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Removal of total organic carbon from surface waters by $Ag_2O/CuFe_2O_4$-loaded silica media filters

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ABSTRACT

This pilot-scale research was conducted at Ahvaz water treatment plant to investigate the removal of total organic carbon from surface waters by $Ag_2O/CuFe_2O_4$-loaded silica media filters. $Ag_2O/CuFe_2O_4$-loaded silica media filters were made with an internal diameter of 5 cm and a height of 150 cm to investigate the efficiency of filter-loaded media on the removal of total organic carbon, and the filter media put to place at 100 cm height inside the filter. The influent TOC of pilot filter was 8 to 10 mg/L. The maximum TOC removal by the silica sand filter was about 38%, while the $Ag_2O/CuFe_2O_4$-loaded silica sand filter removed more than 97% of the influent TOC in the first 4 h at 120 $m^3/m^2 \cdot d$ filtration rate, pH 7, temperature 25°C and turbidity 10 NTU. Also, the results of TOC removal in different turbidity (10, 25, and 50 NTU) showed that the most TOC removal was in 10 NTU turbidity and then in 25 and 50 NTU, the rate of TOC removal in 8th hour was 97% and 96.7%, respectively. The obtained results of this study confirmed that the studied nanoparticles had positive effects on the reduction/removal of TOC in surface water sources. Also, the results of our study showed that the use of $Ag_2O/CuFe_2O_4$-loaded silica media filter can remove a significant amount of TOC in various conditions such as turbidity, loading rate, pH and temperature from raw water.

Keywords: Water treatment; $Ag_2O/CuFe_2O_4$-loaded silica sand filter; Filtration; Turbidity; Total organic carbon

1. Introduction

Approximately 71% of the earth's surface is covered by water, and the ocean holds about 96.5% of the earth's water that with 97.5% being salt water and 2.5% being fresh water. Climate change, population growth and increasing urbanization have posed great challenges to water supply systems and have always increased the demand for freshwater resources. The World Health Organization (WHO) estimates that 844 million people worldwide lack access to safe drinking water, of which 159 million are dependent on surface water. In the Sustainable Development Plan, the United Nations calls for equitable and universal access to safe and affordable drinking water by 2030 for all countries. Investing in efficient and cost-effective treatment technologies is essential to tackle the effects of water scarcity, especially in low- and middle-income countries [1,2]. Nowadays, the increasing global population and the promoting industrial and agricultural activities have led to inducing pollution loads and hazardous pollutants in the environment, especially the water sources. In the meantime, some evidence reported that organic compounds (OC), some of which have poisonous

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and carcinogenic activities, composed a principal portion of the mentioned pollutants [3,4]. Organic matter refers to the significant source of carbon-based compounds in aqueous environments. Furthermore, these substances can be derived from synthetic organic compounds and various human activities, for example, sewage discharge, misuse/excessive use of fertilizers, insecticides, pesticides, and factors reducing the surface tension of water [5,6]. In general, some researchers stated that the concentration of organic compounds identified in surface sources is higher than groundwater sources [7,8]. The mentioned compounds can also react with chlorine to produce potentially carcinogenic compounds, for example, trihalomethanes.

Organic matter (OM) is predominantly responsible for water quality issues such as color, odor and taste. Colorless water may also have OM present in significant levels. OM can act as a major source of microbial re-growth in the water distribution system, if present in treated water. The major issue with OM is formation of unwanted products such as DBPs upon reaction with chemicals like chlorine. Therefore, it can be concluded that OM can pose serious water quality issues in any drinking water treatment industry if not well treated [9,10].

In general, several technologies have been developed for water purification. Coagulation, flocculation, and sedimentation processes can lead to removing a large percentage of water turbidity-producing colloidal substances. Nevertheless, they have inadequate efficiency in eliminating organic compounds in purifying water process. In this regard, filtration using modified media is known as one of the most common and simplest methods [11]. The filtration process is a way to achieve higher water purification efficiency, and its combination with nanotechnology can have unique impacts on increasing its efficiency. One of the basic approaches to providing safe and drinking water is filtration-based emerging technologies, such as nanoparticles-loaded media filters to treat/purify contaminants originated by chemical and microbial substances in water sources. Nanocatalysts have been extensively studied in the decomposition of many pollutants. Heterogeneous catalysts are one of the most widely used nanocatalysts. Among them, metal oxides have attracted many researcher's attentions due to several properties such as high stability, higher catalytic recovery potential, and reusability in several cycles. In recent years, spinel compounds with the general chemical formula AB_2O_4 have been used to activate a variety of oxidants [12,13].

Ferrite nanoparticles are involved in treating environmental pollutants and are widely employed as adsorbents, catalysts, and photocatalysts. Copper ferrite ($CuFe_2O_4$), also called magnetic copper ferrite nanoparticles (MCFNs), is one of the most famous spinels that has long been used in the metallurgical industries. Due to the high magnetic properties of this compound, it has also been considered in advanced oxidation processes to oxidize organic compounds. Copper ferrite nanoparticles are composed of hematite (Fe_2O_3) and copper oxide (CuO), which can show the property of both metal oxides [14,15] and have been widely used as a heterogeneous catalyst for water and wastewater treatment [16,17].

In this research, a sand filter is made with engineered silica media and modified with a multi-purpose application for advanced removal of water contaminants in the filtration system. Therefore, the present study aimed to investigate the performance of $Ag_2O/CuFe_2O_4$-loaded silica media filters to remove the total organic carbon (TOC) compounds of water supplied from the Karun River.

2. Methodology

2.1. Study design

This research is an experimental-applied research that was conducted on an experimental scale at Ahvaz water treatment plant to investigate the removal of organic matter from Karun River water.

2.2. Pilot characteristics

A twin filter set including silica sand media and $Ag_2O/CuFe_2O_4$-loaded silica media each with an internal diameter of 5 cm, a height of 150 cm and a media height of 100 cm was used to purify TOC containing water (Fig. 1). Turbidity and total organic carbon contents were measured in two stages, before and after passing through $Ag_2O/CuFe_2O_4$-loaded silica media filters. All sampling steps were performed based on the standard methods for the examination of water and wastewater [18].

2.3. Procedures

The first variable was the filtration rate and the filter was operated in three different filtration rates including low (120 $m^3/m^2\cdot d$), medium (240 $m^3/m^2\cdot d$) and high (360 $m^3/m^2\cdot d$). During the 60 h filtration period of the filter at each filtration rate, the turbidity of the effluent was measured hourly and filter effluent TOC was measured with 4 h interval times. Filter was used in three different influent turbidity, low turbidity (10 NTU), medium turbidity (25 NTU) and high turbidity (50 NTU) with 120 $m^3/m^2\cdot d$ filtration rate.

The third variable was the inlet water pH to the filter and the filter will be operated in three different pH including slightly acidic (5), neutral (7) and slightly alkaline (9) with 120 $m^3/m^2\cdot d$ filtration rate and turbidity 10 NTU.

The fourth variable was the inlet water temperature, and the filter was operated in two temperature conditions: summer (August, water temperature 25°C) and winter (February, water temperature 15°C) with 120 $m^3/m^2\cdot d$ filtration rate, 10 NTU turbidity and neutral pH 7. In all the aforementioned conditions, the filter was operated until the effluent turbidity exceeded 1 NTU.

2.4. Experiments

In general, the method 2,130 presented in standard methods for the examination of water and wastewater was used to estimate the turbidity of the samples [18], and the SGE ANATOC™ Series II TOC analyzer was employed to measure the concentration of total organic carbon in raw and filtered samples. For this purpose, 30 ml of water sample was first poured into a laboratory beaker, and its pH value dropped below 3 (using 5% perchloric acid) to eliminate the possibility of mineral carbon interference. In the next step, 1 mL of the resulting sample was injected into the device. In addition, titanium dioxide solution at a rate of

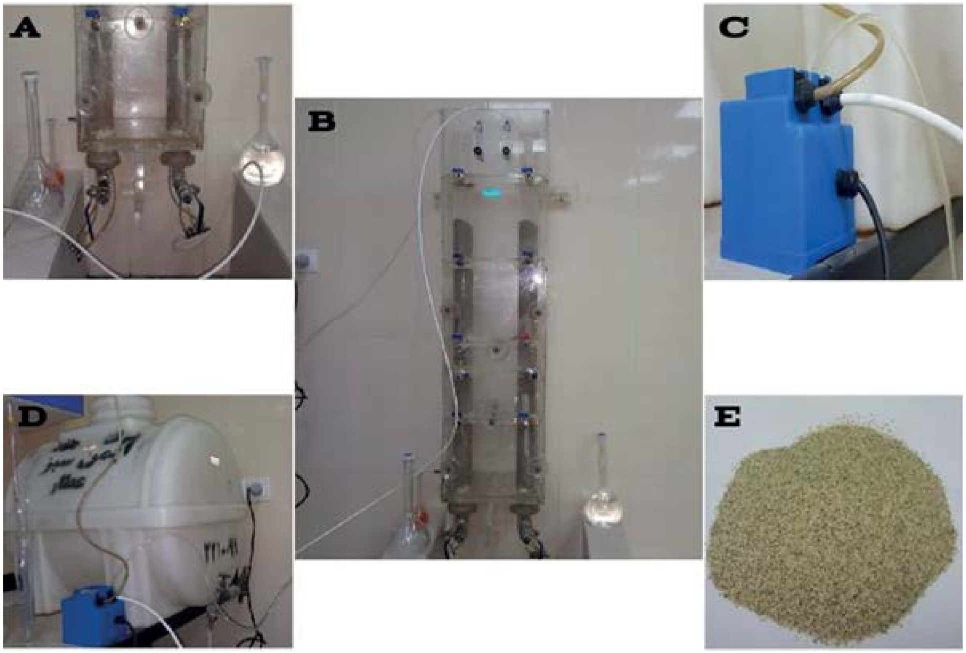

Fig. 1. Photos of the pilot filtration used in the present study.

1 g/L was used for organic matter oxidation. The resulting solution was homogenized before in an ultrasonic device for 15 min following the injection process [8]. It should be noted that the device was calibrated once a week before the tests to reduce the total organic carbon analysis error of the samples. All the statistical analyses and charts were done by Microsoft Excel 2019 software.

3. Results

The present study aimed to evaluate the efficiency of $Ag_2O/CuFe_2O_4$-loaded silica media filters to remove total organic carbon from water samples taken from Karun River. Experimental conditions related to the temperature, pH, TOC, raw water turbidity, filtration rate, and inlet flow to the filter are listed in Table 1. In addition, the data obtained from TOC removal efficiency by the different filters, and the various turbidity, pH, surface overflow rate (SOR) and temperature at different times are presented in Table 2.

The following mentions the results obtained from a pilot laboratory run based on different filters and other conditions. According to Table 2, results showed that the average percentages of TOC removal by $Ag_2O/CuFe_2O_4$-loaded silica sand and silica sand media in 4 to 24 h of the experiment ranged from 97.9%, 95%, 90.8%, 87.5%, 78.3%, 70.4% and 35%, 34.6%, 31.3%, 38.3%, 31.3%, and 28.8%, respectively. The average rate of total organic carbon removal in temperature 15°C and 25°C was 92.4%, 89.5%, 85.3%, 82%, 72.8%, 64.9% and 97.9%, 95%, 90.8%, 87.5%, 78.3%, 70.4%, respectively. Moreover, at pH 5, 7, and 9 and at different times, the results of TOC removal are as follows: Most removal occurred in the first 4 h and at pH 7 (97.9%). Also, the results of TOC removal in different turbidity (10, 25, and 50 NTU) showed that the most TOC removal was in 10 NTU turbidity and then in 25 and 50 NTU, the rate of TOC removal in 8th hour was 97% and 96.7%, respectively. Therefore, the percent of TOC removal in different SOR showed that the maximum of removal occurred at SOR 120 $m^3/m^2·d$ in the first 4 h (97.9%), after that the maximum rate of TOC removal in SOR 240 and 360 were 93.6% and 92% in 8 h after filtration, respectively.

Table 1
Quality of the raw water before filtration process

Parameter	Quantity
Inlet flow, L/h	9.8, 19.6 and 29.5
Filtration rate, $m^3/m^2·d$	120, 240 and 360
Turbidity, NTU	10, 25 and 50
TOC, mg/L	8–10
pH	5–9
Temperature, °C	15 and 25

As shown in Fig. 2, the maximum rate of TOC removal by the silica sand filter (38.75%) occurred during the filtration time of 16 h. However, the rate of TOC removed by silica sand filter (removal efficiency of filter) was again decreased. Also, results related to the turbidity level in the treated water samples showed that the above parameter affected the rate of TOC removal at $p < 0.05$.

The obtained results also show that the average percentage of TOC removal by $Ag_2O/CuFe_2O_4$-loaded silica sand filter in pH 7 at 4th hour is 97.9% (Fig. 3).

As it is shown in Fig. 4, the average rate of TOC removal by $Ag_2O/CuFe_2O_4$-loaded silica sand filter in temperature 25°C was higher than 15°C at the same time.

The results of the $Ag_2O/CuFe_2O_4$-loaded silica sand filter operation in different turbidities showed that the TOC removal decreased during filtration time (Fig. 5).

Table 2
TOC removal efficiency with various situation at different times

Time (h)	TOC removal (%)												
	Type of filter		Temperature (°C)		pH			Turbidity (NTU)			SOR (m^3/m^2·d)		
	Silica sand	$Ag_2O/CuFe_2O_4$-loaded silica	15	25	5	7	9	10	25	50	120	240	360
4	35.0	97.9	92.4	97.9	96.3	97.9	94.2	97.9	97.0	96.1	97.9	93.2	89.3
8	34.6	95.0	89.5	95.0	96.7	95.0	90.8	95.0	97.0	96.7	95.0	93.6	92.0
12	31.3	90.8	85.3	90.8	93.8	90.8	87.5	90.8	95.0	95.5	90.8	89.3	87.0
16	38.3	87.5	82.0	87.5	90.4	87.5	83.8	87.5	93.0	93.3	87.5	83.9	80.3
20	31.3	78.3	72.8	78.3	82.9	78.3	74.2	78.3	89.5	90.6	78.3	69.6	65.2
24	28.8	70.4	64.9	70.4	77.1	70.4	62.1	70.4	81.8	85.1	70.4	66.8	59.9

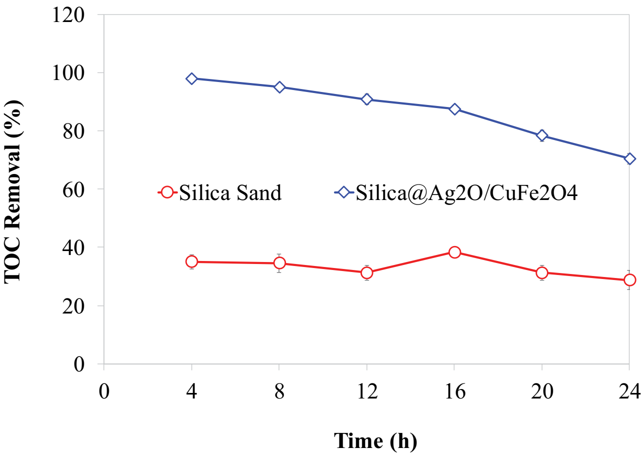

Fig. 2. TOC removal by the silica sand and $Ag_2O/CuFe_2O_4$-loaded silica sand filter.

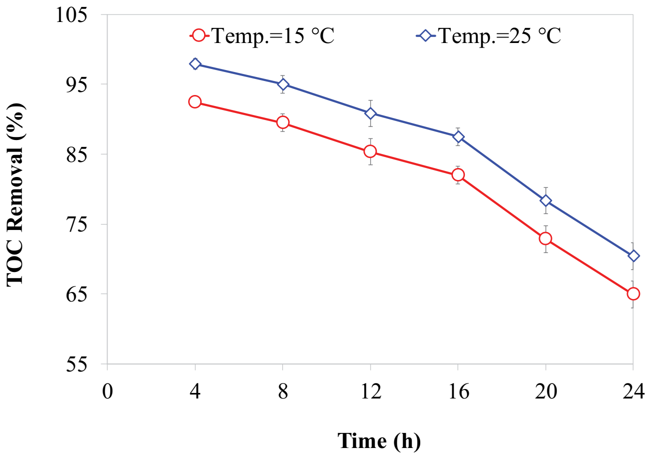

Fig. 4. TOC removal by the $Ag_2O/CuFe_2O_4$-loaded silica sand filter over time at different temperatures.

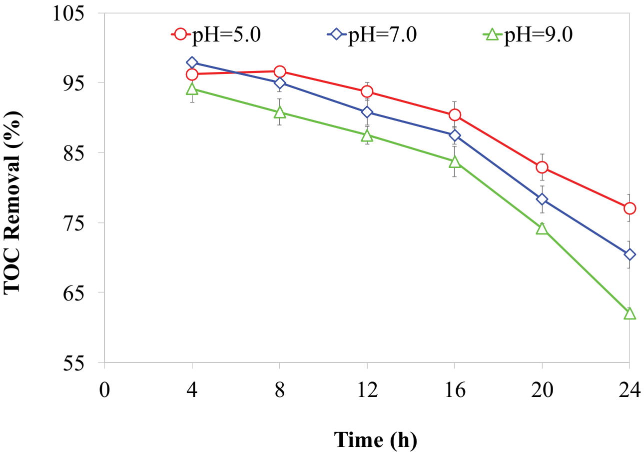

Fig. 3. TOC removal by the $Ag_2O/CuFe_2O_4$-loaded silica sand filter at different pH.

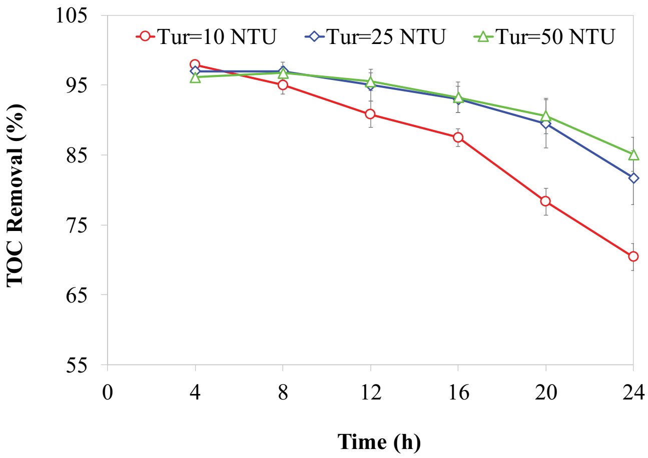

Fig. 5. TOC removal by the $Ag_2O/CuFe_2O_4$-loaded silica sand filter over time at different turbidities.

In this research, we also measured the average of TOC removal in different SOR over time. According to the results, the maximum rate of TOC removal occurred in SOR (120 m^3/m^2·d) (Fig. 6).

4. Discussion

Surface water is more exposed to pollutants than groundwater. Accordingly, the processes of treatment and removal of contaminants for these water resources are more

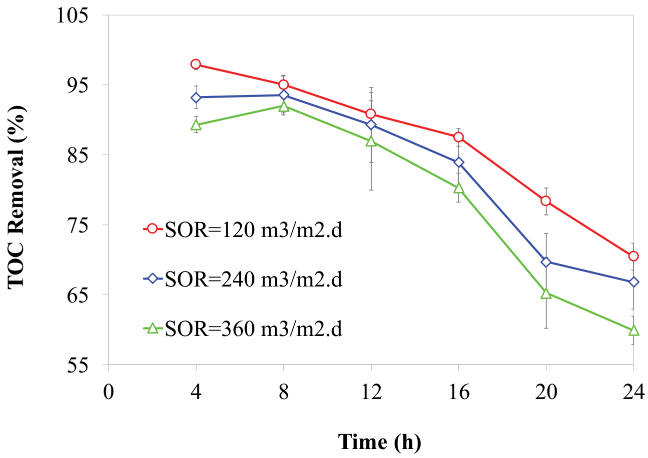

Fig. 6. TOC removal by the $Ag_2O/CuFe_2O_4$-loaded silica sand filter over time in different SOR.

complex. Most surface water resources have more organic compounds and turbidity than the specified amount by standards presented for drinking water quality [19]. On the other hands, since the main purpose of conventional water treatment plants is to eliminate turbidity, a significant amount of TOC concentration is reduced as a result of turbidity removal. Many of studies showed that there is a direct relationship between turbidity and TOC [20,21]. To improve the TOC removal efficiency in water treatment plants, it is necessary to use processes such as enhanced coagulation, chemical oxidation, adsorption and membrane methods. According to previous studies, membrane methods offer high efficiency to reduce TOC from surface water in different conditions [15–17]. Hence, in the present study, the reduction of organic carbon in the water sample prepared from Karun River was examined using a filter containing modified $Ag_2O/CuFe_2O_4$-loaded silica media. According to the results, in influent TOC concentration of 8 to 10 mg/L the maximum TOC elimination by $Ag_2O/CuFe_2O_4$-loaded silica sand filter was much more than silica sand filter, which was >97% and about 38%, respectively. Since media components of the $Ag_2O/CuFe_2O_4$-loaded silica sand filter have unique properties including oxidation and adsorption, therefore probably oxidation and adsorption processes combined with filtration can be considered as active factors improving the TOC removal efficiency. Results of the present study were consistent with the findings of the study presented by Kazemi Noredinvand et al. [22]. Latter researchers reported that the average removal of organic compounds by the filtration process was equal to 34.1%.

As shown in Fig. 3 in the current study, by increasing inlet water pH from 5.0 to 9.0, after 24 h filtration time the TOC removal was decline 15%. Likewise, Jeong et al. [23], observed only 2% improvement in TOC removal by increasing the pH from 6.5 to 7.5. However, Pirouz Hamidi [24] reported that by further increasing the water pH to 9.0 and then 10.0, TOC removal significantly decreased to 53% and 31%, respectively. In their study, the carbon mixture was primarily composed of carboxylic functional groups; thus, the decrease in performance is partially attributed to this factor. Moll and Summers [25] reported that carboxylic and amino acids were better utilized in a neutral or acidic pH environment, while microorganisms used carbohydrates better in filters operated at a pH value of 7.5. Alkaline pH levels lead to a reduction in TOC removal efficiency, which is likely due to the oxygen scavenging effect of carbonate ions that remove oxygen or prevent oxidation.

According to Fig. 4, in our study and past studies, temperature effect as a water quality parameter has also been evaluated. Some authors found that fewer organic matters were removed at low temperatures (≤5°C) than high temperatures (20°C) in anthracite or sand filters [26,27]. In some studies, low temperature (≤5°C) was reported to reduce TOC removal by 9% [25] and 6% [27]. Also, Liu et al. [26] indicated that the removal of various fractions of organic carbons in the range of 70% to 90% at 20°C reduced to 10% to 50% at 5°C. However, this adverse temperature impact was not observed in GAC filters [27]. On the other hand, TOC removal was not affected by temperature changes at values higher than 10°C [28,29]. As the studies reviewed in the previous section were mainly indoor bench scale or pilot scale studies, the seasonal temperature variation could not be accounted to be a compelling factor for the different results obtained in different studies.

As we shown in Fig. 5, the results of our study showed that the maximum TOC removal occurred in 50 NTU turbidity. In other studies, that is, Kazemi Noredinvand et al. [22] and Kiashemshaki et al. [30], it was stated that with increasing turbidity, the rate of TOC removal was higher, which is consistent with the results of the present study. The most important possible reason for these observations is the reduction of filter pore size with increasing turbidity and therefore increasing the filter performance to remove organic compounds [22]. In this study, the rate of TOC reduction over time at different pH was also investigated. The highest removal of TOC obtained in pH 5.0 over time. On the other hands, the results showed that there is a relationship between the temperature and TOC removal, thus, by increase the temperature, the average of TOC removal also increased over time. In 2005, in a study in Iran, Torabian et al. [31] concluded that there is a relationship between air temperature and the efficiency of TOC elimination. This research also reported that the processes employed at the Tehranpars water treatment plant did not achieve a good efficiency in terms of TOC removal. The results of Kiashemshaki et al. [30] conducted that when the input TOC was at its maximum value, TOC removal efficiency was around 80%. This could potentially be increased by augmenting the amount of coagulant substance or through the use of coagulant aid. The removal efficiency reduced as the concentration of TOC input reduced. Unfortunately, the influent TOC concentration was almost constant in the present study (8–10 mg/L). Therefore, we could not investigate the role of TOC concentration and the effect of its removal from water. Many studies have shown that low loading rate (120 $m^3/m^2{\cdot}d$) can enhance the efficiency of TOC removal over time, which was also shown in the present study [22,30,32].

5. Conclusion

The obtained results of this study confirmed that the studied modified silica sand media had positive effects on the reduction/removal of TOC from surface water sources. Also, the results of our study showed that the use of $Ag_2O/CuFe_2O_4$-loaded silica media filter can remove a significant amount of TOC in various conditions such as turbidity, loading rate, pH and temperature from raw water. Since nanomaterials have reasonable prices, high environmental compatibility (environmentally friendly), and effectiveness in reducing water pollution, these materials can be used in the structure of filters used in water purification.

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Conflict of interest

All authors of this study have equally contributed to designing, conducting, and preparing this manuscript and declare no conflict of interest in publishing this manuscript.

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