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Effect of water treatment residuals and cement kiln dust on COD adsorption and heavy metals from textile wastewater

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ABSTRACT

Water treatment residual (WTR) is a by-product after settling of suspended particles and colloids by alum in drinking water treatment plants. The reuse of cement kiln dust (CKD) and WTR wastes for treatment purposes is a good solution to remove the dye, chemical oxygen demand (COD) and heavy metals from the textile wastewater. A batch adsorption experiment was performed to evaluate CKD and WTR at different rates, on COD adsorption, color and heavy metals from the textile wastewater. The amount of COD adsorption increased with the addition of CKD and WTR, but the highest adsorption was obtained at 2 h. COD adsorption on CKD was successfully fitted to both Langmuir and Freundlich isotherm models. The maximum adsorption of Langmuir was higher with the addition of WTR (100.0 mg g^{-1}) than CKD (14.3 mg g^{-1}). Heavy metals in wastewater treated by WTR or CKD were below Egyptian and United States Environmental Protection Agency irrigation guidelines. In this study, the addition of alum to WTR at 200 mg L^{-1} was effective in the removal of turbidity, color and COD from textile industrial effluents compared to 500 mg L^{-1} of WTR only. Therefore, WTR and alum can be used as an effective and low-cost adsorbent for the removal of inorganic and organic pollutants from textile wastewater.

Keywords: Cement kiln dust; Water treatment residual; Textile wastewater; Maximum adsorption; Chemical oxygen demand; Heavy metals

1. Introduction

The textile and clothing sector is the second-largest industrial sector in Egypt after the agro-industry. It represents around 3.5% of the GDP, 34% of the industrial output and 14% of the total Egyptian exports. Today, the textile sector is formed in Egypt from more than 3,000 companies [1]. The textile industry uses high volumes of water throughout its operations, from the washing of fibers to bleaching, dyeing and washing of finished products. On average, approximately 200 L of water are required to produce 1 kg of textiles. The large volumes of wastewater generated also contain a variety of chemicals used in all stages of treatment. These damages can cause if not treated properly before being discharged to the environment. The aquatic toxicity of wastewater from the textile industry varies greatly between production facilities. The sources of aquatic toxicity can include salt, surfactants, and ionic minerals and their components complex metals there, toxic organic chemicals, biocides, and toxins [2]. Generally, textile wastewater has a high pH value, high concentration of suspended solids, chlorides, nitrates, metals like manganese, sodium, lead, copper, chromium, iron, and high biochemical oxygen demand and chemical oxygen demand (COD) value. That may pose a toxic environmental hazard and lead to potential bioaccumulation risks, which

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may ultimately affect humans through the food chain [3]. Cotton-textile wastewater contains a variety of polluted compounds from treated chemicals, dyes such as (disperse, vat, direct, acid, basic, and azo-reactive dyes) and natural impurities extracted from cotton fibers. The remaining dyes in the discharged wastewater cause aesthetic problems and harm the quality of the receiving water. A big problem in the textile industry is the presence of color in the wastewater. The highly colored effluents from textile industries into the environment have an aesthetic negative effect and eutrophication in the receiving waters. However, it has been found that many of the dyes and degradation products used in the textile industry are toxic and genotoxic [4]. Recently, state and federal agencies in the United States have requested low effluent color limits (b200 units from the American Dye Manufacturers Institute, ADMI) [5]. Toxic metals present in textile wastewater may typically include copper, cadmium, chromium, nickel, and zinc. Sources of metals found in textile mill effluents may include fiber, incoming water, dyes, plumbing, and chemical impurities. Dyes may contain metals such as zinc, nickel, chromium, and cobalt. Metals may be difficult to remove from wastewater [6]. So, heavy metals in treated wastewater of textiles must be lower than the Egyptian guideline for irrigation before their discharge.

Many methods used to remove dyes and heavy metal ions from textile wastewater include chemical coagulation, oxidation, coagulation, ozone, membrane/nanofiltration, radiation, ion exchange, and biological treatment technologies. However, most of these technologies have defects such as insufficient removal efficiency, large sludge production, the need for high capital, operating costs, and high technology [7]. Consequently, these methods are not applicable to small industries in developing countries. The biggest challenge in implementing this strategy is the adoption of low-cost wastewater treatment technologies that will increase the efficiency of the use of limited water resources and ensure compliance with all health and safety standards regarding the reuse of treated wastewater [8].

Adsorption is an effective process for removing dye and other contaminants and one of the best methods, due to its high efficiency, low cost, and ease of operation [9,10]. Activated carbon is usually used to remove toxic pollutants from textile wastewater, but it is higher costs. Therefore, attempts are being made to develop low-cost adsorbents from agricultural and industrial waste materials [11]. As shown recently, research efforts have been directed towards the use of industrial waste as a sorbent in an effort to reduce treatment costs and protect the environment and public health [12]. Using waste products as adsorbents materials provide many benefits. It decreases the cost of removing dyes and heavy metals from textile wastewater, as well as being conducive to reducing the amounts of residuals that are accumulated in the environment.

Water treatment residual (WTR) is a by-product after settling of suspended particles and colloids by aluminum sulfate (alum) in drinking water treatment plants and is disposed of in huge amounts [13]. Egypt, water treatment plants are consuming above 365,000 tons of alum and produce more than 100 million tons of water residues per year. The common method of WTR disposal in Egypt is discharging into waterways or landfilling which can have negative environmental impacts [14]. In addition, the continued rise in WTR generation will require the construction of a new landfill, which will add to the cost. The cost of disposal (by landfill) WTR, is about $50 per metric ton, which can significantly increase the cost of drinking water treatment. Therefore, WTR can be used as a safe, inexpensive and effective sorbent to remove heavy metal and phosphorus from contaminated water or soils. It also indicates that adsorption on WTR can be used as a primary treatment for discoloration of wastewater [15]. In recent years, WTRs have been used to remove pollutants from aqueous solutions in most studies. Most research focuses on phosphorus adsorption [16,17].

Cement kiln dust (CKD) the by-product of Portland cement plants and its disposal in huge amounts [18]. CKD is a fine-grained, solid, highly alkaline particulate material. CKD production is around 30 million tons worldwide annually [19], with over 2.5 million tons per year produced in Egypt, which are hazardous and costly to dispose of [20]. The common way to get rid of CKD in Egypt is the landfill that has negative and costly environmental effects. The use of CKD in high way applications, adsorption of heavy metals and dyes, waste treatment and soil stabilization has become an attractive alternative for disposal. The heavy materials removal such as Cr by CKD is due to its high adsorption capacity [21]. Mahmoud [22] observed that CKD filters could greatly reduce heavy metals and other pollutants in the raw wastewater of textiles. CKD may be used as an immobilizing material to reduce heavy metal levels in contaminated soils as well as sewage water and sludge [23].

Drinking water and Portland cement wastes have not been used to remove toxic pollutants from textile wastewater. Thus, the main objective of this study was to evaluate the potential of drinking water and Portland cement wastes as a low-cost sorbent to remove dyes and heavy metal ions from the wastewater generated from Amriya Spinning and Weaving, Egypt.

2. Material and methods

2.1. Raw sewage and CKD samples

Textile wastewater samples were collected from El-Amriya Company for Cotton Spinning and Weaving, Alexandria Governorate, Egypt, after the wet processes. Several types of dyes used in El-Amriya Company, such as disperse (Terasil Red R, Terasil Yelly 4G, Terasil Rubin 2GFL, Terasil Black RM, Dianix Red CBN and FBN), reactive (Cibacrone blue FGFN, Cibacrone Black W-HF, Livafix Brown ERN, Livafix Navy EBNA, Livafix Red ERN, etc), vat (different type from ISMA and INDANTH) and naphtha (Naphthol A, Naphthol B, Fast RED KB and Fast Bordeaux GB). The laboratory examination was performed immediately (within 24 h) after collection to reduce any changes in the sewage characteristics. CKD was obtained from Amriya Cement Factories. CKD is an alkaline waste material (pH 12.3) and contains 14.9% SiO_2, 48.8% CaO, 4.2% Al_2O_3, 2.8% FeO_3 and 2.3% MgO, 4.8% lime-free and some alkaline salts such as sodium and potassium. The specific gravity of CKD is 2.92 and the specific surface area is 4,440 $cm^2\ g^{-1}$. WTR, which is a by-product, resulted from drinking water treatment plants. WTR was manually taken using shovels and buckets from the bottom of the sedimentation basins at Tanta City water treatment facility, El Gharbia, Governorate, Egypt, air-dried, and then passed through a 2-mm sieve prior to characterization or reuse. WTR is an alkaline (pH 7.55) and contains 6.33 meq L^{-1} Ca^{2+}, 2.82 meq L^{-1} Mg^{2+}, 32.4 mg kg^{-1} KCl-Al, 2.9% organic matter and 68% clay and the specific surface area is 1,660 m^2 g^{-1}.

2.2. Adsorption experiments

Batch adsorption experiments tested to remove the dye (color and turbidity), and heavy metals and their ability to adsorb COD from the textile wastewater. Raw textile wastewater samples were mixed with different rates of CKD and WTR at 120 revolutions per minute (rpm) using a jar test for 1, 2, 4 and 24 h at room temperature. The treated samples were allowed to settle for 15 min and the supernatant was taken to measure contaminants in textile wastewater. The color was assayed at wavelength 455 nm on a spectrophotometer. COD, color, turbidity and heavy metals concentration were measured according to standard methods [24]. The amount of adsorbed COD was calculated as the difference between the concentration of COD initial and the final concentration in the equilibrium solution.

The amount of adsorption at equilibrium was calculated using the following equation q_e (mg g^{-1}):

$$q_e = (C_0 - C_e) \times \frac{V}{M} \quad (1)$$

where C_0 and C_e (mg L^{-1}) are the concentrations of COD at initial and equilibrium and, respectively, V is the volume of solution (mL) and M is the adsorbent mass (g).

2.3. Adsorption isotherm models

Langmuir and Freundlich's models are used to study the COD adsorption isotherm for CKD and WTR samples as follows:

The Langmuir model has the form:

$$q_e = \frac{q_{\max} K_L C_e}{(1 + K_L C_e)} \quad (2)$$

The Freundlich model has the form:

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

where q_e is the amount of the adsorbed COD (mg g^{-1}), C_e is COD concentration (mg L^{-1}) at the equilibrium in solution, K_L (L mg^{-1}) and K_F (mL g^{-1}) are the constants of Langmuir and Freundlich, respectively. $q_{\max}$ (mg g^{-1}) is the maximum adsorption capacity of CKD or WTR as adsorbents and $1/n$ is the adsorption intensity.

3. Results and discussion

3.1. Effect of equilibrating time on adsorption of COD on CKD and WTR

Fig. 1 shows that the amount of adsorption of COD from textile wastewater (raw, COD = 760 mg L^{-1} average of six samples) on CKD and WTR as a function of time at pH = 7.0. The amount of COD adsorption increased with the addition of CKD and WTR, but the highest adsorption was obtained at 2 h. This increase in adsorption with the adsorbent dose can be attributed to the increase in the adsorbent surface and the availability of more adsorption sites [25]. At 500 mg L^{-1}

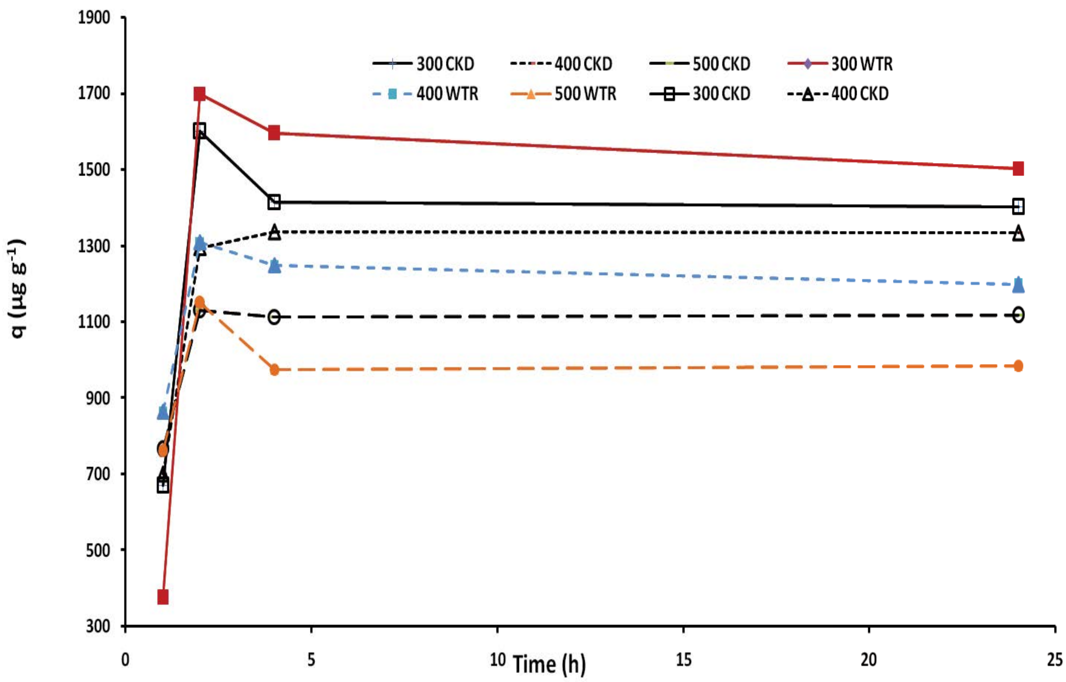

Fig. 1. COD adsorption on CKD and WTR with different rates as affected by equilibrating time.

concentrations, the COD removal of CKD was 49.1%, 59.2%, 58.9% and 58.6%, while, the COD removal of WTR was 50.0%, 60.5%, 59.2% and 58.9% to 1, 2, 4 and 24 h of equilibrating time, respectively (Table 1). The high removal of COD was 59.23% and 60.5% with the addition of 500 mg L^{-1} of CKD and WTR at 2 h, respectively, while the reduction in removal with increased equilibrating time was due to the release of contaminants that could be absorbed from wastewater. It is seen that the equilibrium was achieved within 2 h. The reduction of COD on CKD was reported by Mahmoud [26], who found COD removal above 69% from municipal wastewater. This removal can be accounted for adsorption or adsorption/precipitation mechanism. The COD adsorption mechanism is a mixture of physical, chemical and electrostatic interactions between CKD and organic compounds. Adsorption of organic matter by precipitation is the most dominant mechanism at a higher pH [27]. Similar effects have been observed by Babatunde et al. [28] 72% removal of COD with WTR as an absorbent medium was found in the laboratory scale wetlands. Mountassir et al. [29] found that the use of clay particles in the synthetic textile effluent treatment increased the removal efficiency of color and COD as like WTR used in the study.

3.2. Adsorption isotherm of COD

The correlation coefficient in Langmuir isotherm was higher than Freundlich isotherm by which we can conclude that the Langmuir isotherm model was more appropriate to describe COD adsorption on WTR, but the COD adsorption on CKD was successfully fitted to both Langmuir and Freundlich isotherm models ($R^2 > 0.90$) (Table 2).

Table 1
Effect of equilibrating time on COD concentrations (ppm) by CKD and WTR

Treatments	Conc. (mg L^{-1})	1 h	2 h	4 h	24 h
	300	559	420	412	410
CKD	400	480	355	349	340
	500	387	310	312	314
	300	647	458	462	478
WTR	400	414	362	350	347
	500	380	300	310	312

CKD: cement kiln dust;
WTR: water treatment residual.

The maximum adsorption of Langmuir was higher with the application of WTR than CKD. In the COD adsorption, the values of K_F are 7.96 and 14.16 mL g^{-1} for CKD and WTR, respectively. Water treatment residues have a high adsorption capacity due to their large surface area and chemical composition [30]. The maximum adsorption capacity of the WTR for COD (100.0 mg g^{-1}) was observed to be higher than the adsorption of COD onto peanut activated carbon (52.9 mg g^{-1}) [31], activated carbon (38.96 mg g^{-1}) and wheat straw (55 mg g^{-1}) [32]. In this study, q_{max} of the CKD for COD is 14.3 mg g^{-1}. Magdy et al. [33] found that the q_{max} value of CKD was 100 and 125 mg g^{-1} for *o*-cresol and *o*-nitrophenol (phenolic compounds), respectively. The difference with our study was due to CKD dose and type of raw sewage samples. The adsorption intensity of WTR was higher than CKD, and this indicates that WTR was higher than CKD for COD adsorption in wastewater. The adsorption intensity of COD adsorption on WTR was greater than 1. This indicates that COD adsorption by WTR is a physical process, while the adsorption intensity for COD adsorption on CKD was less than 1, indicating that the adsorption of COD on CKD is a chemical process [34]. X-ray diffraction (XRD) analysis of CKD with the dye has been shown to decrease in the peak intensity of the CH stage, indicating that the removal process is mainly accompanied by a chemical reaction between the dye molecules and the dissolved portion of the CKD [35].

3.3. Effect of CKD and WTR on heavy metals removal

Table 3 shows the effect of CKD and WTR on heavy metals adsorption such as zinc (Zn), manganese (Mn), cadmium (Cd) and lead (Pb) from textile wastewater at equilibrium compared to water criteria for irrigation. Heavy metals concentration in the treated wastewater decreased with the addition of CKD or WTR. Heavy metals in treated wastewater of textiles had lower than the Egyptian guideline (48/1982) [36] and the United States Environmental Protection Agency [37] for irrigation. Similar results with Zaki et al. [38] found that the efficiency of removing Cu, Cd, Ni and Zn by CKD was about 100%. CKD can be used as an immobilizing material to reduce heavy metal levels in contaminated soils as well as wastewater and sludge [23]. The adsorption of heavy metals on CKD may be attributed to the adsorption of heavy metals on $CaCO_3$ existing in CKD. In the formation of surface metal-complexes, these complexes may be formed due to the interaction of the metal with surface sites of oxides such as Fe–OH, Al–OH

Table 2
Chemical oxygen demand (COD) adsorption isotherm model parameters described by Langmuir and Freundlich equations in the textile wastewater

Treatments	Langmuir				Freundlich			
	q_{max} (mg g^{-1})	K_L (L mg^{-1})	R^2	SE	K_F (mL g^{-1})	$1/n$	R^2	SE
CKD	14.286	0.4356	0.9452	0.000043	7.9615	0.9362	0.9455	0.00496
WTR	100.0	6.0533	0.7653	0.000097	14.1619	1.0728	0.7422	0.00571

q_{max} the maximum adsorption capacity; K_L the Langmuir constant related to adsorption energy; $1/n$ the adsorption intensity; K_F the Freundlich constant indicative of adsorption affinity; R^2 the determination coefficients.

and Si–OH that are found in CKD and may also be reduced by precipitation with high pH [22]. Larous and Meniai [39] it was found that the amount of copper removed from aqueous solutions increased with increasing pH.

Irregular surfaces and edges of WTRs, which provide a highly reactive surface to adsorb for heavy metals [40]. Adsorption of the metal cations (Pb, Cd, Co, and Ni), on WTRs depends on pH, with an increasing pH of the solution the removal efficiency was higher [41]. Fourier-transform infrared spectroscopy analysis of WTR showed that the functional groups such as OH^- or COO^- on the surface can the formation of inner or outer-sphere complexes with heavy metals. The adsorption mechanism of metals on WTRs was chemisorption, which involved an exchange between metal anions/cations, surface ligands and covalent formation with the surface [42]. The feasible mechanism of Cd ion adsorption on WTR is the ion exchange model [43]. Wołowiec et al. [30] stated that the removal of heavy metals by WTR could be due to the presence of different oxides in their components and because of their large surface area.

3.4. Effect of alum and WTR on color, turbidity and COD removal

The COD, color and turbidity removals increased with increasing WTR rates in the textile wastewater (Table 4). XRD analysis of Al-WTR showed that it contains quartz, calcite, feldspar, smectite/illite, and kaolinite and these components act as adsorbents for color, COD and heavy metals [44]. The results reveal that the WTR at dose 500 mg L^{-1} could remove about 61.3% of COD, 13.5% of color and 10.8% of turbidity as average (Table 4). While, COD, color and turbidity removals were 82.2%, 85.6% and 79.4% for treated wastewater by 500 mg L^{-1} WTR + 200 mg L^{-1} Alum, respectively. In this study, the removal efficiency of color, COD and turbidity increased, when the alum was applied. Similarly, the combined application of alum and lime enhanced the removal efficiency of COD and color in textile wastewater as compared to alum alone [45]. Another study also revealed that the removal of turbidity, COD, and color were 99.5%, 99.1%, and 99.5%, respectively, of the combination of alum and activated carbon used to treat pulp and paper mill wastewater [46], which were approximately equal to the result obtained in this experiment conducted on textile wastewater. From this study, water treatment residues can be used as activated carbon to remove color, COD and turbidity.

4. Conclusions

The results demonstrated that the adsorption of COD and heavy metals increased with increasing CKD and WTR addition. The removal efficiency of turbidity, color and COD increased with WTR and alum addition. The maximum adsorption of Langmuir was higher with the addition of WTR (100.0 mg g^{-1}) than CKD (14.3 mg g^{-1}). The maximum adsorption of Langmuir was higher with the addition of WTR than CKD. The values of K_F are 7.96 and 14.16 mL g^{-1} for CKD and WTR, respectively. Increases in the COD and heavy metals adsorption in the textile wastewater treated by the WTR or CKD are important for reduction of pollution in the drainage drains. The results confirmed that both CKD

Table 3
Effect of CKD and WTR on heavy metals concentration (ppm) removal from textile wastewater for average five samples

Treatments	Conc. (mg L^{-1})	Zn	Mn	Cd	Pb
	300	0.05	0.12	0.01	0.07
CKD	400	0.03	0.18	0.01	0.05
	500	0.01	0.15	0.01	0.01
	300	0.04	0.026	0.03	0.06
WTR	400	0.03	0.24	0.01	0.03
	500	0.01	0.20	0.01	0.01
FAO (1985)		2.0	0.2	0.01	5.0
Egypt (48/1982) [36]		5.0	1.0	0.05	0.5

Table 4
Effect of alum and WTR on color, turbidity and COD removal from textile wastewater

Treatments	Average (n = 5 samples)					
	Color		Turbidity		COD	
	PCU	Removal, %	NTU	Removal, %	mg L^{-1}	Removal, %
Raw	550		59		2,160	
300 mg L^{-1} WTR	519.57	5.5	53.1	10.0	1,002.2	53.6
400 mg L^{-1} WTR	498.30	9.4	52.8	10.5	980.6	54.6
500 mg L^{-1} WTR	475.75	13.5	52.6	10.8	842.0	61.3
300 mg L^{-1} WTR + 200 mg L^{-1} alum	282.15	48.7	31.4	46.8	654.5	69.7
400 mg L^{-1} WTR + 200 mg L^{-1} alum	245.85	55.3	28.7	51.3	527.0	75.6
500 mg L^{-1} WTR + 200 mg L^{-1} alum	79.20	85.6	12.2	79.4	384.5	82.2

and WTR are excellent feasibility to remove organic and toxic metal contaminants. However, there is still a need to study, develop, apply and reuse waste materials to absorb various environmental pollutants, which increases pollution control.

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