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Removal of heavy metals from groundwater using silica/activated carbon composite

Mutairah Shaker Alshammari[a], Wassila Derafa[a,b,*], Eiman A.A. Elshaygi[a]

[a]*Chemistry Department, College of Science, Jouf University, P.O. Box: 2014, Sakaka, Saudi Arabia, Tel. +966 557326506; email: Wderafa@ju.edu.sa (W. Derafa), Tel. +966 553118365; email: msshamari@ju.edu.sa (M.S. Alshammari), Tel. +966 508914544; email: eimanelshaygi@gmail.com (E.A.A. Elshaygi)*
[b]*Laboratoired' Electrochimie, d'Ingénierie Moléculaire et de Catalyse Redox (LEIMCR), Faculté des Sciences de l'Ingénieur, Université Farhat Abbas, Sétif 19000*

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ABSTRACT

Contaminant monitoring is critical for ensuring the safety of drinking water. This work aimed to study the physicochemical characteristics of five wells used as water resources in the Al-Jouf region (Guara, Tabarjal, Skaka, Dumah Al Jandal, and Alakait), Saudi Arabia. The study extended to investigate the treatment of the contaminated water wells. The physicochemical characteristics include pH, conductivity, turbidity, and some selected heavy metals (cadmium, chromium, lead, and arsenic). The quality of the treated wells' water was correlated with the guidelines of the Environmental Protection Agency (EPA), the World Health Organization (WHO), and Saudi Arabia's standards. The results indicated that the concentrations of the measured heavy metals were higher than the National Standards of Saudi Arabia and that recommended by the EPA and WHO. The capacity of activated carbon (AC) combined with silica (silica/AC) and AC composite as adsorbents for the removal of the selected heavy adsorption. The silica/AC ratio was 2:3, respectively. The silica/AC was more efficient than active carbon in removing the selected heavy metals. Scanning electron microscope photos were taken for the adsorbents used for the removal of toxic metals. This work concluded that the use of silica/AC composite is a promising technique for the treatment of contaminated water wells.

Keywords: Heavy metals; Treatment; Adsorption, Activated carbon (AC); Silica/Activated carbon; Composites

1. Introduction

Drinking water is subjected to various pollutants, contingent upon its resources. Surface water tainting happens when water goes on or through the dirt surface. Thus, pollution occurs in many ways, such as the discharge of untreated sewage-water, industrial, agricultural wastewater, and leachate of landfill sites. While moving, it disintegrates pollutant particles and gets minerals coming out. The pollutants in groundwater require more effort to be scrubbed [1]. The appearance, taste, turbidity, and smell of the water control its appeal as a source of drinking. But the appearance of water doesn't reflect the quality of water [2]. The World Health Organization (WHO) and Environmental Protection Agency (EPA) set guidelines for pollutants concentration in drinking water [3,4]. When the water quality decreased, it needs to be treated to comply with the International Regulatory Standards [3,4]. Heavy metals pollute water resources such as anthropogenic and geological sources [5,6].

* Corresponding author.

The discharge of heavy metals (HM) into the environment has the potential to endanger water, soil quality, and plant, animal, and human health, both directly and indirectly. If heavy metal levels reach particular thresholds, they have a negative impact on soil fertility, plant, and animal output, and the environment as a whole [7]. Due to their intractable nature, toxicity to accumulate in organisms, food chain, and non-biodegradability, HMs have been identified as one of the most harmful minerals. It is very poisonous to humans, causing harm to the circulatory, digestive, neurological, and endocrine systems, as well as the kidneys, liver, lungs, and bones [8–10]. Drinking water contains trace amounts of heavy and trace metals that help the human body function normally, but too much can be harmful to one's health. Ingestion of these HMs incorporated with food and water allows them to enter the human system [11]. So far, various strategies for removing heavy metals from water and other aqueous solutions have been investigated and implemented. Adsorption, ion exchange, precipitation, electrochemical treatment, and membrane filtration are some of the most commonly used procedures [12].

However, adsorption has proven effective and successful in heavy metals, natural pollutants, and colors from polluted water [13]. A few adsorbents, for example, activated carbon, silica and graphene can be utilized in water purification. Activated carbon has been demonstrated to be an effective adsorbent for the removal of heavy metals due to its high surface area which ranges from 500 to 1,500 m^2/g [14–17].

The aim of this work was to measure the adsorption efficiency of activated carbon (AC) combined with silica (silica/AC) and AC composite for the removal of heavy metals (lead, arsenic, cadmium and chromium). Groundwater samples were collected from five locations at Al-Jouf City, Saudi Arabia.

Table 1 shows the permissible values of the studied heavy metals according to the World Health Organization (WHO) [18] and Saudi Arabia standards [19].

2. Materials and methods

2.1. Sampling

The samples were collected from 5 wells (with a depth of about 25–75 m). These wells are the main drinking water source in Al-Jouf City. The wells located in Guara, Tabarjal, Skaka, Dumah Al Jandal, and Alakait, in Al-Jouf City. The tests cover measuring the concentrations of lead, cadmium, arsenic and chromium well's water as well as the pH, turbidity, and conductivity. Within 2 h, the samples were delivered in an icebox to the laboratory. The containers used to handle the samples (1 L volume) were rinsed many times with double distilled water. The abbreviations of samples take as this (WG: Well Guara, WT: Well Tabarjal, WS: Well Skaka, WD: Well Dumah Al Jandal and WA: Well Alakait).

2.2. Samples preparation for heavy metals determination

The samples of 100 mL were digested with 5 mL nitric acid then cooled. About 5 mL of hydrogen peroxide were added to the cooled samples and continue the digestion of the samples. The samples were filtered using filter paper (Whatman 4) [20]. The samples were stored in the refrigerator at +4°C when not in use. The standard heavy metal solutions were used to get the calibration curve.

2.3. Chemicals

All the chemicals and AC used in this study were bought from Sigma-Aldrich (Germany) and were of analytical grade. Atomic absorption spectrophotometer (AAS) technique type AA-6800, Shimadzu (Japan), assessed the amounts of the heavy metals under analysis. The pH 4, 7, and 9 buffer solutions for pH-meter calibration and concentrated nitric acid (63%) were of high quality and obtained from Sigma and Fisher (Germany).

2.4. Instrumentation

All heavy metal investigations were conducted on Agilent 5100 inductively coupled plasma – optical emission spectrometer (ICP-OES) with a synchronous vertical dual view. For every series of assessment intensity calibration curves were established from a blank and three or more standards from Merck Company (Germany). The exactness and accuracy of the metals estimations were affirmed utilizing external reference standards from Merck (Germany), and standard reference material for the following components in water and quality control tests from the National Institute of Standards and Technology (NIST) were utilized to affirm the instrument perusing. The pH of the water tests were initially assessed utilizing

Table 1
Guideline values for the recommended concentration of examined heavy metals in μg[18,19]

Metal	Concentration according to WHO (μg)	Concentration of heavy metals according to the Saudi Arabia standards (μg)								
		Bottled			Tap			Treated		
		Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Pb	ND		ND			ND			ND	
Cd	3 μg		ND			ND			ND	
As	10 μg	0.235	4.08	1.51	0.45	3.65	1.87	0.788	2.878	1.585
Cr	50×10^3 μg/L		ND			ND			ND	

the pH – Hanna pH 211, conductivity meter EC-Jenway 4510 (Germany) was utilized and Lovibond as turbidimeter.

2.5. Preparation of AC, and silica/AC (2:3) composite

The AC was modified followed the same technique referenced by Yin et al. [21] by a heating process where it was rinsed with water and thereafter heated at 450°C for 4 h. Silica/AC (2:3) composite was set up with the same method introduced by Wang et al. [22]. The surface alteration of silica (SiO_2) with (3-Aminopropyl)triethoxysilane (APS) was completed in the fluid stage. 10 g of SiO_2 was then mixed with 150 mL ethanol, under continuous stirring for 30 min to get very much scattered SiO_2 suspension. After the option of another 150 mL ethanol, 0.5 mL APS was included in the suspension. The blend was mixed, heated to 50°C for 12 h, the product was then separated, rinsed with ethanol followed by deionized water and dried under vacuum. Air conditioning covered silica composite (SiO_2/AC) was set up as silica/AC (2:3 W/W) proportion. Under gentle blending for 1 h till the arrangements got straightforward, and AC accelerated with SiO_2-NH_2. The resultant SiO_2/AC was gathered and rinsed with double distilled water several times and dried at 60°C.

2.6. Removal of Pb, Cd, As, and Cr by AC, and silica/AC (2:3) composite

Removal of Pb, Cd, As, and Cr by AC and silica/AC (2:3) composite was tried utilizing the recently referenced adsorption strategy [23]. The dose of 20 mg of the prepared composite was used for the treatment of water samples of 20 mL. The arrangements were gently sonicated for 20 min, at the ambient temperature. Solids were allowed to settle down, and the supernatant was collected. The AAS was used to determine the concentration of the pre-mentioned heavy metals.

2.7. Microstructure of the materials

The microstructure of the materials was observed with scanning electron microscopy (TESCAN SEM, INCA, Germany).

3. Results and discussions

3.1. Significant physicochemical parameters as affected by activated carbon and silica/AC (2:3) composite

The quality of the collected water samples from different wells in the Al-Jouf region was first tested by measuring the pH, turbidity and conductivity, the results are shown in Table 2.

3.1.1. pH values

Measuring pH is important for determining the acidity or alkalinity of water. The water sample has a pH of less than 7.0 and is considered slightly acidic. The pH values were found to be lower than those obtained by El-Hajoj et al. [6]. The pH results correlated with the standard specifications outlined by the Saudi Arabia specification; (6.5 to maximum 8.5). It was found that samples of untreated water were to be within the permissible limits (6.70 to 8.15) as shown in Table 2. The obtained pH values after treatment with AC were within the recommended range also (8.1 to 6.5) as shown in Fig. 2a. The silica/AC composite is very effective in balancing the pH of water samples. Acidic water leads to metal corrosion for pipes in potable water plants, this may liberate metal ions such as lead in treated water. Drinking water with a high concentration of heavy metals caused health risks [24,25].

3.1.2. Turbidity

The turbidity was ranged from 0.2 to 0.7 NTU. The turbidity was within the acceptable range for Saudi Standards

Table 2
Physicochemical parameters in drinking water collected from water wells before, after treatment with AC and composite silica/AC

Region	Parameter	Before treatment	After treatment with activated carbon	After treatment with composite silica/AC
Well Guara	pH	8.15	8.03	8.1
	Turbidity (NTU)	0.5	0.4	0.2
	Conductivity (µS/cm)	360	355	350
Well Skaka	pH	7.5	6.7	7.4
	Turbidity (NTU)	0.2	0.1	0.1
	Conductivity (µS/cm)	231	215	210
Well Tabarjal	pH	7.31	7.25	7.3
	Turbidity (NTU)	0.7	0.4	0.2
	Conductivity (µS/cm)	2,334	2,320	2,320
Well Dumah Al Jandal	pH	7.15	7.02	7.1
	Turbidity (NTU)	0.6	0.4	0.3
	Conductivity (µS/cm)	844	810	810
Well Alakait	pH	6.7	6.5	5.6
	Turbidity (NTU)	0.7	0.4	0.2
	Conductivity (µS/cm)	1,616	1,610	1,605

(lower than 5.00 NTU) Table 2. Treatment with AC and composite silica/AC reduced the turbidity of the five wells to a lower reading ≤0.4 and ≤0.3 NTU respectively as shown in Fig. 2b.

3.1.3. Electrical conductivity

Pure water is not a good conductor of electric current and a good insulator. The electrical conductivity (EC) increases with increasing the ion concentration of solutions. In this study, the EC of the water samples before treatment ranged from 360 to 2,334 µs/cm (Table 2). Each of Guara and Skaka recorded levels complying with Saudi Arabia standards (less than 400 µs/cm) while samples collected from Tabarjal, Alakait and Dumah Al Jandal wells recorded higher values (2,334; 1,616 and 844 µs/cm) respectively. This indicated that they have a higher level of ionic concentration activity due to high dissolved solids. After treatment of the samples with AC and composite silica/AC, the EC was reduced significantly but did not reach the permissible range by Saudi Arabia standards ≤400 µs/cm. Karnib et al. [23] showed that the capacity of removal of ions by AC decreased with the high concentration of metal ions. The deionization process is restricted by the total pore volume and size dispersed on the AC. As the sites become saturated with metal ions, the AC cannot eliminate ions and become less adsorption efficiency [26] as in Fig. 1c.

3.2. Heavy metals concentrations removal with AC and silica/AC (2:3) composite

3.2.1. Lead (Pb)

All the samples before treatment contained more than EPA, WHO and Saudi Arabia standards [18,19]. Contamination of lead is the largest in the WT sample and the lowest in the WS sample. The lead concentration in the sample of WT reached the level of 0.04 mg/L. This high level of Pb may be attributable to the well's plumbing systems themselves (pipes and fittings). Lead in drinking water arises from lead-lined pipes within the well unit, welding, and brass bathroom fixtures. The EPA reported 98% of all houses in the well hold piping to have pipes, fixtures or weld joints that can add amounts of lead to the well water [27]. It is because the rate of growth for young children is much higher than for an adult. Lead can grow over a period in the human body and cause significant damage to cells in the brain, kidney, nerve and red blood. Significant amounts of lead can cause problems in brain development for infants [28]. The concentration of Pb in all samples was reduced after treatment with AC. The concentration of Pb obtained was not complying with the permissible levels recommended by the WHO and the Saudi standards except for samples of WS. Whereas after treatment with silica/AC composite, the obtained concentrations were within the range of WHO and the Saudi standards except for samples (WT) and (WA) as shown in Fig. 2a. It was noted that at a concentration higher than 0.03 µg/L the silica/AC composite could not remove Pb completely.

3.2.2. Chromium (Cr)

The concentration of Cr was the same (0.002 mg/L) in samples of WT, WD, and WA and the lowest (0.001 mg/L) in samples of WG and WS, but the concentration was exceeding the Saudi Arabian standards in all well's samples (Table 1). Although the Cr metal is found in all samples, it was below the recommended WHO limit (0.05 mg/L). Yet the amounts of chromium in water are normally very low. Manufacturing systems where sodium dichromate (hexavalent) solutions are often used to avoid deterioration in the piping is considered as a protentional source of Cr metal ions. No Cr involving heavy industry exists in Saudi Arabia. So, the existence of Cr in well water may be attributed to the rocks containing well water [29]. The organic matter reduces chromium VI to Cr(III). The Cr(III) exits at 0.1 mg/L can influence respiratory disorders, cognitive hemorrhage, and kidney failure although chromium VI can cause dermatitis and ulceration [30]. After treatment with AC, the concentration of Cr was decreased in all samples. The residual concentrations of Cr were within the range of WHO and Saudi Arabia standards except for samples WG and WA. Whereas after treatment with silica/AC composite, the residual concentrations for all well water were within the range of both WHO and the Saudi Arabian standards as seen in Fig. 2b.

3.2.3. Cadmium (Cd)

Usually, the concentration of Cd in water is very low. The deterioration of some galvanized piping materials and the main water piping materials may add Cd to drinking water. There may be higher concentrations of Cd in the water close to manufacturing areas or hazardous waste sites. The Cd contents in collected well water samples were very low. The greatest Cr concentration was found in the sample of WA while the lowest concentration was detected in the sample of WS. All samples contain Cd higher than the WHO level and Saudi Arabia standards, except for the WS sample which containing 0.002 mg/L lower than the WHO level. Cd has significant potential for short-term health consequences like vomiting, nausea, muscle cramps, diarrhea, sensory disturbances, salivation, hepatic damage, shock, seizures, and renal failure [31]. Lifelong sensitivity to Cd at concentrations higher than 0.005 mg/L may cause damage to the kidney, liver, bone, and blood [15,17]. After treatment with AC, the concentration of Cd was decreased in all samples. The residual concentrations of Cd were within the range of WHO but higher than Saudi Arabia standards. Whereas after treatment with silica/AC composite, the obtained Cd concentrations were all within the range of WHO and only two samples (WS and WD) were within the range of Saudi Arabia standards but three samples WG, WT, and WA were more than the range of Saudi Arabia standards as represented in Fig. 2c.

3.2.4. Arsenic (As)

Naturally As found in the groundwater with various concentrations. As in its inorganic form, is extremely poisonous. Polluted water used for drinking, preparing

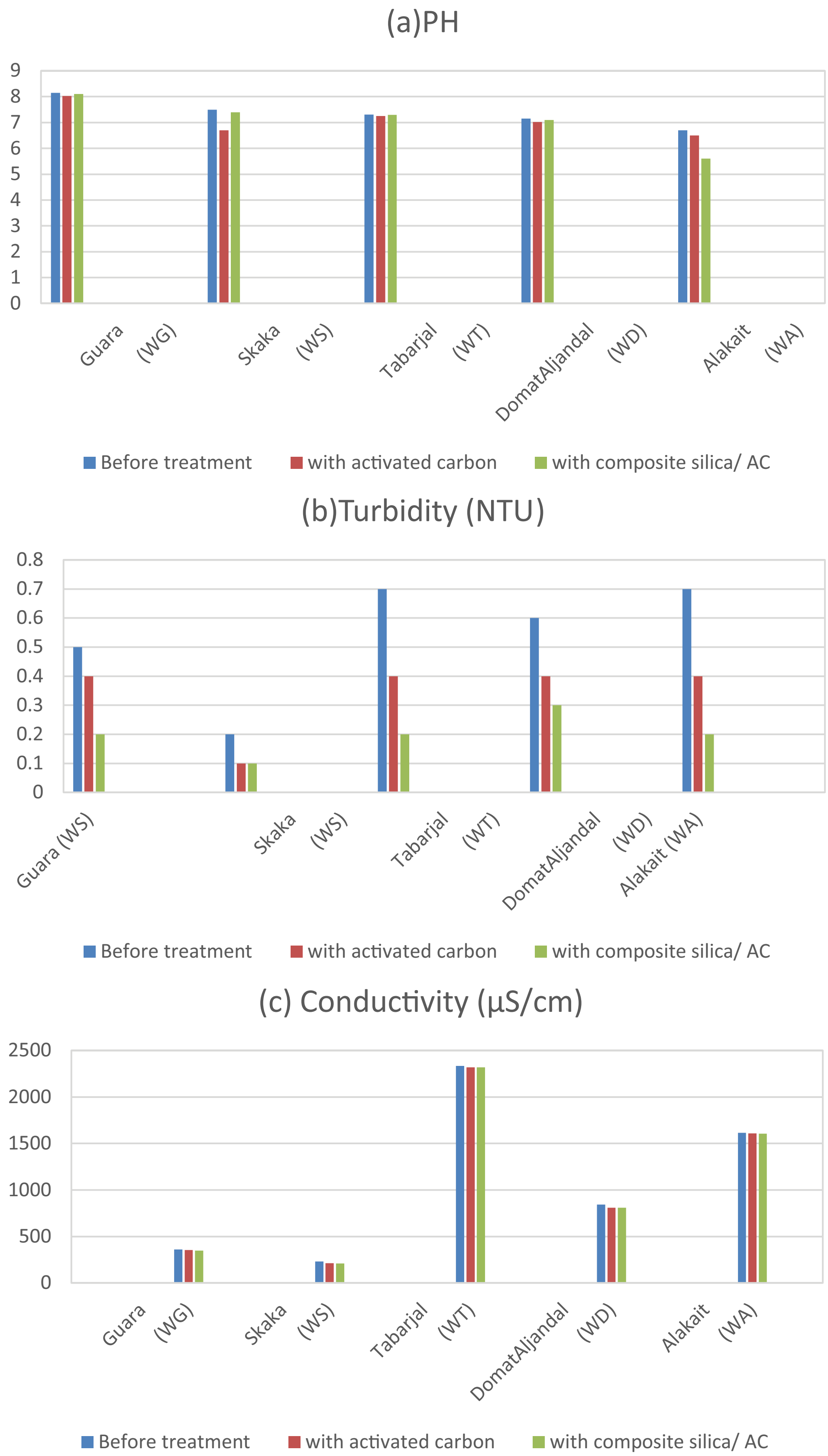

Fig. 1. Values of physicochemical parameters in drinking water well: (a) pH, (b) turbidity, and (c) conductivity.

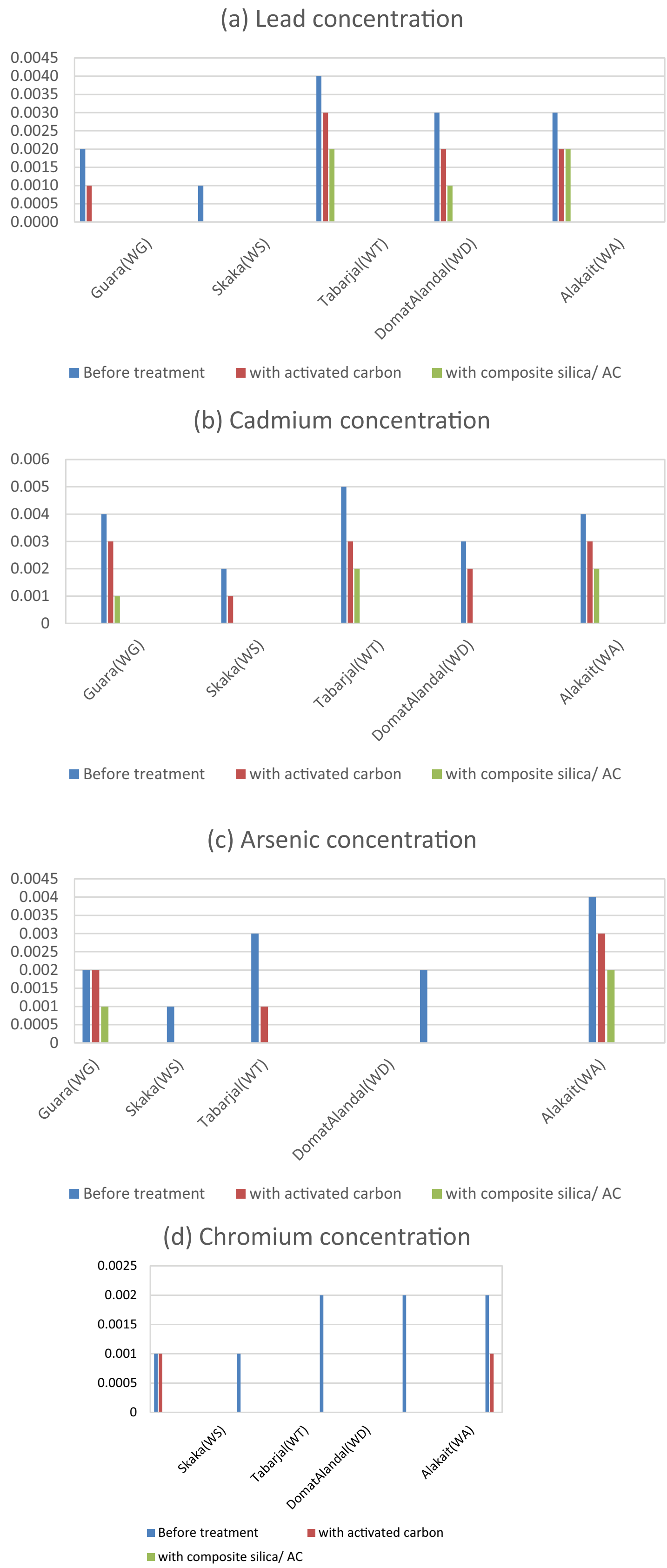

Fig. 2. Heavy metals concentrations in drinking water well: (a) lead, (b) cadmium, (c) chromium, and (d) arsenic concentration.

foods, and irrigating plants pose the danger of arsenic toxicity to public health. In the sample of WT, the highest concentration of As was detected while the lowest concentration was observed at the sample of WS as shown in Fig. 2d. Long-term drinking water and food exposure to As may lead to cancer and skin lesions. It was also related to cardiovascular disease and diabetes [31]. Exposure in utero and early childhood has been associated with adverse effects on cognitive performance and increased deaths of young adults [15,16]. The most effective step in the communities affected is to avoid more exposure to As by ensuring a safe water source. After treatment with AC, the concentration of As was decreased in all samples. The residual concentrations of As were within the range of WHO and Saudi Arabia standards. Also, after treatment with silica/AC composite, the obtained concentrations were all within the range of WHO and of Saudi Arabia standards.

3.3. Characterization of the adsorbents under test

Fig. 3 shows the scanning electron microscopy (SEM) of the adsorbents used in the treatment of water wells at magnifications of 1 and 4 μm. Investigation of the SEM image (Fig. 3a and b) displays the AC exhibited a small size distribution, with an average size of 3.45 μm, so it was a nanoparticle. The silica nanoparticles were indicated by arrows and have an average size of 5.18 nm as shown in Fig. 1c and d that showing the nanoparticles of pure silica appeared in the form of white aggregates at a magnification of 4 μm. While the silica/AC composite nanoparticles (2:3) were represented in Fig. 1e and f, with a magnification of 1 and 4 μm, the silica gel nanopores were clear on the surface of the AC nanoparticles.

4. Conclusion

The current investigation was carried out to determine the quality of drinking well water from various locations in Al-Jouf City. Well water samples were analyzed for the physical examination (pH, turbidity and conductivity) and the concentrations of toxic metals ions before and after treatment with AC and silica/AC composite. Four toxic metals were detected in all samples. The metals were exceeding the WHO and Saudi Arabian standards in water samples before treatment. The presence of toxic metals in drinking water was in the following order Cd > Pb > As > Cr. After treatment with AC and silica/AC composite, the concentrations of these metals were within the range of WHO and Saudi Arabia standards. The composite silica/AC (2:3) displayed the highest removal percentage comparing with AC towards heavy metals following this order Cd < Pb < As < Cr. This means that the metal ions accessible to the adsorbent surface are very small at a high initial concentration of metal so that functional adsorption is dependent on the initial concentration.

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Table 3
Heavy metals concentrations (μg/L) in drinking water collected from water wells before, after treatment with activated carbon and composite silica/AC

Region	Heavy metals	Before treatment	After treatment with activated carbon	After treatment with composite silica/AC
Guara	Lead	0.002	0.001	ND
	Cadmium	0.004	0.003	0.001
	Arsenic	0.002	0.002	0.001
	Chromium	0.001	0.001	ND
Well Skaka	Lead	0.001	ND	ND
	Cadmium	0.002	0.001	ND
	Arsenic	0.001	ND	ND
	Chromium	0.001	ND	ND
Well Tabarjal	Lead	0.004	0.003	0.002
	Cadmium	0.005	0.003	0.002
	Arsenic	0.003	0.001	ND
	Chromium	0.002	ND	ND
Well Dumah Al Jandal	Lead	0.003	0.002	0.001
	Cadmium	0.003	0.002	ND
	Arsenic	0.002	ND	ND
	Chromium	0.002	ND	ND
Well Alakait	Lead	0.003	0.002	0.002
	Cadmium	0.004	0.003	0.002
	Arsenic	0.004	0.003	0.002
	Chromium	0.002	0.001	ND

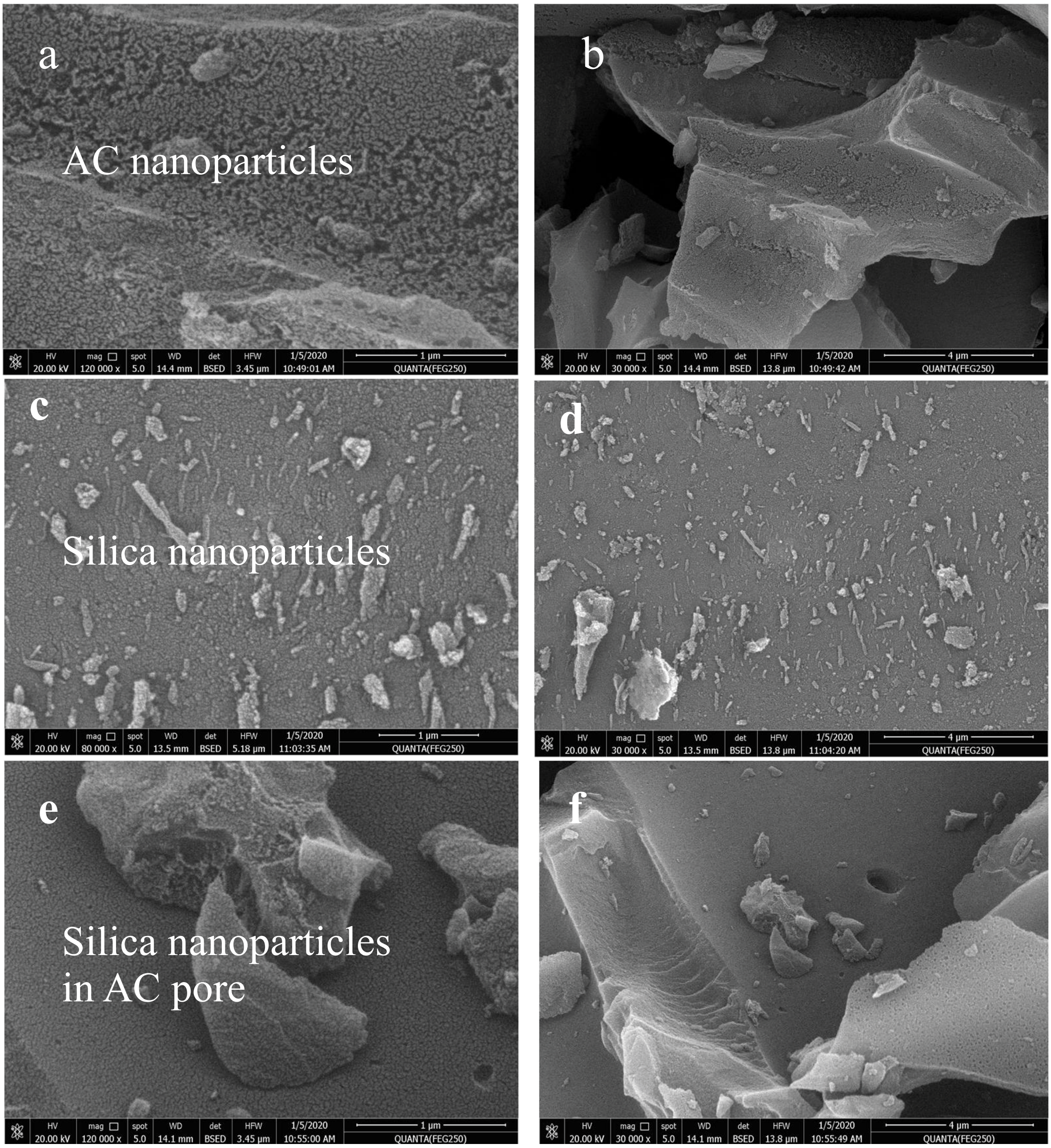

Fig. 3. Scanning electron microscopy images of: (a) AC 1 μm, (b) AC 4 μm, (c) silica 1 μm, (d) silica 4 μm, (e) silica/AC (2:3) composite at 1 μm and (f) silica/AC (2:3) composite at 4 μm.

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