (1, 3, 5, 10, 20, 30, 60, 120 min). For the kinetic studies two different adsorbent dosages, i.e., 0.5 and 3.0 g, and two initial CV concentrations, i.e., 100 and 200 mg/L, were taken into consideration.

Desorption Studies

The CV-loaded biomass of SA, SB, or SC, which was initially exposed to 200 mg/L of CV at pH 6.0 and dosage 0.1g, was contacted with 50 mL samples consisting of distilled water, 0.10 mol/L NaOH, 0.1 mol/L HAc, or 1.0 mol/L HAc as the dye-desorbing agent for 60 min on a rotary shaker at 100 r/min. Then the adsorbent SA, SB or SC was removed by centrifuge and the quantity of CV recovered was determined. The CV concentrations were measured using UV absorption.

Calculation Method

The amount of CV adsorbed onto various adsorbents, qe (mg/g), its desorption amount, X (mg/g), and removal efficiency (%) were calculated using the following equations, respectively:

$$q_e = (c_0 - c_e)V/w \tag{1}$$

$$X=c_{e}V/w$$
 (2)

Removal efficiency =
$$100*(c_0-c_0)/c_0$$
 (3)

...where q_e is the adsorption capacity, c_o and c_e are, respectively, the initial and equilibrium liquid-phase concentrations of CV (mg/L), V is the volume of CV solution (L), and w is the weight of adsorbents (g).

Results and Discussions

Characterization of Adsorbents

Surface Morphology Analysis

Fig. 1 shows the SEM photographs of SA, SB, and SC at 5,000 x magnification. It can be found that SA possessed a rough surface and smaller pores, but SB and SC had more irregular pores, and their surface roughness increased obviously, which suggests erosion occurring on the surface of SB and SC, and it was likely that these larger and rough pores were produced due to activating agent or incineration. Moreover, much more lamellar and crystal pieces were shown in a SEM photograph of SB which could be assigned to much more inner clay particles or metal compounds, while the surface of SC presented much bigger irregular pores. The difference in the surface of various adsorbents would lead to the different adsorption capacity of crystal violet.

FTIR Analysis

The FTIR spectra of SA, SB, and SC were shown in Fig. 2. Based on related reports and map analysis [1, 17], peaks at 3,650-3,200 cm⁻¹ of wavenumber corresponded to the O-H stretching vibration of bound hydroxyl groups, the bands of 1,670-1,650 cm⁻¹ and 1,460-1,400 cm⁻¹ were normally related to the C=O stretching, which can be attributed to the hydroxyl group from primary alcohol and secondary alcohol [18]. As shown in Fig. 2, many more peaks at 3,650-3,200 cm⁻¹ were found in spectra of SB, which signified that many more bound hydroxyl groups originated from previous ZnCl, activation or acidity of H₂SO₄[19]. However, bands at both 1,670-1,650 cm⁻¹ and 1,460-1,400 cm⁻¹ only existed obviously in SC followed by SA, which suggests more presence of basic oxygencontaining functionalities such as carboxyl groups, aliphatic hydrocarbons, and aromatic hydrocarbons [20]

in SC followed by SA. Moreover, the absorption band of 1,200-1,020 cm⁻¹, which were assigned to either Si-O-Si or Si-O-C structures, indicates the association of silicon content, and the C-H bending vibration in the region of 1,000-670 cm⁻¹ was obviously observed in all three adsorbents. It could be obtained that the difference in bound hydroxyl groups and oxygen-containing functionalities may contribute to CV adsorption.

Thermal Analysis

Fig. 3 shows the thermogravimetric (TG) and derivate thermogravimetric (DTG) thermograms of SA, SB, and SC. According to the TG curves, the total mass loss of SA was 38.48%; however, the total weight loss of SB and SC were approximately 9.81% and 10.21%, respectively, which might be caused by their pyrolysis at 550°C for 1 hr during preparation. As shown in Fig. 3, the weight

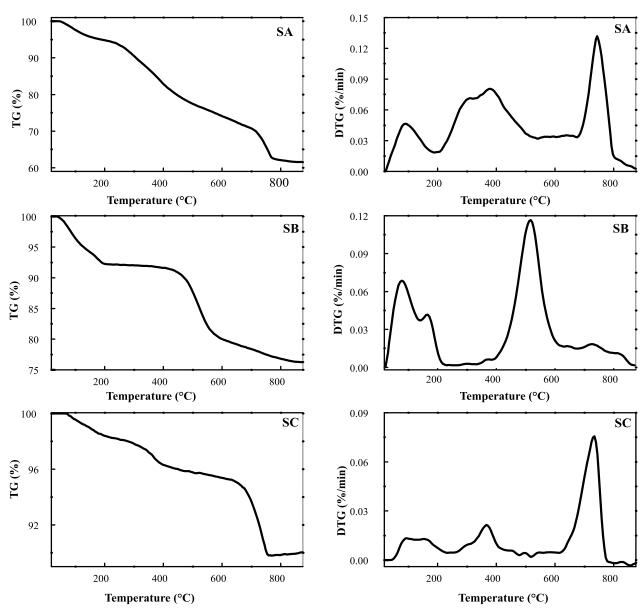


Fig. 3. TG, DTG curves of experimental sludges (SA, SB, and SC).

loss of different adsorbents mainly concentrated at four stages: 1) The moisture evaporation stage happened at approximately 90°C; 2) The second stage happened in the region of 200-400°C, which was ascribed to the breakage of C-C from a carbon-containing compound [21], suggesting the decomposition of a thermolabile component such

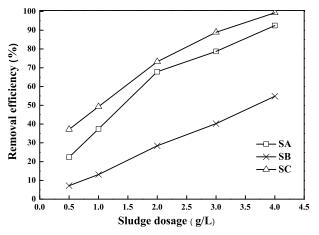


Fig. 4. Effect of sludge dosage on the adsorption of crystal violet.

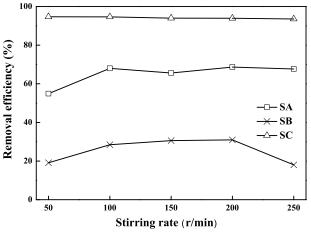


Fig. 5. Effect of stirring rate on the adsorption of crystal violet.

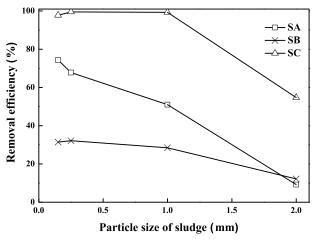


Fig. 6. Effect of sludge particle size on the adsorption of crystal violet.

as carboxyl groups of the organic matter with complex structures [22]; 3) The third fraction decomposed at 400-600°C, which might be evidence for the decomposition of high thermostability oxygen functional groups such as quinines and phenolic hydroxyl group [23]; 4) The weight loss in the region of 600-900°C might be caused by the decomposition of some inorganic substance or the breakage of residual C-C [22]. Further comparison of different thermograms showed that the weight loss of both SA and SC concentrated at temperatures of 80, 200-400 and 600-900°C, while rapid weight loss of SB was mostly found at 80 and 400-600°C, which showed the existing evidence of some carboxyl groups and inorganic substances in SA and SC, while much more high thermostability oxygen functional groups such as phenolic hydroxyl group exists in SB, which was consistent with the FTIR analysis and may contribute to CV adsorption.

Adsorption Studies

Effect of Adsorbent Dosage

The effect of adsorbent dosage on the removal of CV is shown in Fig. 4. The removal efficiency of CV increased with the inceasing adsorbent dosage over the range of 0.5-4.0 g/L, and the removal efficiency of CV increased from 22.35 to 92.5%, 7.17 to 54.82%, and 37.17 to 99.27% for SA, SB and SC, respectively, which could be ascribed to the increase of available surface area and adsorption sites accompanied by the increasing adsorbent dosage [1]. Moreover, the differences of CV adsorption were obviously followed by SC>SA>SB, and the highest removal efficiency for SC could reach up to 99% when the adsorbent dose of 4.0 g/L was used. Recent reports showed that the amount of dye sorbed by dairy sludges approached 99% when sorbent dose of 7.0 g/L was used [24]. Thus, we concluded that adsorbents from SC gave better adsorption capacity and much higher removal efficiency for CV in solution.

Effect of Stirring Intension

As shown in Fig. 5, it was clearly observed that stirring intension indicated an unobvious effect on CV adsorption. The removal efficiency of CV by SC mainly kept around 95% at the experimental condition. However, the removal efficiency of CV by SA and SB was subtle and varied from 55-65% and 15-25%, respectively. The results explained that stirring intension had little effect on CV removal in solution, and a similar observation was obtained for crystal violet adsorption on activated sludge [25].

Effect of Particle Size

A recent article reported that particle size played an important role in the amount of dye adsorbed [17]. The adsorption experiments were carried out using various particle sizes from 0.15 to 2 mm and the results were

Table 2 Completive as	afficients of four	. Irimatiaa aassatiam	a fam amyatal via	let by different sludge.
ranie 2. Correlative co	berncients of four	Kinetics eduation	s for crystal vic	net by antierent sluage.

Sludge		Crystal violet	Elovich equation	Langmuir kinetic equation	First order equation	Two-constant equation	
Sludge quality (g)	concentration (mg/L)	Y = a + blnt	$Y = a + blnt t/Y = t/Y_{max} + 1/k$		lnY = a + blnt		
	0.5	100	0.9851**	0.8747**	0.9851**	0.9843**	
C A		200	0.9704**	0.8133**	0.9768**	0.9770**	
SA	2	100	0.9553**	0.9748**	0.9968**	0.8392**	
	3	200	0.8938**	0.9908**	0.9974**	0.8708**	
		100	0.9134**	0.8241*	0.9249**	0.9232**	
CD	0.5	200	0.8608**	0.4148	0.8158*	0.8290*	
SB 3	100	0.9921**	0.9101**	0.9948**	0.9917**		
	200	0.9918**	0.7944*	0.9940**	0.9919**		
	0.5	100		0.8554**	0.8974*	0.8903**	0.8577**
0.5	200	0.8380**	0.9357**	0.9142**	0.8348**		
SC 3	2	100	0.8762*	0.8561**	0.8112*	0.8892*	
	3	200	0.8255**	0.9704**	0.9846**	0.8254*	

Y represents adsorption capacity (mg/g); t represents contact time (min); a, b, c all represents the model parameter; R represents correlation coefficient, the larger R value, the better the model; * represents p<0.05, significance level; ** represents p<0.01, very significance level.

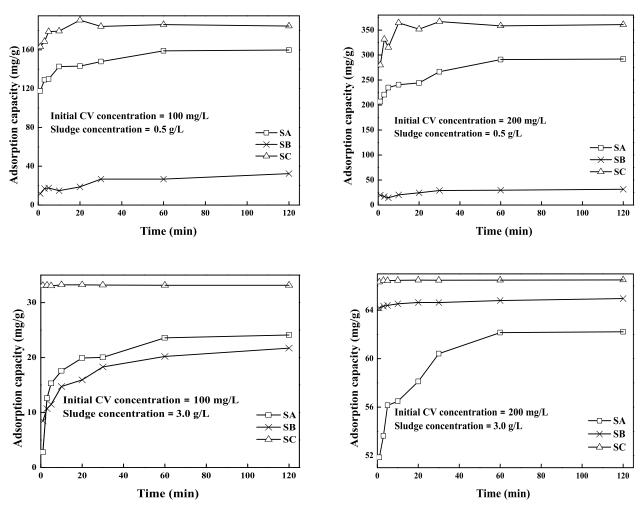


Fig. 7. Dynamic curves of crystal violet adsorption on experimental sludges.

	Henry model		Freundlich model			Langmuir model			
Adsorbents	G=A+K _h C _e			$G = K_f C_e^{1/n}$			$G = G_{o}K_{L}C_{e}/(1 + K_{L}C_{e})$		
	A	K _h	R	K _f	1/n	R	G_{0}	K _L	R
A	11.75	0.083	0.9602**	4.56	0.315	0.9745**	26.75	9.64	0.9770**
В	12.18	0.036	0.8259*	4.39	0.316	0.9692**	22.94	11.98	0.9934**
С	16.92	0.973	0.9515**	3.12	0.793	0.9422**	124.37	50.50	0.9702**

Table 3. Isotherm model parameters for CV adsorption onto adsorbents.

R represents correlation coefficient, the larger R value, the better the model. * represents p<0.05, significance level; ** represents p<0.01, very significance level.

presented in Fig. 6, and the removal efficiency of CV increased with decreasing particle size of adsorbents. Chu and Chen [26] indicated that the decrease in particle size caused an increase in the sludge surface area, and Mittal et al. [17] suggested that the decrease in particle size would promote the accessibility of the adsorbent pores for the dye. Moreover, other investigators [27] indicated that such similar results show that external transport limited the rate of adsorption, and that powdered adsorbent would be advantageous over granular particles and may produce a shorter time to equilibrate. Furthermore, as shown in Fig. 6, a much bigger effect on CV adsorption was caused by particle size of SA than that of SB and SC. And particle sizes of 0.15-1.00 mm were found to yield much higher removal efficiency for CV in solution.

Adsorption Kinetics

The CV uptake by SA, SB, and SC as a function of time was depicted in Fig. 7. It was clear that the adsorption capacity of CV increased rapidly at first, and then the process slowed down and approached a balance with a lapse of time. This result revealed the availability of readily accessible surface sites at the initial stage, and gradually decreased when reaching equilibrium [28]. From the Fig. 7 it can be seen that the uptake of CV rapidly increased in the first 20 min. of the adsorption process

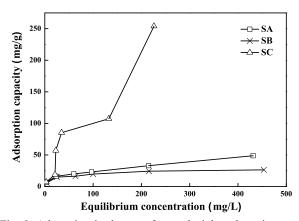


Fig. 8. Adsorption isotherms of crystal violet adsorption onto experimental adsorbents.

and reached equilibrium with dosage of 0.5 g/L for all three adsorbents. Then, with increasing contact time, the adsorption capacity would keep balance and the dynamic curve became flat gradually. However, under the adsorbent dosage of 3 g/L, it can be seen that the uptake of CV by SC reached equilibrium at the first five min. while the uptake of CV reached equilibrium at the first 60 min. both for SA and SB, regardless of the initial CV concentration. Thus it could be concluded that the CV adsorption onto all the three adsorbents is a rapid adsorption process and that CV adsorption onto SC was the most rapid.

The kinetics provide valuable insights of the adsorption reaction mechanisms [29]. And many attempts have been made to formulate a general expression describing the kinetics of adsorption reaction [30]. In order to characterize the adsorption process of the dyes onto solidified landfilled sludges and its modified product, four different kinetic models were investigated, namely the Elovich equation, Langmuir kinetic equation, first- order equation, and twoconstant equation. The calculated parameters were listed in Table 3. The correlation coefficient, R² value, was used to evaluate the applicability of the kinetics equation, the better fitting result, the larger the correlation coefficient R². The R² values in Table 2 suggested that the first-order equation best described the kinetics of CV adsorption irresponsible of adsorbent dosages and initial CV concentrations. In addition, further investigations showed that not only the initial CV concentration (Fig. 7), but also the adsorbent dosage could affect the CV adsorption rate, indicating that the mass transfer of adsorbate molecules from the bulk solutions to the adsorbent surface may govern the ratecontrolling step during the CV adsorption process.

Adsorption Isotherms

The equilibrium isotherm is important for model building in the design of adsorption systems [2]. As presented in Fig. 8. The CV adsorption capacity increased with increasing initial CV concentration, and the CV adsorption capacity of SC increased sharply and was much higher than that of others under high initial CV concentration. Three models, including Henry, Langmuir, and Freundlich were employed to fit the data and describe the adsorption equilibrium. They were expressed in Eqs. (4), (5) and (6), respectively.

$$G = A + K_h C_e \tag{4}$$

$$G = G_{o}K_{T}C_{e}/(1 + K_{T}C_{e})$$
 (5)

$$G = K_f C_e^{1/n} \tag{6}$$

...where G is the amount of CV adsorbed at equilibrium, C_{\circ} is the adsorbate equilibrium concentration, G_{\circ} is the maximum adsorption capacity according to the Langmuir model, K_{h} , K_{L} and K_{f} signals the constants for Henry Langmuir and Freundlich models, respectively, and n is the reciprocal reaction order for Frundlich models.

As shown in Table 3, both Henry and Langmuir isotherm models provided reasonable fits to analyze the characteristics of CV adsorption onto experimental adsorbents. However, Freundlich models couldn't fit very well based on present reports that only the value of n > 1represents a favorable adsorption condition [31]. The applicability of the models was also established from the correlation coefficient, R. The results showed that the correlation coefficient for the Langmuir models were between 0.9770 and 0.9934, while those of the Henry models were between 0.8259 and 0.9602, Compared with the R value, it could be concluded that the Langmiur model best describes CV adsorption. This could speculate that the homogeneity model was more favorable for CV adsorption onto the surface of SA, SB, and SC. Moreover, the maximum adsorption capacities (G) were about 26.75, 22.94, and 124.37 mg/g for SA, SB and SC, respectively. These results predicted that the conventional modification of solidified landfilled sludge by ZnCl₂ and H₂SO₄ might be not efficacious for improving the removal efficiency of CV in solution. On the contrary, the removal efficiency of CV in wastewater could be developed by directly incineration of solidified landfilled sludges.

Desorption Studies of CV Adsorbed

Recovery of CV from exhausted biomass of SA, SB, and SC was tested using aqueous solutions of distilled

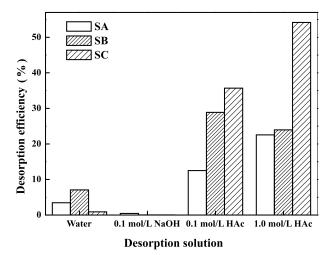


Fig. 9. Desorption efficiency of crystal violet from solidifyinglandfilled sludge and its modified products.

water, 0.10 mol/L NaOH, 0.1 mol/L HAc or 1.0 mol/L HAc. Fig. 9 revealed that the desorption efficiency of CV was followed by acetic acid > distilled water > sodium hydroxide, which indicated that the alkaline solution restrained while the acetic acid promoted CV desorption from the three adsorbents. Moreover, the desorption rate increased obviously for SA and SC while decreasing for SB when the concentration of acetic acid increased from 0.1 mol/L to 1.0 mol/L, and the desorption rate of CV from SC reached up to about 55% with 1.0 mol/L acetic acid used as desorption solution. Since the desorption rate represents the rapidity of active surface sites contacted with the dye [12, 32], the highest desorption efficiency of CV from SC in solution of 1.0 mol/L acetic acid may be explained by its rapid adsorption kinetics (Fig. 7) and its much more carboxyl groups or its higher inorganic contents (Fig. 2 and Fig. 3), such as oxides, which can be dissolved by acid.

Conclusion

As a by-product of wastewater treatment, sewage sludge is commonly treated with the landfilling method. However, the problems encountered with landfilling are its high moisture content, poor mechanics, and space limitations. Since sewage sludge is often solidified with a dehydrating agent before landfilling, the development of a method to recycle the solidified landfilled sewage sludge (SA) to conserve space and reduce costs is very important. It was revealed that the treatment of solidified landfilled sewage sludge with ZnCl₂ and H₂SO₄ activation (SB) or direct incineration (SC) resulted in the formation of more irregular pores and higher surface roughness, especially on the surface of SC. Compared with SA, SB possessed more high thermostability bounded hydroxyl groups while SC had more oxygen-containing functionalities. CV adsorption was considerably affected by the initial adsorbent dosage and particle size, but not the stirring intension. And CV adsorption onto the three adsorbents was a rapid adsorption process, with CV adsorption onto SC being the most rapid and the first-order kinetic model provided the best fit. Moreover, the Langmuir isotherm model fit the experimental data best, based on Langmiur model, the practical limiting adsorption capacities (Q) of SA, SB, and SC were found to be 26.75, 22.94 and 124.37 mg/g, respectively, exhibiting much higher CV adsorption capacity and better adsorption conditions for SC. the desorption rate of CV adsorbed onto SC by acetic acid was the highest, which reached up to about 55% and may be explained by its rapid adsorption kinetics and its much more carboxyl groups or its higher inorganic contents such as oxides, which can be dissolved by acid.

In summary, solidified landfilled sewage sludge and its modified products have the potential for CV removal. In particular, the direct incinerated sludges can achieve greater CV adsorption capacities. However, further study is still needed to determine the ability of solidified landfilled sewage sludge to act as an adsorbent for other pollutants.

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References

- SENTHILKUMAAR S., KALAAMANI P., SUBBURAAM C. V., Liquid phase adsorption of crystal violet onto activated carbons derived from male flowers of coconut tree. J. Hazard. Mater., 136 (3), 800-808, 2006.
- MA W., SONG X. Y., PAN Y. Q., CHENG Z.H, XIN G., WANG B.D., WANG X.G., Adsorption behavior of crystal violet onto opal and reuse feasibility of opal-dye sludge for binding heavy metals from aqueous solutions. Chem. Engin. J. 193-194, 381-390, 2012.
- RAFATULLAH M., SULAIMAN O., HASHIM R., AHMAD A., Adsorption of methylene blue on low-cost adsorbents: A review. J. Hazard. Mater. 171 (1-3), 70-80, 2010.
- KUNZ A., MANSILLA H., DURÁN N., A degradation and toxicity study of three textile reactive dyes by ozone, Environ. Technol. 23 (8), 911-918, 2002.
- GARG V. K., GUPTA R, YADAV A.B., KUMAR R., Dye removal from aqueous solution by adsorption on treated sawdust. Bioresour. Technol. 89 (2), 121-124, 2003.
- MOHAN N., BALASUBRAMANIAN N., AHMED BASHA C., Electrochemical oxidation of textile wastewater and its reuse. J. Hazard. Mater., 147 (1-2), 644-651, 2007.
- KORNAROS M., LYBERATOS G., Biological treatment of wastewaters from a dye manufacturing company using a trickling filter. J. Hazard. Mater. 136 (1), 95-102, 2006.
- GUPTA V.K., CARROTT P. J. M., RIBEIRO COARROTT M. M. L., SUHAS, Low-cost adsorbents: growing approach to wastewater treatment-a review. Critical Reviews in Environ. Sci. and Technol. 39 (10), 783-842, 2009.
- BASAR C.A., Applicability of the various adsorption models of three dyes adsorption onto activated carbon prepared waste apricot. J. Hazard. Mater. 135 (1-3), 232-241, 2006.
- FUAT G., HASAN S., GUHBAHAR A.S., FILIZ K., Decolorisation of aqueous crystal violet solution by a new nanoporous carbon: Equlibium and kinetic approach. J. Indus. Engin Chemi. 20, 3375-3386, 2014.
- LUYI Z., HUAYONG Z., WEI G., YONGLAN T., Removal of malachite green and crystal violet cationic dyes from aqueous solution using activated sintering process red mud. Appli Clay Sci. 93-94, 85-93, 2014.
- ZHU W., LI L., LIN C., Biochemical effects on permeability of solidified sludge. Chin. Rock and Soil Mechanics, 27 (6), 933-938, 2006.
- ZHENG X. J., ZHU W., LI L., XU Z. R., QU Y., Optimization experiment study of sludge solidifying materials. Chin. Rock and Soil Mechanics, 29 (S1), 571-574, 2008.
- 14. SMITH K. M., FOWLER G. D., PULLKET S., GRAHAM N. J. D., Sewage sludge-based adsorbents: a review of their production, properties and use in water treatment applications. Water Res., 43 (10), 2569-2594, 2009.
- ZHU W. Q., SHAN J. L., ZHANG Z., GUO D., ZHAO Y. C., Comparative study on adsorption behaviour of copper and zinc from aqueous solution by solidifying landfilled sludge. China Environ. Sci., 31 (9), 1503-1508, 2011.

- 16. SHI Q. Y., LI B., ZHAO Y. C., Influence factors of the low-temperature pyrolysis process for oil production of sewage sludge. Chin. Environ. Sanitation Engin. 14 (5), 4-6, 2006.
- MITTAL A., MITTAL J., MALVIYA A., KAURA D., GUPTA V. K., Adsorption of hazardous dye crystal violet from wastewater by waste materials. J. Colloid and Interface Sci. 343 (2), 463-473, 2010.
- WHITE J. L., ROTH C. B., Infrared spectrometry, in: Methods of soil analysis, Part 1, second ed., Agron. Monogr. 9. Americ. Soci. Agronomy-Soil Sci. Soci. Americ. Madison. 291-330, 1986.
- HAN R. P., DING D. D., XU Y. F., ZOU W. H., WANG Y. F., LI Y. F., ZOU L. N., Use of rice husk for the adsorption of congo red from aqueous solution in column mode. Bioresour. Technol. 99 (8), 2938-2946, 2008.
- SINGH K. P., MALIK A., SINHA S., OJHA P., Liquid-phase adsorption of phenols using activated carbons derived from agricultural waste material. J. Hazard. Mater. 150 (3), 626-641. 2008.
- BAGREEV A., BANDOSZ T.J., H₂S adsorption/oxidation on materials obtained using sulfuric acid activation of sewage sludge-derived fertilizer. J.Colloid and Interface Sci. 252 (1), 188-194, 2002.
- FRANCIOSO O., RODRIGUEZ-ESTRADA M.T., MON-TECCHIO D., SALOMONI C., CAPUTO A., PALENZO-NA D., Chemical characterization of municipal wastewater sludges produced by two-phase anaerobic digestion for biogas production. J. Hazard. Mater. 175 (1-3),740-746, 2010.
- YUE Z. R., CHRISTIAN L. M., JAMES E., Characterization of surface chemistry and pore structure of H₃PO₄-activated poly (vinyl alcohol) coated fiberglass. Carbon, 42 (10),1973-1982, 2004.
- 24. SUMANJIT, TEJINDER P. S. W., Use of dairy sludge for the removal of some basic dyes. J. Environ. Engin. and Sci. 7 (5), 433-438, **2008**.
- WANG H. J., DONG W. Y., LI W. G., YIN W. C., TAO X., Adsorption of crystal violet on activated sludge. Chin. Environ. Sci. 29 (10), 2856-2859, 2008.
- CHU H.C., CHEN K.M., Reuse of activated sludge biomass: The rate processes for the adsorption of basic dyes on biomass. Process Biochem. 37 (10), 1129-1134, 2002.
- WANG S. B., LI H. T., Kinetic modelling and mechanism of dye adsorption on unburned carbon, Dyes and Pigments, 72 (3), 308-314, 2007.
- FOO K.Y., HAMEED B.H., Coconut husk derived activated carbon via microwave induced activation: Effects of activation agents, preparation parameters and adsorption performance. Chem. Engin. J. 184, 57-65, 2012.
- CHOWDHURY S., SAHA P., Sea shell powder as a new adsorbent to remove basic green 4 (Malachite green) from aqueous solutions: equilibrium, kinetic and thermodynamic studies. Chem. Engin. J. 164 (1), 168-177, 2010.
- MONA S., KAUSHIK A., KAUSHIK C.P., Waste biomass of Nostoc linckia as adsorbent of crystal violet dye: optimization based on statistical model. Intern. Biodeterior. & Biodegrad. 65 (3), 513-521, 2011.
- MCKAY G., BLAIR H. S., GARDNER J. R., Adsorption of dyes on chitin-1: equilibrium studies. J. Appli. Polymer Sci. 27, 3043-3057, 1982.
- 32. HU X .N., NAN Z. R., WANG S. L., HUANG H., HU Z. Y., Sorption and desorption of copper, zinc and lead in the irrigated desert soil from the oasis in the arid regions, northwest China. Chin. Ecol. and Environ. Sci. 18 (6), 2183-2188, 2009.