

Decontamination of Water from Dissolved Pollutants by Special Ceramic (AKP1 & AKP2)

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Abstract

In this study, contaminated water was purified by ceramic purifiers produced from a mixture of Iraqi raw materials. These raw materials were activated kaolinite, Porcelanite, and Limestone. They were mixed with two different mixing percentages to produce two types of ceramic (AKP1 and AKP2). Ceramic filters were evaluated based on a number of parameters that were: hydraulic conductivity, purified water properties, and adsorption ability. Results indicated that ceramics hydraulic conductivity have values 0.0024, and 0.0032 m/hr, respectively. The filtered water were tested for water quality parameters that the ceramic filters could remove, as an average percentages of removal, 99.9% to 3.26% by AKP1 and 100% to 64.67 % by AKP2. The adsorption ability of the ceramics for solutes of seven heavy metals, Mn, Fe, Pb, Cd, Co, Cu and Zn, were measured. Results showed that most of the heavy metals ions were removed by the both types of filters that the removal varies between 98.18 and 100% of the initial concentration of each metal. The adsorption capacities of each filter to adsorb the heavy metals were computed according to Langmuir model and Freundlich models. The results showed The data were most fitted to Freundlich model.

Key words: Ceramic, Purifiers, Kolinite, Adsorption, Heavy metals.

1. Introduction

Water is essential to survival of humans and all the living beings. Since the recent century, the percentage of contaminated water increased due to industrial and agriculture development.

Water filtration is one of the easier and cheaper methods for purifying contaminated water. Water filtration is a very important subject due to lack of water in some regions that require circulation and reuse of water.

Ceramic is a very important and useful material which used for water filtration (Jassim, 2010). It was used to remove heavy metals from their solutions (Jassim et al., 2014, Jassim, et al., 2017). Dyes were also removed by special kinds of ceramic (Al-Robai, et al., 2018, and Jassim, et al., 2013). Its properties can be controlled by developing technology to meet certain purposes (Zereffa, Bekalo, 2017). The pore size of the ceramic material can be made homogeneous and small within the range of microfiltration (Jassim, 2010), which is excellent to remove most of the suspended solids and bacteria (Nurmiyanto, Prasetya, 2012).

Some of particles smaller than the ceramic pore size could be adsorbed by ceramic. Ceramic can withstand solutions of high temperatures and a wide range of acidity that other microfiltration media can't withstand (Jassim, 2010). Ceramic can be operated with a proper maintenance or stored for a long time without losing its filtration efficiency. Ceramic can be used easily and they have no side effects, they are environmentally friendly (Ajayi, and Lamidi, 2015).

Generally, this research aims to study the hydraulic performance, purifying and adsorption properties of the ceramic purifiers made of available local materials.

2. Materials and methods

The ceramic raw materials that were used in this study were Activated Kaolinite, Porcelanite, and Limestone. General Company of Geological Surveying and Mineralization provided them. **Table 1** shows the chemical analysis of raw materials

Table 1: Chemical analysis of raw materials. (GCGSM, 2008).

Compound	Kaoliniteite	Porcelaniet	Limstone
SiO ₂	54.5	75.3	3.16
Al ₂ O ₃	35.7	1.95	0.12
Fe ₂ O ₃	1.83	0.77	0.22
CaO	0.94	4.6	54.04
TiO ₂	1.2	0.11	0.02
MgO	0.24	4.7	0.67
Na ₂ O	0.24	0.1	0.38
L.O.I	13	8.5	41.74

Kaoliniteite is one of the clay minerals in Iraq. It is found in Iraq at the west desert at Duakhlia location. It composed of Kaoliniteite and Quartz. The structure of the Kaoliniteite is a gibbsite sheet with a single tetrahedral silica (OH)₈Al₄Si₄O₁₀, (GCGSM, 2008). It can be used as adsorbent when it is activated by heating or by using acid, (Bello, Ajibola and Idris, 2011; and Al Bakain, 2014). It is widely used in ceramic industries, (Gajic, et al. 2014).

Porcelanite is a siliceous rock. These rocks found in Iraq in Akashatat the western desert. It is largely composed of sponge spicules and some other siliceous microfossils (Diatoms and Radiolarian). It is composed of the following minerals: Cristobalite, Tridymite, and Calcite,(GCGSM, 2008).

Limestone is a white rock found at Kerbela and the western desert in Iraq. It is mainly composed of Alcaisiet, CaCO₃,(GCGSM, 2008).

Kaolinite was activated by 0.1 M HCl (Edama,2014). Then two different mixes were prepared. The main part of the first mix was kaolinite. While the main one of the second mix was porcelinet.

Semi dried compression method were used to prepare the mixtures with 10%by weight of water content[Al-Robai, et al. 2018]. Some of the prepared wet mixture was placed in the mold and pressed with an electrical hydraulic jack at 250kg/cm².

Ceramic purifiers samples were manufactured in this study as cylindrical disc tablets of a diameter of 50 mm and thickness of 10 mm.

The firing process of the dry discs, previously prepared, was carried out at at 1200 C°. Three ceramic discs were produced for each mix.

3. Physical test

Tests were conducted to find the physical properties of the produced ceramic. The physical properties that may affect the durability and hydraulic conductivity of ceramic are the apparent density, and porosity.

3.1. Density and porosity tests

Apparent density means dry density. It affects the compressive and tensile strength of Ceramic (Nurmiyanto, and Prasetya, 2012). It also gives the indication to the porosity of ceramic. Apparent density of ceramic body depends on firing program, size graduation, applied pressure and water content during shaping process, (Jassim, 2010).

Porosity refers to the percentage of pore space in a material. An open pore is a cavity or channel that communicates with the surface of the particle.

The apparent density, apparent and true porosity of ceramic discs were measured according to(ASTM-C 373 standards, 2006).

3. 2. Hydraulic conductivity test

The hydraulic conductivity of a porous medium defines its ability to transmit water. The hydraulic conductivity is calculated by constant head method by applying the following equation, (Pal, et. al, 2006):

$$K = \frac{Q \times L}{A_c \times t \times h} \tag{1}$$

Where: A_c= cross section area of ceramic sample, m², h = constant head exerted by water level, m, L=length of sample, m, Q= water discharge passed ceramic at time t, m³, and t=time at which water discharge Q passed through ceramic. Constant head method was employed to measureK of the whole ceramic disc production using distilled water. Each test of hydraulic conductivity of the ceramic disc test was carried out with one replication. Computing the average saturated hydraulic conductivity HC_a for each disc. Computing the standard hydraulic conductivity, HC_s,for each disc according to equation, (Nicolaidis, et. al., 2015)

$$HC_s = HC_a \frac{\mu}{\mu_{20}} \tag{2}$$

HC_a=average saturated hydraulic conductivity,m/hr,μ=viscosity of water at any temperature,Pa.s, and μ₂₀ =viscosity of water at 20°C,Pa.s.

3.3. Mineralogical tests

Mineralogical tests of the produced ceramic discs were done by the x-ray reflection device at Baghdad University College of Science Geological Department x-ray laboratory (Mohamed, 2005).

3.4. Adsorption test

Adsorption test was carried for each produced ceramic disc. The examination were performed by passing polluted water through the ceramic disc and measuring the concentration of pollution before and after passing. Make up solutions of Pb(II), Co(II), Fe(II), Mn(II), Cd(II), Zn(II), and Cu(II) with varyingt concentratio1,10, 20, 40, 60, and 80 mg/l of each of these ions . Adsorptiontest was carried out at room temperature.The test was carried out for all filters together at one time with one replication for each test. The residual focus of metal ions in the solution were measured by atomic absorption spectrometer, AAS. The percentage adsorption was calculated as follows, (Foo, and Hameed, 2010):

$$AP = \frac{Co - Ce}{Co} \times 100 \tag{3}$$

where: Ap=percentage of adsorption, %. Co =initial concentration of metal ion in the queous phase, mg/l, Ce =final concentration of metal ion in the aqueous phase, mg/l.

3.4.1. Adsorption capacity

Two of the more common mathematical formulations used in establishing adsorption isotherms are the Freundlich and Langmuir equations.

3.5. Water purification tests

The principal factors that are taken into consideration when determining water quality are, (WHO, 2006): Turbidity, PH, EC, TDS, Ca⁺², Mg⁺², Na⁺, Cl⁻, HCO₃⁻, SO₄⁻², NO₃⁻.

4. Results, and discussion

4.1. Physical properties

Eight experiments were done to determine the physical properties of the produced ceramics. The results are listed in Table 2.

Table 2: Average Physical Properties of Ceramic purifier.

Filter type	ρ _a , apparent density, gm/cm ³	na, apparent porosity	nt,true porosity	wa,% absorbed water
AKP1	1.48	47.14	14.9	31.82
AKP2	1.29	51.47	17	39.95

4.2. Hydraulic conductivity test

Hydraulic conductivity test was carried out on the produced ceramic, including two replications, to evaluate the hydraulic performance of the produced ceramic. The tests were carried following the constant head method which was viewed previously. The results showed that the average hydraulic conductivities for the two mixes were 0.0024m/hr, and 0.0032 m/hr respectively.

These results shows that when Porcelenite was the main component of the ceramic mix the hydraulic conductivity increased by 33.3%.

4. 3. Mineralogical test

Mineralogical test was carried on the produced ceramic purifier according to the procedure explained before. The resulting minerals were found as shown in figures 1, and 2, which showed the following:

Ceramic purifier AKP1 had formed after burning:

Anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), Beta-Quartz (SiO_2), Low Cristobalite (SiO_2), Tridymite (SiO_2), Enstatite (MgSiO_3), and Larnite syn (Ca_2SiO_4). While ceramic purifier AKP2 had formed after burning: Anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), Beta-Quartz (SiO_2), Low Cristobalite (SiO_2), Tridymite (SiO_2), Diopside ($\text{CaMg}(\text{SiO}_3)_2$), and Larnite syn (Ca_2SiO_4). Here also the three phases of silica (SiO_2), Beta Quartz, Tridymite, and Low Cristobalite, in addition to Anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$) and Diopside ($\text{CaMg}(\text{SiO}_3)_2$), and Larnite syn (Ca_2SiO_4), had appeared. It means that Tridymite and Low Cristobalite remained with their original form and didn't transform because they were the phases of silica which formed at high temperature ($1470, 1705\text{ }^\circ\text{C}$ respectively and since the firing temperature did not reach these high temperatures so they remained in the new composition of ceramic body. The Silica in initial composition transferred to Alpha-Quartz when temperature rose until $573\text{ }^\circ\text{C}$ then it started to transform to Beta-Quartz until $870\text{ }^\circ\text{C}$. Then it reversed to Tridymite then Cristobalite. The formation of Tridymite and Cristobalite were speeded up by the addition of limestone and they were occurred at lower temperatures, (Zereffa, Bekalo, 2017). The Al^{+3} in initial Kaolinite appeared at Anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$) which is also called lime feldspar and it works as Flux in the ceramic body, (Rayon, 1978).

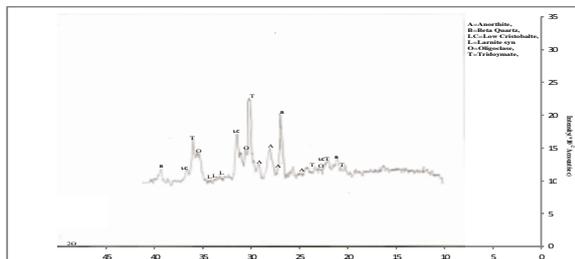


Fig. 1: X-Ray diagram of ceramic type AKP1.

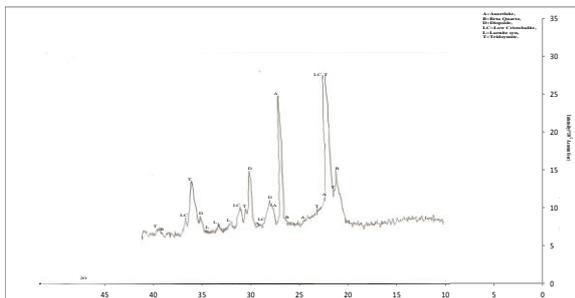


Fig. 2: X-Ray diagram of ceramic type AKP2.

4.4. Heavy metals adsorption tests

Two adsorption tests, including two replication, were carried out to examine the adsorption of heavy metals properties of the produced CP disc. One, including one replication, were carried out on a pre prepared solution of 1 mg/l of each of seven heavy metals, Mn, Fe, Pb, Cd, Co, Cu and Zn. Other one test, including one replication, were carried out on pre prepared solution of 10 mg/l of each of these seven heavy metals.

According to (Iraqi Specifications No 417, 1974), limits of these heavy metals, Mn, Fe, Pb, Cd, Co, Cu and Zn in drinking water are listed in Table 2.

Table 2. Iraqi specifications limits of heavy metals in drinking water.

Heavy metal	Pb	Cd	Zn	Cu	Fe	Mn
Concentration, mg/l	0.05	0.01	1.0	0.5	0.5	0.1

Average results of test that were carried out to examine heavy metals adsorption properties of the produced ceramic discs using a pre prepared solution of 1 mg/l of each of the used seven heavy metals, Mn, Fe, Pb, Cd, Co, Cu and Zn, are shown in figure 3. Generally, the results showed that the produced ceramic adsorbed most of the heavy metals ions. The removal varies between (99.40 - 100%) of the initial concentration. Figure 4 showed the average results of test that were carried out to examine heavy metals adsorption properties of the produced ceramic discs using a pre prepared solution of 10 mg/l of each of the used seven heavy metals, Mn, Fe, Pb, Cd, Co, Cu and Zn. Generally, the results indicated that both filters adsorbed most of the heavy metals ions. The adsorption varies between 98.72 and 100% of the initial concentration of each metal.

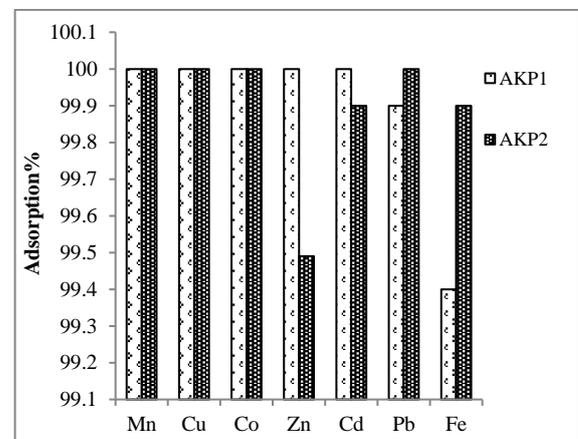


Fig. 3: % of removal of heavy metals at concentration 1 mg/l .

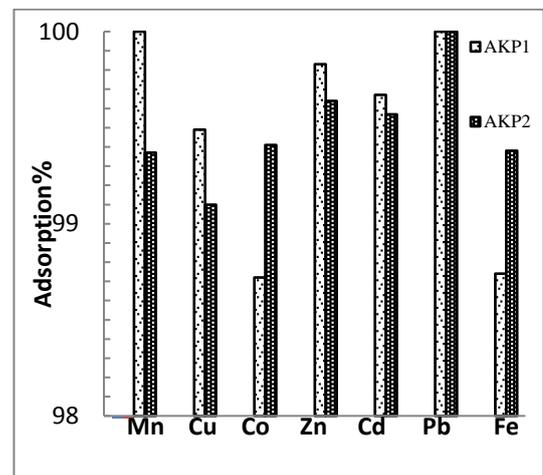
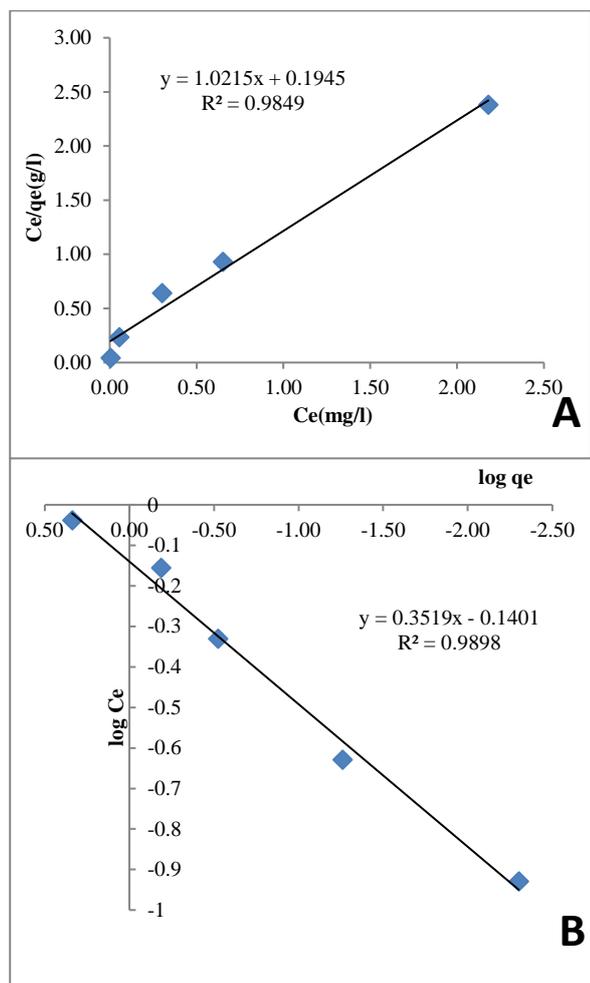


Fig. 4: % of removal of heavy metals at concentration 10 mg/l .

4.4.1. Adsorption Capacity

Adsorption capacities of the produced ceramic, was measured according to the procedure mentioned before. The polluted water with seven heavy metals (Mn, Fe, Pb, Cd, Co, Cu, and Zn), at (20, 40, 60, and 80 mg/l) of each metal. The remaining concentrations ($\text{C}_e\text{ mg/l}$) of these metals after passing through these filters were measured. Adsorption capacity ($q_e\text{ mg}$ of metal adsorbed by one gram of adsorbent) at each concentrate were calculated as mentioned. Then graphs were plotted according to both Langmuir and Freundlich methods. Langmuir and Freundlich graphs were

plotted as shown in figure 5 for Manganese adsorption data of first product. The correlation coefficient R^2 was 0.9849 for Langmuir graph and 0.9898 for Freundlich graph. The same thing was done for all adsorption data for each heavy metal adsorption and for each filter. Adsorption capacity results of first product were shown in Table 3. Results of adsorption capacities according to Freundlich model were: Pb>Cd>Mn>Co>Cu>Zn>Fe, and maximum capacity was for Pb (4.294 mg/g), and minimum was for Fe (1.962 mg/g). While the results according to Langmuir model were: Fe>Zn>Cu>Mn>Co>Pb>Cd, and maximum capacity was for Fe (2.221 mg/g), and minimum was for Cd (0.916 mg/g). For 2.nd mix, adsorption capacity results were shown in Table 4. Results of adsorption capacities according to Freundlich model were: Pb>Mn>Co>Cd<Cu>Zn>Fe. Where maximum capacity was for Pb(4.1771mg/g), and minimum was for Cd(1.8808mg/g). While Langmuir model's results were: Zn>Cu> Fe> Cd> Co> Mn > Pb, and maximum capacity was for Zn (1.1273 mg/g), and minimum was for Pb (0.951 mg/g).



Figs.5: A: Langmuir graph, B: Freundlich graph of data for adsorption capacity of Mn by AKP1

Table 3: Adsorption Capacities of AKP1 for Seven Heavy Metals

metal ion	Adsorption capacity (mg/g) Langmuir model	R^2	Adsorption capacity (mg/g) Freundlich model	R^2
Mn	0.979	0.9849	2.842	0.9898
Cu	1.062	0.9781	2.272	0.9801
Co	0.973	0.9754	2.634	0.9978
Zn	1.066	0.9857	2.101	0.9509
Cd	0.916	0.970	4.093	0.991

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Pb	0.947	0.9847	4.294	0.9852
Fe	2.221	0.6648	1.962	0.9709

Table 4: Adsorption Capacities of AKP2 for Seven Heavy Metals

metal ion	Adsorption capacity (mg/g) Langmuir model	R^2	Adsorption capacity (mg/g) Freundlich model	R^2
Mn	0.9944	0.9717	3.1348	0.9803
Cu	1.1242	0.9484	2.0370	0.9879
Co	1.0381	0.9696	2.5813	0.9784
Zn	1.1273	0.9795	1.9069	0.9914
Cd	1.0654	0.9705	2.1939	0.9923
Pb	0.9510	0.9864	4.1771	0.9941
Fe	1.0714	0.9606	1.8808	0.9779

4.5. Water Purification Tests

Ten water quality tests, including two replication, EC, TDS, Ca^{+2} , Mg^{+2} , Na^{+1} , NO_3^{-1} , SO_4^{-2} , PH, Cl^{-1} , and turbidit. Table 5 showed the instrument used for carrying out each test. The results showed variable ability of purifying of produced filters and this could be caused to their variability in composition of these filters. The concentrations of EC, TDS, Ca^{+2} , Mg^{+2} , Na^{+1} , NO_3^{-1} , SO_4^{-2} , PH, Cl^{-1} , and turbidity of allowable concentrations according to (Iraqi specification No.417, 1974) and concentrations of raw water were as shown in Table 6 .

Table 5: The Instrument Used For Carrying Each Test.

Water quality parameter	Instrument
EC, $\mu m/cm$	Hanna Instrument 3 in one(PH, EC, and TDS), cod No.HI-9811-S
TDS, mg/l	Hanna Instrument 3 in one(PH, EC, and TDS), cod No.HI-9811-S
pH	Hanna Instrument 3 in one(PH, EC, and TDS), cod No.HI-9811-S
Ca^{+2} , mg/l	complexometric titration using EDTA
Mg^{+2} , mg/l	complexometric titration using EDTA
Na^{+1} , mg/l	Flame Photometer Corning-EEL Scientific Instruments
NO_3^{-1} , mg/l	spectrophotometer instrument model VARAN CARY 100conc
SO_4^{-2} , mg/l	spectrophotometer instrument model VARAN CARY 100conc
HCO_3^{-1} , mg/l	complexometric titration with diluted Hydrochloric Acid
Cl^{-1} , mg/l	Argentometric titration method
Turbidity, NTU	Hanna Instrument LP 2000

Figure 6 showed the variable ability of the ceramic products to remove different contaminations. they had removed: 99.99-100 %of turbidity, 79.19-77.52 %of EC, 82.76- 81.03 %of TDS, 80.09- 82.3%of Ca^{+2} , 83.383- 63.75%of Mg^{+2} , 67.18- 59.54%of Na^{+} , 75.23- 71.96%of Cl^{-} , 3.26- 94.37%of NO_3^{-} , 82.56-70%of SO_4^{-} , and 39.75- 64.67%of HCO_3^{-} , respectively. This means they have good to very good purification properties due to forming Tridomyite in their final mineralogy.

Table 6: List of tests carried on raw water, allowable concentrations according to the Iraqi specification and the concentrations in the used raw water.

Water quality parameter	Allowable concentrations according to the Iraqi specification	Concentrations in raw water
EC, $\mu\text{m/cm}$	Not available	2980
TDS, mg/l	1000	1740
Ca^{+2} , mg/l	200	678
Mg^{+2} , mg/l	150	331
Na^{+1} , mg/l	200	131
NO_3^{-1} , mg/l	45	5.06
SO_4^{-2} , mg/l	400	820
HCO_3^{-1} , mg/l	Not available	317
PH	6.5-8.5	7.8
Cl^{-1} , mg/l	200	214
Turbidity, NTU	5	123

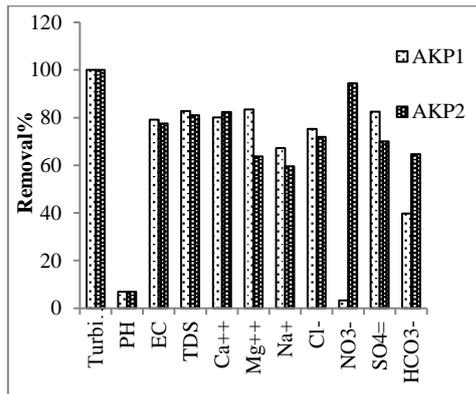


Fig. 6: Percentage of removal of different contaminations by the ceramic products.

5. Conclusion

The following could conclude from this research:

Ceramic water purifiers can be produced by using Iraqi Kaolinite.

The average hydraulic conductivities are 0.0024 m/hr, and 0.0032 m/h r respectively

The ceramic products composites of Tridymite and Low Cristobalite, which have adsorption properties.

The ceramic products could adsorb seven heavy metals, Mn, Fe, Pb, Cd, Co, Cu and Zn, from their solutions at 1.0 mg/l, and 10.0 mg/l concentrations. maximum adsorption capacity of heavy metals for AKP1 according to Freundlich model was for Pb (4.294 mg/g), and minimum was for Fe (1.962 mg/g). And according to Langmuir model maximum capacity was for Fe (2.221 mg/g), and minimum was for Cd (0.916 mg/g). While for AKP2 maximum capacity was for Pb(4.1771mg/g), and minimum was for Cd(1.8808mg/g). While the results according to Langmuir model maximum capacity was for Zn (1.1273 mg/g), and minimum was for Pb (0.951 mg/g).

Purification properties of ceramic products are removing: 99.99-100 %of turbidity, 79.19-77.52 %of EC, 82.76- 81.03 %of TDS, 80.09- 82.3%of Ca⁺, 83.383- 63.75%of Mg⁺, 67.18- 59.54%of Na⁺, 75.23- 71.96%of Cl⁻, 3.26- 94.37%of NO₃⁻, 82.56-70%of SO₄⁼, and 39.75- 64.67%of HCO₃⁻, respectively.

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