GENERAL ASPECTS OF THE COAGULATION PROCESS FOR THE REMOVAL OF ARSENIC FROM SIMULATED GROUNDWATER

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Abstract
Removal of As from the simulated groundwater contaminated with arsenic by coagulation process was examined in the present study using Jar-test procedure. Aluminium sulfate, polyaluminium chloride, ferric chloride and ferric sulfate were comparatively tested as coagulants, in order to investigate their performance on arsenic removal.
The experimental study results have showed that all the coagulants tested are found to be effective in removing As from simulated groundwater with initial As concentration of 20 μg L^{-1}, the best results being obtained with ferric chloride.

Introduction
Providing drinking water with safe arsenic levels in the West Region of Romania is a current challenge, due to arsenic's presence in some specific groundwater sources.
Low-cost, easy to use, efficient, and sustainable solutions are needed to supply arsenic safewater to the rural and peri-urban population in the affected areas [1,7].
Various technologies are available for the removal of arsenic from contaminated groundwater using physicochemical methods including oxidation, chemical precipitation or coagulation-flocculation, adsorption, lime softening, ion exchange, and membrane separation processes like reverse osmosis, nanofiltration and electrodialysis [2,3,4].
Coagulation is a simple method for As removal process from contaminated groundwater and most used coagulants are aluminum salts such as aluminum sulfate [$Al_2(SO_4)_3.18H_2O$] and ferric salts such as ferric chloride [$FeCl_3$] or ferric sulfate [$Fe_2(SO_4)_3.7H_2O$]. Ferric salts have been found to be more effective than alum removing As on a weight basis and effective over a wider pH range. With low cost and relative ease of handling in arsenic removal by this process, the coagulants transform dissolved arsenic into an insoluble solid which is precipitated later. Dissolved arsenic may also be adsorbed on the solid hydroxide surface site and be coprecipitated with other precipitating species [4] and the solids can be removed through sedimentation and/or filtration. Optimized operating coagulants doses needed are dependent on the matrix quality of the groundwater source.
Groundwater monitoring is nowadays mandatory in all member state level in European Union [6,7] and our previous studies was focused to act in accordance with the legal provisions of this directive adopted. After an adequate conventional monitoring in groundwater, in the Western Region of Romania, in the catchment areas for abstraction points, or in groundwater sources, of some relevant parameters, substances or pollutants, we concluded that the groundwater sources from the West Region of Romania that require water treatment, are generally characterized by high concentrations of iron and manganese, also arsenic being present in fairly high concentrations and in certain situations exceedances may occur for ammonium [5].

Based on these results, the present study investigates the treatment of a simulated groundwater contaminated with arsenic (20 µg L^{-1}) by using a coagulation process, using Jar-test procedure and aluminium sulfate, polyaluminium chloride, ferric chloride and ferric sulfate as coagulants.

Experimental

Coagulants and water samples

For comparison purposes, commercially available PAC (>19% Al_2O_3, basicity >80,0%, density >1.20 kg/dm^3) was a UNICHEM product (Hungary). Alum stock solution was prepared from liquid aluminium sulphate (approximately 339 g/L $Al_2(SO_4)_3.18H_2O$) obtained from a local Bega water treatment plant. $FeCl_3$ stock solution (approximately 40%) and $Fe_2(SO_4)_3$ stock solution (approximately 40%) were prepared by adding commercial $FeCl_3$ product (Chimcomplex S.A. Borzesti, Romania), respectively $Fe_2(SO_4)_3.7H_2O$ (Kemwater Cristal, Romania).

The simulated groundwater chemistry composition was given by 4 simulated pollutants added: arsenic (20 µg L^{-1}), iron (0,4 mg L^{-1}), manganese (0,2 mg L^{-1}), and ammonium (0,5 mg L^{-1}), into deionized water. After that, this simulated groundwater was stirred for 5 min., at 300 rpm.

Experimental and analytical methods

Coagulation experiments were carried out at room temperature using Jar-test on a six-paddle gang stirrer (Floculator Jar-test, WiteStir JT M6). The 800mL simulated groundwater was added into the 1000mL beaker. A measured amount of coagulant was added by a calibrated pipette (Multipette stream Electronic hand dispenser, Eppendorf, Germany) into the working simulated groundwater under rapid stirring. The simulated groundwater was stirred rapidly at 150 rpm for 2 min after coagulant dosing, followed by slow stirring at 45 rpm for 10 min.

For 20 min after settling, supernatants were collected to measure residual turbidity using a Turbidimeter (HI 88713, HANNA Instruments). Total organic carbon (TOC) was analyzed after filtration through a 0.45 mm membrane and were determined using a TOC Analyzer (Multi N/C 2100 S, Analytik Jena). pH and conductivity were determined on a laboratory pH-meter (Thermo Orion, Cole-Parmer) and conductometer (Starter 3100C, Ohaus). Colour in Hazen units and residual aluminium was measuring using a photometer (Move 100, Merck). The absorbance at 254 nm (due to the Natural Organic Matter/NOM content) was measured with a Spectrofotometer UV-VIS (Specord 205, Analytik Jena), using a 1 cm path length quartz cuvette. The detection technique used for As was Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES Avio 500, Perkin-Elmer) and the concentration of iron and manganese were determinated with a Spectrofotometer (sAA-280FS, Agilent).

Results and discussion

The comparison of the efficiency of the coagulation-flocculation process by using the 4 types of coagulants (selected according to the data from the specialized literature) was carried out by the Jar-test method, in 2 situations, respectively without and with pre-oxidation with sodium hypochlorite applied as oxidant before coagulation.

Sodium hypochlorite (concentration 14.6%) was added to each sample before the introduction of coagulants, pre-oxidation which was practically carried out under continuous stirring at a speed of 600 rpm for 2 minutes.

Added doses (similarly established with documented literature data) are specified on each work set in Tables 1,2.

Coagulants were added under rapid stirring simultaneously to all samples using the automatic dosing pipette. Three sets of analysis were performed for each type of coagulant.

Table 1: Comparative analysis for quality parameters of AS1 (simulated groundwater) treated with coagulants SA, PAC, $FeCl_3$, $Fe_2(SO_4)_3$.

Parameter, unit	AS1	SA	PAC	FeCl₃	Fe₂(SO₄)₃
Dose, mg/L	-	10	10	10	10
Turbidity, NTU	0,79	0,48	0,24	0,84	0,316
pH	7,54	7,24	7,16	6,206	6,239
Temperature, °C	22	22	22	22	22
Color, grd. Hz	<25	38	<25	32	27
Conductivity, μs/cm	258	228	234	247	249
MTS, mg/L	<2	8,1	11,5	34,8	26,4
UV_{254nm}, cm^{-1}	0,050	0,031	0,023	0,028	0,051
Aluminium dissolved, μg/L	-	<20	<20	-	-
Iron dissolved, mg/L	0,40	0,32	0,268	0,293	0,123
Manganese dissolved, mg/L	0,20	0,20	0,20	0,20	0,20
Amonium, mg/L	0,50	0,50	0,50	0,50	0,50
Arsen, μg/L	20	<2	<2	<2	<2

The application of preoxidation with sodium hypochlorite (doses of 0.1 mg/L NaOCl) also significantly improved the degree of reduction of iron, manganese and ammonium ions (table 2).

Table 2: Comparative analysis for quality parameters of AS1 (simulated groundwater)

after peroxidation with 0.1 mg/L NaOCl and treated with coagulants SA, PAC, FeCl₃, Fe₂(SO₄)₃.

Parameter, unit	AS1	SA	PAC	FeCl₃	Fe₂(SO₄)₃
Dose, mg/L	-	10	10	10	10
Turbidity, NTU	0,79	0,32	0,21	0,54	0,286
pH	7,54	6,58	6,75	6,17	5,99
Temperature, °C	22	22	22	22	22
Color, grd. Hz	<25	37	34	30	28
Conductivity, μs/cm	258	266	283	293	298
MTS, mg/L	<2	7,3	9,9	38,9	21,8
UV_{254nm}, cm^{-1}	0,050	0,040	0,032	0,031	0,067

Aluminium dissolved, µg/L	-	33	<20	-	-
Iron dissolved, mg/L	0,40	0,27	0,218	0,18	0,178
Manganese dissolved, mg/L	0,20	0,18	0,16	0,176	0,18
Amonium, mg/L	0,50	<0,028	0,043	0,060	0,040
Arsen, µg/L	20	<2	<2	<2	<2

Conclusion

Concluding this study results:

- Differences were observed between the performances of arsenic removal for the four coagulants compared, ferric chloride proven to be the most effective coagulant.

- However, the degrees of reduction to match the quality of the coagulated and then decanted water within the norms required by the drinking water standards were not achieved, even for iron chloride coagulant applied to treat the simulated selected chemistry matrix for groundwater, with similarity into part of the underground aquifer in the West region of Romania.

- Continue research are needed in finding the best available and cost-effective treatment process to remove As and its co-contaminants, for As-free safe and healthy drinking water.

Acknowledgements

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