### UTILIZATION OF CEMENT KILN DUST IN DOMESTIC SEWAGE SLUDGE TREATMENT FOR SAFE DISPOSAL AND ENVIRONMENTAL PROTECTION

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### ABSTRACT

Recent years, wastewater treatment plants are producing huge amounts of primary and secondary sewage sludge. On the other hand, large quantities of cement kiln dust (CKD) were produced from cement factories with negative impacts on the environment. CKD is a considerable by-product waste material of the industrial process. At the time being, remarkable advances have been achieved in the management and reuse of CKD. Due to its high alkalinity, huge amounts, negative impact and abundant source; it is considered as noncontrolled source of pollution. Reuse of CKD in presence of waste sludge has high potential as an additional low cost raw material, fertilizer, constructing material, adsorbent and improving sandy soil properties. In this study, CKD was used as chemical coagulant / flocculent sewage sludge and adsorbent for soluble pollutants as well as microbial disinfectants. The optimum operating conditions were adjusted using Jar test experiment and tested sludge was analyzed. Results showed that on addition 8.0 g/l CKD to primary sludge and 7.0 g/l to return activated sludge (RAS) achieved good efficiency for solids separation, BOD5, COD, PO4, and NH4; respectively. Under the effect of high pH value, it was also beneficial in heavy metals immobilization and minimization of metal solubility in the treated sludge matrix. Consequently, metal ions were precipitated as hydroxides. Moreover, all pathogenic and viral species were destructed including total and fecal coliforms.

**Keywords**: Sewage sludge – Cement Kiln dust – Reuse – Adsorption - pH.

### **INTRODUCTION**

The constituents removed in wastewater treatment plants include screenings, grit, scum, solids, and bio-solids. Solids were formed as a result of wastewater treatment plant activities. They are usually in the form of liquid or paste phases. Bio-solid is typically contains from 0.25 to 12% solids by weight depending on the used treatment process. The term bio-solids, as defined by the Water Environment Federation (WEF), reflected the fact that solids content are organic in nature that can be used beneficially after treatment using stabilization and composting. The term "Sludge" is generally used in conjunction with a process descriptor, such as primary sludge and waste-activated sludge (George et al., 2003). According to Yurtsever, 2005; the quantity and characteristics of sludge generated in a wastewater treatment plant was dependent on the physic-chemical composition of the wastewater, the type of wastewater treatment used, and the type of subsequent treatment applied to the sludge. The characteristics of the sludge produced can be changed annually, seasonally, or even daily because of continuous variations in the incoming wastewater composition as well as in treatment processes. The composition of the wastewater reflected on all the substances that were used in the community, due to wide variations in the wastewater contents. Cleverson et al., 2007 showed that the evaluation of alternatives for sewage sludge treatment and final disposal was usually complex, due to the interaction of technical, economic, environmental and legal aspects. Although

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complex and expensive, final sludge disposal was often neglected in the conception and design of wastewater systems, operators sometimes need to handle the final disposal of the sludge on an emergency basis, with all the burden of high costs, operational difficulties and undesirable environmental impacts that might undermine the benefits of the wastewater treatment system. Along Egypt about 3938  $m^3/d$  of primary sludge were discharging from three primary wastewater treatment plants with solid content of 4%, and near 619253  $m^3/d$  of secondary sludge were produced from 270 secondary wastewater treatment plants with total solid percentage of 2%, (Holding Company for Water and Wastewater-Technical Report, 2013). On the other hand, El-Mahllawy, 2013 reported that in Egypt, every year a huge amount of CKD was produced from cement industry during the manufacture of cement clinker via the dry process. The annual production of cement reached 30 million tons, with at least 3 million tons of CKD. It is generally gravish in color and consists predominately of silt-sized, non-plastic particles representing a mixture of partially calcite and un-reacted raw feed. Clinker dust and fuel ash enriched with alkali sulfates, halides and other volatiles. The environmental problems arise when CKD transported to be dumped as land filling that cause remarkable pollution to all surrounding environment. Reuse options were limited and the bulk of CKD was not reused in the cement manufacturing process but sent to landfills or stored on-site (Mackie et al., 2010). X-ray diffraction analysis of cement kiln dust showed that the main constituent is calcite (CaCO<sub>3</sub>), quartz (SiO<sub>2</sub>) and calcium sulfate (CaSO<sub>4</sub>). CKD has potential as an amendment for the neutralization of acidity with high adsorption capability for aqueous metals and nutrients removal

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efficiencies (El-Awady, 1998b; Salem *et al.*, 2015). Adsorption is the most effective and widely used technique for the removal of toxic heavy metals from wastewater as a reliable approach for sewage treatment in developing countries (El-Awady, 1998a; Selvi *et al.*, 2001; El-Awady and Ali, 2012). Due to the high cost and difficult procurement of activated carbon as an adsorbent material, efforts were directed to find efficient and low-cost adsorbent materials (Mahmoud, 2010 and 2014). The high adsorption ability of cement kiln dust, and its availability in high quantities, as an inexpensive by-product of cement industry, it could be effective in the removal of all heavy metals with special efficiency for chromium from tannery wastewater (El-Awady and Sami, 1997; Al-Meshragi *et al.*, 2008). The present study aims to study the role of cement kiln dust in the treatment of primary and return activated sludge as well as the possibility of reuse the treated sludge for agricultural purposes.

### MATERIALS AND METHODS

### **1- Materials:**

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### 1.1.Sewage sludge samples:

Two types of sewage sludge were collected from; (i) Abu-Rawash wastewater treatment plant (ARWWTP). It is operating as primary treatment plant located in Giza, Egypt. Its production rate reached 5000 m<sup>3</sup>/day liquid primary sludge with total solids percentage of 4%, where around 90 liters of sample was collected. (ii) Zeinin wastewater treatment plant (ZWWTP) is operating as secondary treatment plant located in Giza, Egypt. It produces

more than 20,000  $\text{m}^3$  /day liquid RAS with solids percentage of 0.5 %, where about 75 L of sample was collected.

### 1.2.Cement kiln dust (CKD):

CKD was collected from Helwan Cement Company, located in Helwan District, Cairo, Egypt. It was dried for 24 h at 105°C. The characterization of

### **2- Methods:**

### 2.1. Jar test procedures:

The Jar test procedure was conducted using a standard apparatus (Velp scientific, Europe, c26- six paddle stirrer, with lamps to light during mix an alarm for timing) was used in all experiments. Paddles attached to shafts made from stainless steel. The stirrers feature variable speeds from 0 to 300 rpm, with digital read out panel. Jar test has been used to determine the optimum conditions (CKD dose; RPM; flocculation time and settling time) for CKD addition to primary and return activated sludge samples. After adjusting optimal operating conditions, complete analyses were carried out for raw and treated samples.

### 2.2. Physico-chemical analyses of raw and treated samples

The physical and chemical parameters have been carried out according to APHA (Standard Methods for the Examination of Water and Wastewater, 22<sup>nd</sup> Edition, Eugene *et al.*, 2012). Metals concentrations were measured using Inductively Coupled Plasma (ICP), Optical emission Spectrometer (7300DV, Perkin Elmer). pH's were measured using pH-Meter (Thermo scientific, USA). Total solid (TS) was the term applied to the material residue left in the vessel after evaporation of a sample and its subsequent drying in an oven at a defined temperature. Total suspended solids (TSS) is the portion of

total solids retained by a filter, and Total dissolved solids (TDS) is the portion that passes through a filter of 2.0m (or smaller) nominal pore size under specified conditions. Sludge volume index (SVI) is the volume in milliliters occupied by 1 g of a suspension after 30 min settling. Chemical Oxygen Demand (COD) was measured using Block heater of model Thermo Scientific, USA and colorimeter (Lamotte, USA). Biological oxygen demand (BOD<sub>5</sub>) measured using incubator (WTW, Germany), DO meter (Thermo Scientific, USA) and bottles 300-mL (Wheaton, USA). Ammonia (NH<sub>3</sub>) measured using Ion Selective electrode (thermo-orion star). Phosphate (PO<sub>4</sub><sup>-3</sup>) digested on a hot plate using potassium persulphate and sulfuric acid then determined using double beam spectrophotometer (Lambda 25, Perkin Elmer, USA). Turbidity measured using colorimeter (lamotte, USA). Finally, sulphide (S<sup>-2</sup>) was measured according to iodometric titration method.

### 2.3. Determination of total and fecal Coliform:

The total coliform (TC) and fecal Coliform (FC) were performed according to APHA, 22<sup>nd</sup> Edition, 2012. The used media were M-Endo agar and M-FC agar. For detecting TC, the collected samples were serially diluted using sterile saline water 10 fold, 1 ml of each dilution was seeded by membrane filter technique on the testing culture medium (M-Endo Medium).The inoculated plates were incubated for 24 hours at 35°C. The typical coliform colonies on M-Endo medium were counted. For detecting FC, the collected samples were serially diluted using sterile saline water 10 fold; 1 ml of each dilution was seeded by membrane filter technique on the testing sterile saline water 10 fold; 24 hours at 35°C. The typical coliform colonies on M-Endo medium were counted. For detecting FC, the collected samples were serially diluted using sterile saline water 10 fold; 1 ml of each dilution was seeded by membrane filter technique on the testing culture medium (M-FC Medium). The inoculated plates were incubated for 24

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hours at 44.5°C, where the typical FC colonies on m-Endo medium were counted.

### **RESULTS AND DISCUSSION**

### 1. Characterization of cement kiln dust:

Table (1): Chemical composition of CKD

Constituent	Weight (%)	Constituent	Weight (%)
SiO <sub>2</sub>	9.85	SO <sub>3</sub> <sup></sup>	7.45
$Al_2O_3$	2.27	Na <sub>2</sub> O	1.74
Fe <sub>2</sub> O <sub>3</sub>	1.98	K <sub>2</sub> O	4.32
CaO	33.02	Insoluble Residue	
MgO	2.25	Loss of Ignition	10.85

The chemical composition of CKD in Table (1) shows that it has multiple coagulants (CaO, MgO,  $Al_2O_3$  and  $Fe_2O_3$ ; respectively), in addition to be an adsorbent at the same time due to presence of SiO<sub>2</sub> (Rahman *et al.*, 2011).

# 2. Evaluation of physical-chemical and microbiological quality of treated sludge

### 2.1. Jar test procedures

The jar test procedures were conducted to detect the optimum conditions for CKD addition to primary and return activated sludge to obtain the best quality of the treated supernatant under investigation. Figure (1) shows the optimum dose of CKD for: (a) the primary sludge and (b) return activated sludge

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Figure (1): a.Optimum CKD dose for primary sludge treatment



Figure (1): b. Optimum CKD dose for return activated sludge (RAS) treatment

Figures (1.a - 1.b) show that the NTU decreases with increasing CKD dose, then increased again on extra addition. The optimum conditions for CKD addition to primary sludge found to be 8 g/L at 1 min flash mixing, 2 8 Vol.33, No.2, June, 2016

min flocculation, 15 min settling time, 15 RPM, at final pH value 12.6. On the other hand, the optimum conditions for CKD addition to RAS found to be: 7 g/L at 2 min flash mixing, 10 min flocculation, 30 min settling time, 10 RPM, at final pH value 12.1. Results showed an increase of pH up to 12.1and 12.64 at the CKD optimal doses; whereas Usama and Rafik (2013) observed that when 1.2 g/l CKD added to raw samples, the pH increased to be 8.22. Moreover, El-Awady and Samy, 1997 found that the chemical analysis of CKD showed that sodium and potassium oxides have dominant effect on the increase of the pH of the aqueous CKD soaked solution.

### 2.2. Physico-Chemical and Microbiological Analyses:

They were carried out on raw primary and RAS samples, in addition to their treated supernatants. The optimum operating conditions have been determined. The jar test was used and the produced samples were analyzed to determine the effect of CKD addition to primary and RAS, respectively. Three Jar test runs were conducted for three different composite sludge samples. The averages of analyses were calculated to detect the removal efficiencies. Table (2) shows the physico-chemical analyses for primary sludge and its treated supernatants, respectively.

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Parameter	Unit	primary sludge*	Treated samples with CKD	R%
PH		6.83	12.64	
TSS	mg/l	5237	384.7	92
TS	mg/l	6572	4633.3	29.5
SV	ml/l	233	178.5	23.4
SVI		38.1	29.1	23.6
COD	mg/l	10397	1209	88.4
BOD <sub>5</sub>	mg/l	2063	362	82.4
NH <sub>3</sub>	mg/l	28.8	3.6	87.6
$PO_4^{-3}$	mg/l	23.3	9.4	<b>59.8</b>
$S^{-2}$	mg/l	24.7	3.9	84.3
Cu	mg/l	0.42	0.1	77.2
Fe	mg/l	30	2	93.2
Pb	mg/l	0.13	0.02	88.4
Mn	mg/l	0.5	0.13	74.5
Ni	mg/l	0.1	0.01	80
Zn	mg/l	1.5	0.2	88.5

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Table (2): Physico-chemical analyses of primary sludge and treated

\*The average of three samples

supernatant

The TSS removal was 92% which is comparable results to that stated by El-Awady and Samy, 1997, where the percent removal was characterized as 92.8 %. The high removal efficiency of TSS proved that CKD is relevant as a coagulant due to presence of CaO, MgO,  $Al_2O_3$  and  $Fe_2O$  in its composition, in addition to its good adsorbing capability (Rahman *et al.*, 2011). The removal of TS was 29.5% and this is due to the high ability of increase the TDS concentration after addition of CKD dose. The decrease in Sludge volume (SV) by 23.3% and the decrease of Sludge volume Index (SVI) by 23.6% were

matched with El-Awady, 1998b. On the other side, Allison et al., 2010 used CKD and quicklime slurries to treat mine wastewater who indicated that the lower sludge volumes could be generated through the use of CKD slurry if compared to traditional quicklime treatment. Organic load like COD and BOD<sub>5</sub> were the basic and important parameters, so the removal of COD was up to 88.4 % which is close to that concluded by El-Awady and Samy, 1997. They also declared that the use of CKD with 20 g/L as an adsorbent has the capability to increase pH-value from 3.64 to 8.2; with a removal efficiency of COD reached to 55.6 %, while the removal of TOC exceeded 88.8%. The BOD<sub>5</sub> removal was found to be 82.4% that matched with El-Awady (1998) where he reported that the effect of CKD as an efficient alternative coagulant in sewage treatment. He carried out a comparison in chemical coagulation between ferric chloride, lime and CKD and found that the removal efficiency of BOD<sub>5</sub> using CKD was 77% from the initial value. The results obtained from this study showed that NH<sub>3</sub> removal efficiency of 87.6%, where EL-Awady (1998) found that ammonia removal was 66.5%. Phosphate removal efficiency found to be 59.8% which matched with El-Awady (1998). The sulphide content decreased with 84.3% which gave an indicator about the good quality treatment and this matching with Rahman, 2011 who found that CKD make good chemical elimination for hydrogen sulphide. Removal of metals like Cu, Fe, Pb, Mn, Ni, and Zn were 77.16, 93.23, 88.39, 74.50, 80, and 88.47 %, respectively. Results show that the final concentration of metals is within the limits. El-Awady and Samy (1997) reported the mechanism of heavy metals removal from acidic wastewater may be explained as follows:

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### (1) Heavy metals hydrolysis:

 $X_2O + H_2O \rightarrow 2X$  (OH) ; X (OH) + MSO<sub>4</sub>→ M (OH)<sub>n</sub>↓ + XSO<sub>4</sub> Where X: K, Na, M: Cr, Fe, Cu, Co and n: 2, 3.

### (2) Adsorption of heavy metals on the CKD fine particles :

 $M (OH)_n + CKD \rightarrow CKD - (-M (OH)_n)$ 

It was reported that the removal of Mn, Cu, Pb, Cd, Zn, and Cr were 100% at 20g CKD/ L and 150 rpm for 30 minutes, while the removal of iron and nickel were 98% and 80 %, respectively. Physico-Chemical analyses for RAS and its treated supernatant samples are graphically represented in Figure (2).





Figure (2) shows TSS removal efficiency up to 94.3% which refers to the characteristics of CKD as a good coagulant and flocculent. The R% of SV and SVI was 39.4 and 37.4%, respectively. Allison and Margaret, 2012, compared between CKD and quicklime slurries in the treatment of mine 12 Vol.33, No.2, June, 2016

water and they found that sludge volumes were generated in CKD-treated samples were only 50-60 % of those generated with the quicklime slurry. The removal efficiencies of BOD<sub>5</sub>, COD, and PO<sub>4</sub><sup>-3</sup> were 99.2, 99.5 and 93.9 % respectively. These results is matching with Mostafa (2012) who enhanced the removal efficiencies of BOD<sub>5</sub>, COD, TSS, phosphate and pathogenic bacteria by adding CKD to wastewater as a chemical coagulant in a designed module at temperature ranges of (30-35) °C and (15-20) °C to be complied with the Law 501/2005 for reuse limits for agricultural purposes. Mostafa (2012) stated that phosphate removal by chemical precipitation is the best known process and is widely used despite its relatively high costs. When 2 g CKD/L was added for wastewater in his designed model, the average removal efficiencies for BOD<sub>5</sub>, COD, TSS and phosphate were found to be 94.1%, 94.1%, 94.2% and 87.9%, respectively. Also, CKD addition decreased the bulking problems occurred at temperature range (15–20) °C, where SVI was decreased from 149 to 63 and as a result, bulking did not occur. The ammonia removal was 61.6%. Mostafa, 2012 showed that CKD contain high ratio free of CaO which act as a good coagulant and flocculent also have cations as potassium and manganese which define cell membrane permeability and leads to high phosphate removal. Removals of heavy metals (Cu, Fe, Pb, Mn, Ni, and Zn) from treated RAS with CKD were consequently 77.1, 93.23, 88.39, 74.5, 80 and 88.47 %. Taha et al. (2007) studied the adsorption of Cd (II), Al (III), Co (II) and Zn (II) using CKD and synthetic stock solution of heavy metals and found that the removal of zinc was about 80% at pH 6.5 and it increased to 99% at pH 8. For aluminum, 85% was removed at pH 5 and it increased to 99% at pH 6. For cadmium, 90% was removed at pH 5.5 and it

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increased to 99% at pH6.2.For Cobalt, removal increased proportionally with increasing pH from 50% at pH 6 to 90% at pH 8. The obtained results of this work are in agreement with Salem *et al.*, (2015) who found that mixing of sewage water with CKD (10 g/l) completely removed manganese, nickel, lead, zinc and iron (100% removal) after 3 days of treatment. The removal of cadmium and cobalt were proportionally increased with increasing pH up to 8. Above pH 8, the adsorption ability was decreased due to increasing in OH–ions.

Table (3) shows the microbiological analyses for the primary, return activated sludge and their treated supernatants, an average of three results and the efficiency of removal.

Parameter	Parameter primary sludge			Return activated sludge		
	Raw	Treated	<b>R%</b>	Raw	treated	<b>R%</b>
ТС	6.3×10 <sup>5</sup>	0	100	$2.4 \times 10^{7}$	5000	99.98
FC	$1.8 \times 10^{6}$	0	100	13.3×10 <sup>5</sup>	1000	99.92

Table (3): Microbiological analyses of primary and treated sludge with CKD

The obtained results showed high removal efficiency of total coliform and fecal coliform with 99.9% and 100%, respectively which has remarkable effectand it is due to the strong effect of the high pH value. Results matched with El-Awady 1998 a, b; El-Awady and Ali 2012 who carried out bacteriological examination on composite samples of primary and RAS for TC and FC with removal efficiencies 100%.

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### Physico-chemical and microbiological analyses for the dried raw primary and RAS and dried treated sludge with CKD

The precipitated sludge at the end of each test procedures has been collected and dried at 105°C. Physico-chemical analyses were shown in Figure (3)



Figure (3): Efficiency of removal for dried raw and treated sludge

Sludge weight of primary sludge increases to 124 % after treatment which matched with Salah and Sabah (2012) who observed that sewage sludge treatment by addition of CKD resulted in an increase in solid content of the sludge. Also the results matched with Allison *et al* (2010) who use CKD-generated slurry for the treatment of mine wastewater, resulted in efficient precipitation of metals in the coagulation stage. Specifically, over 99 % of soluble iron and zinc in the raw mine wastewater were transformed into insoluble compounds (precipitates).

The microbiological analyses for dried sludge and the treated one were shown in table (4)

 Table (4): Efficiency of removal for total coliform and fecal coliform

Parameter	<b>R%</b> (primary sludge)	<b>R%</b> (RAS)
TC	99.9	99.7
FC	99.9	99.2

The results show high removal efficiency of FC and TC up to 99% after applying the optimum conditions and this is due to the high pH. Results matched with El-Awady and Ali (2012) who found that CKD addition to sewage sludge destructed all types of viruses, parasites, pathogenic bacteria, and all types of other harmful microorganisms.

### SUMMARY

### From the previous results, the following can be concluded:

- The obtained results of the treatment of primary sludge and return activated sludge using CKD as a coagulant at their optimum operating conditions, with the high pH value which control the release of heavy metals via changing the cationic ions into a hydroxide forms.
- The silicate as well as calcium compounds contained in CKD behaved as an adsorbent material. Moreover, the high pH value destructed all types of pathogenic viruses. As a result of these effects, the CKD behaved as an adsorbent material, in addition to help suspended materials to be compacted and reduce the fluffy volume of sludge to the minimum. The solid compaction helps the water part to be separated from sludge matrix.

The produced water can easily recycled and the solids be dried in few days.

- Addition of 8 g/l CKD to the primary sludge and 7 g/l to the return activated sludge achieved an excellent removal of different organic and inorganic analytes.
- The chemical treatment of sewage sludge using CKD as coagulant and flocculants is preferred because of the high removal of solids, BOD5, COD, Nitrogen, sulphide and phosphates.

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## استخداء ترابع أفران الأسمنت في معالجة حمأة الحرف الحدي بمدف التخلص الآمن وحماية البيئة

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### المستخلص

يهدف هذا البحث إلى دراسة تاثير إضافة تراب أفران الأسمنت إلى حمأة الصرف الصحى بهدف التخلص الآمن وأيضا إمكانية استخدامها فى الأغراض المختلفة. تنتج مصانع الأسمنت كميات هائلة من تراب أفران الأسمنت فى مصر سنويا لا نقل عن ٣ مليون طن ومن المعلوم أن لها تأثيرات سلبية على البيئة. على الجانب الآخر تقوم محطات معالجة الصرف الصحى بانتاج الاف الأمتار المكعبة من الحمأة السائلة الإبتدائية والثانوية يوميا والتى تحتوى على كميات من الأحمال العضوية والمعادن الثقيلة والعديد من الكائنات الممرضة والفيروسات. تم جمع عينات من الأحمال العضوية أبورواش وزنين للصرف الصحى، وتراب أفران الأسمنت من مصنع حلوان. وباستخدام جهاز اختبار الجرعات وجد أن الجرعة المثلى المضافة من تراب أفران الأسمنت هى ٨ جم/ لتر من الحمأة الإبتدائية السائلة و ٧ جم/ لتر من الحمأة الثانوية السائلة، وبعد إجراء القياسات المختلفة على العينات الخام والمعادين الأمونيا والفوسفات المضافة من تراب أفران الأسمنت هى ٨ جم/ لتر من الحمأة وإزالة الأمونيا والفوسفات الكلى بالإضافة لازالة المواد العضوية متراب أفران الأسمنت وبعد إجراء القياسات المختلفة على العينات والأكسجين الكيماوى المستهاكي المضافة من تراب أفران الأسمنت هى ٢ جم/ لتر من الحمأة وإزالة الأمونيا والفوسفات الكلى بالإضافة لازالة المواد العضوية متمتلة فى الأكسجين الحيوى الممتص وازالة الأمونيا والفوسفات الكلى بالإضافة لازالة المواد العضوية متمتلة فى الأكسجين الحيوى الممتص وازالة الأمونيا والفوسفات الكلى بالإضافة لازالة المواد العضوية متمتلة فى الأكسجين الحيوى الممتص وازالة الأمونيا والفوسفات الكلى بالإضافة لازالة المواد العضوية متقلة فى الأكسجين الحيوى الممتص وازالة عالية المعادن الثقيلة نتيجة ارتفاع الأس الهيدروجينى حيث تحولت من الصورة الذائبة إلى نسب إزالة عالية المعادن الثقيلة نتيجة ارتفاع الأس الهيدروجينى حيث القران يرفع الأس المواد النائبة إلى نسب إزالة عالية المعادن الثقيلة نتيجة ارتفاع الأس الهيدروجينى حيث تحولت من الصورة الذائبة إلى الصورة غير الذائبة وترسبت فى صورة هيدروكسيدات.

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