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Arsenate Removal from Wastewater Using Solid Phase Extraction (SPE) Onto Functionalized Polyvinylbenzylchloride-Based Diquaternary Ammonium Nanofibres

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Abstract:

An estimated 30 to 35 million Bangladesh people are exposed to arsenic concentrations above 50 $\mu g l^{-1}$ (World Health Organization (WHO) guideline value prior to 1993), and the number of people exposed to more than 10 $\mu g l^{-1}$ (current WHO guide line value) is 46 to 57 million. The problem of wastewater is acute among developing and industrialized countries that are still struggling with the toxic effect of waste water. Efforts are being made to remove these contaminants, arsenic (v) from wastewater using low cost adsorbents. The removal of arsenic(v) from wastewater using two novel fabricated sorbent materials; aminated polyvinylbenzylchloride (PVBC) prepared through a substitution reaction and then quaternized using three alkyl groups ($R' = CH_3$, C_2H_5 and C_3H_7) were investigated. The experiments were carried out in solid phase extraction mode. Preliminary experiments were done with series of three sorbent materials ($R' = CH_3$, C_2H_5 and C_3H_7) in order to find out a suitable and low-cost adsorbent for the effective removal of arsenic (v). The best sorbent materials were obtained after the optimization and preparation experiments. The $R' = CH_3$ were effective for arsenic (v) removal from wastewater. The effect of pH, initial sorbate concentration, amount of sorbent, effect of co-existing anions and the reusability in solid phase extraction mode were also investigated. From the experiment conducted, the pH shows a decisive factor on arsenic removal. Analysis of adsorbent dosage, Kinetic studies and co-existing anions were carried out in order to evaluate sorption parameters.

1. Introduction

The presence of arsenic in wastewater has reached calamitous proportions in many parts of the world. Arsenic exists as both inorganic and organic compounds and its toxicity is related to its chemical form (Mazan et al., 2002).

It is widely distributed in nature in the form of either metalloids or chemical compounds, primarily present in inorganic forms and exists in two predominant species: arsenate and arsenite. Arsenite is much more toxic (Ferguson and Gavis, 1972), soluble and mobile than arsenate. Arsenate ($H_2AsO_4^-$ and $HAsO_4^{2^-}$) is the predominant form of arsenic in well-oxidized waters, while arsenite occurs predominantly as H_3AsO_3 and $H_2AsO_3^-$ in reduced environments (Ferguson and Gavis, 1972). The speciation of inorganic arsenic, As species is important when considering toxicological studies and remediation techniques, particularly since the latter is highly species dependent. Two factors (pH and redox potential) are very important in controlling As speciation (Mohan et al., 2007). As⁵⁺ species ($H_3AsO_4^-$, $HAsO_4^{2^-}$, and $AsO_4^{3^-}$) are dominant under oxidizing conditions, with $H_2AsO_4^-$ dominates at low pH (pH < 6.9) and $HAsO_4^{2^-}$ at higher pH, while H_3AsO_4 and $AsO_4^{3^-}$ may occur in extremely acidic and alkaline conditions, respectively. On the contrary, As^{3+} species (H_3AsO_3 , $H_2AsO_3^-$, and $HAsO_3^{2^-}$) are stable under reducing (or anoxic) conditions, with the most common being the uncharged species H_3AsO_3 at pH < 9.2 (Smedley et al., 2002).

In wastewater, arsenic occurs both naturally and through anthropogenic activities, that is, human influences. Arsenic can be naturally derived from several parent materials in industrial wastes, including magnetic sulphides and iron ores. The most important arsenic ores are the arsenopyrites, which are often present in sulfide ores. Human influence can result in a build-up of arsenic, which includes the use of arsenical pesticides and the application or disposal of industrial and animal wastes (Xie et al., 2006).

Arsenic is a highly toxic metalloid, it is brittle, crystalline, odourless and tasteless in its elemental form (William and Keneet, 1989). Arsenic rarely occurs in a free state, it is largely found in combination with sulphur, oxygen, and iron (Jain and Ali, 2000).

In groundwater, arsenic combines with oxygen to form inorganic pentavalent arsenate and trivalent arsenite. Unlike other heavy metalloids and oxyanion-forming elements, arsenic can be mobilized at the pH values typically found in surface and groundwaters (pH 6.5 to 8.5) and under both oxidizing and reducing conditions. While all other oxyanion-forming elements are found within the μ g/L range, arsenic can be found within the mg/L range (Smedley and Kinniburg, 2002).

1.1. Health Effect of Arsenic

Arsenic (As) is a Class "A" human carcinogen as classified by the USEPA and its presence in the environment at specific concentrations, either as a dissolved contaminant in water, food web bioaccumulation, or as an inhaled particulate in the atmosphere, is of great concern to the well-being and health security of humans, with the situation being at its worst in Asia (Stroud et al., 2011; Marchiset-Ferlay et al., 2012). The ingestion of As into the human body takes place either by direct intake of arsenic rich water (via drinking or cooking foods) or by indirect intake through crops grown using arsenic-contaminated water.

Chronic exposure to low concentrations of arsenate can cause dermal changes like skin pigmentation; hyperkeratosis, cancer, melanosis, and ulceration (see Figure 3). Other chronic non - dermal effects of arsenic reported are diabetes (Weng-Ping, 2007), damage to the cardiovascular system (Hussam et al., 1991), hypertension, respiratory effects (Ann et al., 1996), adverse pregnancy outcome (Ann et al., 2005) and increased carcinogenic rates in humans at exposure to high concentrations of arsenic (World Health Organization, 2000). However, discussion is still on going about the threshold limit of carcinogenic action of arsenate, since both US and non-US based studies showed only a positive correlation between exposure to arsenic and the occurrence of cancer on arsenic concentrations above 100 μ g/g. Exposure to arsenic can also occur through food, water and air (Gorby et al., 1994). However, attention has been given to it since more than 90 % of arsenic in food is in form of organic arsenic. Water is the second major route of exposure to total arsenic and as it is mainly present in inorganic arsenic in natural water (Shakya et al., 1996).

1.2. Conventional Methods of Arsenic Removal

There are several methods of arsenic removal, which include ion exchange, coagulation precipitation, lime softening, membrane separation and adsorption. Some of these removal techniques are quite simple, but the main disadvantage associated with them is because of large amounts of toxic sludge they produce which need further treatment before disposal into the environment. Besides, these systems must be economically viable and socially acceptable activities (Salaún et al., 2007 and Villaescusa and Bollinger, 2008). The objective of this work was to synthesized an improved tailor-made material, which will meet several requirements such as; regeneration capability, easy availability, cost effectiveness, optimized masses and applied as adsorbents for uptake of arsenate. The aim is to use of solid-phase extraction cartridges for speciation purposes. These disposable cartridges are inexpensive and can potentially provide selective retention of arsenic species. As(V) is relatively easy to remove from water by using processes such as ion exchange

2. Experimental

2.1. Chemicals and Reagents used for the Experiments

polyvinylbenzylchloride (PVBC), N, NDimethylformamide (DMF), Tetrahyhdrofuran (THF), ethylenediamine (EDA), were purchased from Sigma Aldrich (Johannesburg, (South Africa), Hydrochloric acid, (Methyl, ethyl and propyl) iodide were purchased from merck chemicals and used as obtained.

2.2. Fabrication of PVBC

There are many solutions and process parameters that govern the outcome of morphology of nanofibers (Son et al., 2004; Li and Xia 2004]. Polymer solution parameters mainly include viscosity, conductivity and surface tension (Liang et al., 2007; Pillay et al., 2013; Li and Xia, 2004). One of the principal parameters that impact the process of electrospinning is viscosity of the polymer solution, which depends on polymer concentration and polymer type. The process of electrospinning is also based on the stretching of a charged jet of polymer solution (Greiner and Wendorff, 2007).

Different polymer concentrations of (20, 25, 30, 35, 40 and 45) %w/v of PVBC was dissolved in ratio 1:1 DMF: THF in a flask. Magnetic stirrer was in the flask because the weighted of polymer and the flask was fitted with a stopper to avoid evaporation of the solvent. The solution was stirred for 24 h, after complete dissolution, it was then transferred into a syringe. The positive electrode of a high voltage supply was applied to the metal spinneret (0.5 mm) tip of the syringe. The applied voltage used for the electrospinning was 15 kv, and the tip to collector distance was 13 cm. An electrically grounded, detachable, flat metal screen that is adjustable to a desired height and flat metal screen was used to collect electrospun nanofibers.

2.3. Functionalization/Quatenization of Polyvinyl benzyl chloride (PVBC)

2.15 g of PVBC nanofiber was cut out in circular portions of (1.5 cm) of the nanofiber sheet soaked in 8.0 g of ethylenediamine (EDA) in 15 mL of ethanol and shaken on a mechanical shaker for five days (Scheme 3.2) and the reaction in the vessel was allowed to proceed at 80 - 100°C for 36 hrs for the amination reaction. After the reaction, the modified fibers were removed from the solution and the fibers washed with methanol, Soxhlet extracted with methanol, and then air dried.

A mass of about 2.0 g each of the functionalized polyvinylbenzylchloride were prepared in different 100 mL reaction vessel. A 50mL of ethylenediamine was introduced into the 100-mL vessel containing 7 .0 g of methyl iodide. 4.0 g of lutidine and ethanol was added into 100 mL reaction vessel with a stirrer and was shaken on a mechanical shaker at 50°C to 60°C for 24 hrs. The quaternized fibre was filtered under vacuum and, washed extensively with water. The iodine was removed by washing the quaternized fibers with 100 mL of 0.1 M FeCl₃ in 6 M HCl, followed by using a hot 100 mL of 0.05 M Na₂S₂O₅ solution and finally 50 mL of conc. HCl was used to protonate the sites and the fiber was dried in a desiccator for 24 hrs.

2.4. FT-IR spectroscopy

The Fourier transform infrared (FT-IR) spectra of PVBC, functionalized PVBC quaternized PVBC and desorption of PVBC were obtained using a Perkin Elmer Spectrum 100 FT-IR spectrometer with an Auto IMAGE System.

2.5. Adsorption of Arsenate Using Solid Phase Extraction (SPE)

To optimize extraction, it is important to be aware of the nature of the sorbent used with respect to physicochemical characteristics. The most important physicochemical characteristics for optimal extraction is porosity and specific surface area.

The feasibility of using quaternized CH₃, C_2H_5 and C_3H_6 electrospun nanofibers as an SPE-sorbent material for the extraction of arsenate from wastewater is shown below. The nanofiber-sorbent beds were "packed" by cutting out circular portions (1.5 cm) of the nanofibers sheets from the aluminum foil into the SPE disk for extraction procedure for a flow through the process as shown in Figure 1



Figure 1: Shows the removal of arsenate using solid phase extraction (SPE)

SPE method was conducted to examine the adsorption isotherm effect of pH and the competitive anions on the adsorption of arsenic(v) from wastewater, using three different quaternized nanofibers (R': CH₃, C_2H_5 and C_3H_7). 5 mg each adsorbent material was chopped in a circular portion (1.5 cm) in the SPE disk. 100 mL of arsenate solutions were prepared in 250 mL flask. In the sorption experiments, and the solution pH was adjusted to 2.5 - 6.5 and was controlled throughout the filtration experiment.

After the sorption of arsenate, the fiber in the SPE disk was rinsed with Millipore water to remove the solution trapped in the disk and prepared it for other analysis. The initial and final arsenate concentrations in the solution from each of the collected flasks were determined using (ICP-OES).

2.6. Adsorption/Desorption Studies for Arsenate Removal Using Quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) Nanofibers

The effect of the functionalized (adsorbent) was observed on the uptake of arsenate present in the contaminated water sample. About 5 mg of the circular portions (1.5 cm) of the quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) nanofibres were added to 25 ml of arsenate solutions in and stirred for 30 min. The loaded nanofiber materials were filtered off, washed with Millipore ultra-pure water and dried in the desiccators using vacuum. The used dried fire was placed in 25 ml of 0.1 M NaOH solution and stirred for 30 min. in order to desorb the arsenate adsorbed on the nanofibers. The extent to which arsenates were enriched was determined using the ICP-OES analytical technique.

3. Results and Discussion

3.1. FTIR Studies of Unfunctionalized and Functionalized PVBC

The spectrum (A) of the PVBC can be assigned as follows: $_v$ (C-Cl) 670 cm⁻¹, CH (CH₃, C₂H₅ and C₃H₇) corresponds to the 2976 cm⁻¹, while 1451 cm⁻¹ spectrum represents the CH bending vibration of the same CH group in the alkyl groups. After the functionalization, the spectrum shows some significant changes. The $_v$ (C-Cl) at 670 cm⁻¹ disappeared completely to form a new band at peaks 1562 cm⁻¹ which can be assigned to N-H group of the amine group, while the peaks at between 1610 and 799 cm⁻¹ are due to C=O and C=N stretching groups. The presence of band at 3336 cm⁻¹ is due to bonded OH groups, which indicates the presence of water of crystallization.



Figure 2: FTIR absorption spectra for the unfunctionalized polyvinylbenzylchloride (A), functionalized PVBC (B) and the quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) (C, D and E) nanofibres. Moronkola BAet al 2016

3.2. FTIR showing the Desorption Studies of Functionalized PVBC ($R' = C_2H_5$ and C_3H_7) nanofibers using 0.1 M NaOH. The FTIR studies after desorption using 0.1 M NaOH in figure 2 shows some significant changes. The band at peak 1562 cm⁻¹ which can be assigned to N-H group of the amine disappeared completely in the FTIR spectra after desorption as shown in Figure 3



Figure 3: FTIR spectra for (A) and (B) shows before desorption studies, (C and D) after desorption using 0.1 M NaOH for the functionalized polyvinylbenzylchloride PVBC ($R' = C_2H_5$ and C_3H_7) nanofibres Moronkola BAet al 2016.



Figure 4: (e) SEM images (scale bar = $20 \ \mu m$) showing the morphology of fibers electrospun from PVBC solutions at concentration of $40 \ \% w/v$. (voltage of 15 Kv and at a constant spinning distance of 15 cm)Moronkola BAet al 2016.

The morphology of the functionalized PVBC nanofibers is shown by the SEM images presented in Figure 4. The functionalized PVBC nanofiber materials containing the amine (EDA) groups shows a slight difference with a change in morphology during functionalization and the fibers were not damaged. The fiber diameter range was 665-769 nm. This shows a slight increase in the fiber diameter after the functionalization with EDA Moronkola BA *et al* 2016.



Figure 5: (f) SEM images (scale bar = $20 \mu m$) showing the morphology of functionalized fiber with EDAMoronkola BA et al 2016.



Figure 6: SEM images (scale bar = $20 \ \mu m$) showing the morphology of nanofibers quaternized fiber with CH₃ and quaternized fiber with C₂H₅Moronkola BA et al 2016.



Figure 7: SEM images (scale bar = $20 \mu m$) showing the morphology of nanofibers quaternized with C_3H_7 Moronkola BA et al 2016.

The morphology of the sorbent materials (PVBC) changed after the functionalization with amine as shown in figures 5, 6 and 7 When quaternized after the functionalization with the alkyl groups (CH₃, C_2H_5 and C_3H_7) it was observed that the surface area of the sorbent materials was reduced when characterized with Brunauer, Emmett, Teller (BET) analysis from 341.3 m²/g to 243.3 m²/gMoronkola BA *et al* 2016.

3.3. The Effect of Initial Solution pH on the Removal of Arsenate using Quaternized PVBC Nanofibers from Wastewater. Operation Performed Thrice, N = 3.



Figure 8: Effect of initial solution pH on arsenate removal onto quaternary PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) nanofibres (n = 3). (Experiment conditions employed: initial As(v) concentration 10 µg/L, adsorbent dosage 5.0 mg/L, adsorption time 30 min, agitation speed 150 r/min).



Figure 9: Effect of final solution pH on arsenate removal onto quaternary PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) nanofibres (n = 3). (Experiment conditions employed: initial As(v) concentration 10 µg/L, adsorbent dosage 5.0 mg/L, adsorption time 30 min, agitation speed 150 r/min).

Figures 8 and 9 : Shows the optimum pH for arsenate removal was 5.0 for the three quaternized PVBC ($\mathbf{R}' = CH_3$, C_2H_5 and C_3H_7) nanofibres. The solutes interact with mineral surfaces due to their electrical surface charge and because of the ionic interactions involving the functional groups (NH_4^+ , H^+ and OH^-) on the surface of nanofibres ($\mathbf{R}' = CH_3$, C_2H_5 and C_3H_7 quaternized PVBC) as well as ions in the solution. The adsorption amount for arsenate was 99.7 %, 97.31 % and 95.47 % respectively for the quaternized PVBC ($\mathbf{R}' = CH_3$, C_2H_5 and C_3H_7) were obtained. The solution pH does not only affect the charge property on the quaternized PVBC surface through the protonation of the amine groups, but also influenced arsenate speciation in solution.

Arsenate exists in the surface of water as H_3AsO_4 , $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-} in aqueous solution. $H_2AsO_4^-$ is the main species in solution at pH 3 to 6, while $HAsO_4^{2-}$ and AsO_4^{3-} becomes the major species at the pH range studied. The electrostatic attraction plays

an important role in the sorption process. The amine groups were first protonated in the acidic medium of the solution and thus, adsorbed more of negative arsenate which means that more of the arsenate groups were found at lower pH.

When the solution pH was increased, the number of protonated amine groups on sorbent materials appeared to have been dequarernized due to N-H group of the amine group on the sorbent material at a higher pH. As a consequence, the sorption amount on the quaternized PVBC sorbent material for arsenate decreased as the pH increases (Shubo et al., 2008; Ismail, 2012).

3.4. Effect of Adsorbent Dose on Arsenate Removal onto quaternized PVBC Nanofibers

The criteria for selection of suitable adsorbent materials include: the cost of the medium, the ease of operation or handling the cost of operation, the useful service life per cycle the adsorption capacity of the adsorbent, the potential of reuse and the possibilities of regeneration of adsorbent.

Figures 10, 11 and 12 shows the effect of adsorbent dose on percentage removal of As(v). The figure reveals that, uptake of As(v) increases rapidly from 2 to 5 g/l, and marginally thereafter due to more surface area, with increase in adsorbent dose (Gupta et al., 2005). The sorption curve is a typical sigmoidal curve that indicates an increase in sorption as the sorbent dose is increased. The curve beveled off, indicating exhaustion of sorption reaction through a good amount of sorbent surfaces available to the sorbate – solvent interaction.



Figure 10: Effect of arsenate concentration and adsorbent dosage on arsenate removal onto quaternary PVBC R' CH_3 (n = 3). (Experiment conditions employed: initial As(v) concentration10 $\mu g/L$, solution pH 5.0, adsorption time 30 min.



Figure 11: Effect of arsenate concentration and adsorbent dosage on arsenate removal onto quaternized PVBC ($R' = C_2H_5$) (n = 3). (Experiment conditions employed: initial As(v) concentration10 μ g/L, solution pH 5.0, adsorption time 30 min.



Figure 12: Effect of adsorbent dosage on the adsorption of As (V) removal onto quaternary PVBC R' C_3H_7 (n = 3). (Experiment conditions employed: initial As(v) concentration 10 μ g/L, solution pH 5.0, adsorption time 30 min.

3.5. Adsorption Kinetics of Arsenate Removals onto Quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) (n = 3)

As noted above, the adsorption kinetics plays an important role in the efficiency of arsenate removal from a solution. The sorbtive uptake of arsenate by quaternized ammonium nanofiber from water at a solution pH of 5.0 shows a graphic representation of percent of arsenate removal with contact time. In Figure 12 below, the nanofiber quaternized with methyl group is the highest at 99.2 %, followed by ethyl and propyl groups at 97.92 % and 97.01 % respectively. Almost 100% removal occurred within 15 min for the methyl group and between 93% and 97 % was removed in 40 min for ethyl and propyl groups respectively. The percent sorption of sorbate as a function of contact time indicates a triphasic pattern, with an initial step portion, indicating a high rate of uptake. The rate of uptake was constant, which is represented by Figure 12. The high uptake of arsenate was due to the diffusion of arsenate from the bulk of the solution onto the surface of the sorbent, which is described as instantaneous sorption.



Figure 13: Adsorption kinetics of arsenate removals onto quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) (n = 3). (Experiment conditions employed: initial As(V) concentration 10 µg/L, adsorbent dosage 5.0 mg/L, solution pH 5.0.

Sorbent material	Anions	% of As(v) adsorbed
quaternized PVBC	Blank, AsO_4^{3-} , SO_4^{2-} , Cl^{-}	99.97, 97.95, 92.21 89.89, 80.19 and 78.42
$(R' = CH_3)$	and NO ₂	
quaternized PVBC	Blank, AsO_4^{3-} , SO_4^{2-} , Cl^{-}	97.69, 95.01, 86.71 86.10, 76.88 and 74.95
$(\mathbf{R}' = \mathbf{C}_2\mathbf{H}_5)$	and NO ₂	
quaternized PVBC	Blank, AsO_4^{3-} , SO_4^{2-} , Cl^{-}	80.44, 79.00, 66.99 61.22, 60.63 and 57.47
$(\mathbf{R'} = \mathbf{C}_3\mathbf{H}_7)$	and NO ₂ ⁻	

Table 1.0: Effect of co-existing anions on the removal of arsenate onto quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7). (Experimental condition10 µg/L of arsenate and 30 µg/l of co-existing anions dosage 12 mg/L,) (n = 3).

3.6. Desorption Studies of Arsenate onto Quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) Nanofibers (n = 3)

Desorption experiments were carried out to investigate the feasibility of regeneration of the used quaternized PVBC ($R' = CH_3$, C_2H_5 and C_3H_7) adsorbent, after As(v) adsorption.

Figures 13, 14, 15 show the desorption efficiency of As(v) with 40 min desorption time. The recovery efficiencies of As(v), using the three different sorbent materials.

The arsenate sorption capacity of the sorbent material; quaternized PVBC ($\mathbf{R}' = CH_3$, C_2H_5 and C_3H_7) nanofibers which can be used four times. These results agree with the work of Gillman, 2006 and Chiban et al., 2009 that NaOH solution could be used as solvent to regenerate As(v) from the adsorbent materials.



Figure 13: Reusability cycles for arsenate onto quaternized PVBC ($R' = CH_3$) nanofibres pH = 5.0 (Experimental condition 10 µg/L of arsenate adsorbent dosage 5 mg/L (n = 6).

Figures 14 and 15 below are largely determined by the efficiencies of their recovery and the extent to which the adsorbed arsenate are desorbed. There was no significant change in the efficiencies of adsorption and desorption up to the 4th time of use. The decline in efficiencies after the 4th usage of the quaternized PVBC (R' C_2H_5 and R' C_3H_7) nanofibers could be attributed to the losses of amount of the sorbent during usage.



Figure 14: Reusability cycles for arsenate removal onto quaternized PVBC (R' C_2H_5) pH = 5.0 (Experimental condition10 µg/L of arsenate adsorbent dosage 5 mg/L (n = 6).



Figure 15: Reusability cycles of arsenate removal onto quaternized PVBC R' (C_3H_7) pH = 5.0. (Experimental condition10 µg/L of arsenate adsorbent dosage 5 mg/L agitation speed 150 r/min) (n = 6)

4. Conclusion

Arsenic in wastewater is major problems that people are facing worldwide. Arsenic removal from drinking water has become mandatory for many wastewater systems after the U.S. EPA promulgation of the 10 μ g/L final arsenic rule in 2006. This rule was designed to save human lives by reducing the degree of prolonged arsenic exposure, thereby reducing the risk of both internal and skin cancers. The arsenic adsorption capacities of low cost adsorbents presented vary, depending on the characteristics of the individual adsorbent, concentration of arsenic, pH, contact time effect of co-existing anions and reusability.

The solid phase extraction (SPE) experimental results show that, the SPE sorption for arsenate using PVBC $R' = CH_3$ was very fast as well as pH- dependent. The sorption amount of PVBC $R' = CH_3$ for the equilibrium of arsenate concentration using SPE was reduced from 50 µg/l to 10 µg/l within 30 min because of the positive charge on the nitrogen PVBC $R' = CH_3$, which had proved to be better than other conventional adsorbents used for the removal of this toxic anions from wastewater.

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