REMOVAL OF TRICHLOROETHYLENE FROM WATER BY BIOLOGICAL AND ULTRASONIC PROCESSES

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This paper emphasized the new approach of groundwater drinking water source recovery. Experimental tests were focused to remove trichloroethylene from water. Many countries have underground water polluted with TCE and Romania has some. The treatment proposed flow is based on biological treatment: biological granular activated sludge followed by sludge separation (settling/filtration) and ultrasonication (sonolysis) of the liquid separated phase. Laboratory experimental tests showed that removal efficiencies of TCE had the optimal conditions: biological sludge dose 360 mg/L, two hours reaction time, five minutes of ultrasonication (20 kHz, 5000 kJ energy) for 0.262 mg TCE/L initial concentration (removal yield was 97%).

Keywords: trichloroethylene, TCE, sonolysis, granular biological sludge

1. Introduction

Physical-chemical and biological methods are usually used for removal of organochlorinated compounds from wastewater. Only few experimental studies were dedicated to removal of these compounds from drinking water sources by ultrasonic and biological processes in the same drinking water treatment flow.

The ultrasonic method is a member of AOPs water treatment category which is successfully applied in case of micropollutants removal. Ultrasonic cavitation phenomena are the base of organic compounds sonolysis. Similar to other advanced oxidation processes (AOPs), degradation of organic micropollutants take place by radical mechanism (hydroxylic radical is the most efficient) [1].

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Trichloroethylene (TCE) is a toxic and carcinogenic compound which is constant detected in atmosphere, soil and underground water in many regions of the world because of inadequate disposal method in many countries. This aliphatic organochlorinated compound is heavier than water (1.46 g/cm^3) and can penetrate the soil until groundwater level where could generate pollution plumes and contamination of groundwater for a long time in that area and beyond. Romania has some areas even in the proximity of Bucharest. The maximal admitted concentration level in drinking water is 10 μg/L [2].

Remediation of TCE polluted groundwater still generates new technologies on environmental market because of interest to recover new water sources. It is well known that TCE is hard to be oxidize even with advanced oxidation processes (e.g. Fenton oxidation).

Ultrasonic field degrades TCE not only because of hydroxyl radical generation during the water sonolysis but also because the high levels of temperature and pressure inside the microbubbles generated by ultrasonic cavitation [3].

Experimental studies have indicated that ultrasonic pretreatment processes lead to improved biodegradability in conventional aerobic or anaerobic oxidation by increasing the speed and efficiency of the treatment.

Biological methods are known as the most common and cost-effective choices of the treatment. Membranes biorector are also aerobic technologies wich have ben used alone or in combination with aerobic activated sludge process to treat industrial wastewater.

In the last years, in other countries, biological processes were applied in the field of drinking water treatment in special for ammonia removal. In Romania there are some studies on pilot laboratory scale but no application for ammonia or organochlorinated removal from drinking water[4]. Biological treatment step could be done by three principal mechanisms: reductive dechlorination (anaerobic process: *Dehalococcoides ethenogenes, Dehalospirillum multivorans, Enterobacter agglomerans spp*.), co-metabolization (anaerobic process: Pseudomonas putida, Pseudomonas mendocina, Pseudomonas cepacia spp.), direct oxidation (anaerobic or aerobic process).

A hybrid TCE removal path can be biological treatment + sonolysis which could be efficient for advanced removal of specific micropollutant and for disinfection (with no additional pollution as trihalomethanes, specific for classical chlorination of drinking water) [5]. This paper shows laboratory scale experimental results of TCE removal from water by biological process, based on granular activated sludge, followed by ultrasonication.

2. Experimental

The experimental tests were performed in order to establish the optimal conditions for TCE removal from synthetic aqueous solution during the following water treatment scheme:

PHASE 1 -Biological treatment with granulated activated sludge
PHASE 2 - Separation of biological sludge (settling + filtration)
PHASE 3 - Sonolysis of filtered water

The main goal was to determine the optimal process parameters for PHASE 1 and the best TCE removal both for this treatment stage and for all treatment schemes. The optimal sonolysis parameters were considered to be 20 kHz frequency, 5 minutes ultrasonic treatment at 5000 kJ ultrasonic energy and 100% amplitude, based on previously experimental tests.

Experimental tests for the biological treatment phase had the following laboratory procedure:

- ***preparation of biological activated sludge*** from the stock suspension of biological aerated sludge: sampling of 100 mL sludge, settling 20 min., washing of separated wet solid phase (three times with 100 ml distilled water followed each time by 10 min. settling, sieving of the final washed sludge - 1 mm mesh);
- ***experimental batch tests***: 250 mL water sample with TCE content (0.73÷6.78 mg/L) and biological sludge (25.2÷378 mg/L with 3990÷6650 mg/l d.w. - dry substance content), magnetically stirring keeping ~50 mL of air above the reaction mixture; TCE concentrations levels were established by GC-MS method (Head Space);
- ***separation of biological granular sludge*** (10 min settling, 22 μm filtration membrane).

Liquid phases were analyzed for chemical-physical (pH, TCE) and microbiological indicators (total coliforms and enterococci).

A blank sample (without biological sludge add) were done for each batch test in order to find the influence of TCE removal only by aeration (aeration efficiency is inside the tables content).

First experimental series established the influence of reaction/contact time, biological sludge dose and initial TCE concentration for biological treatment step.

The final tests emphasized TCE removal efficiency for entire treatment flow PHASE 1-3 (total efficiency and specific efficiencies) for three initial TCE concentrations: 0.262 mg/L, 0.530 mg/L, 4.607 mg/L, two hours biological treatment for the same biological sludge dose (360 mg/L) and five minutes of ultrasonication. Microbiological analysis was performed (total coliforms, enterococcus).

3. Results and discussions

The experimental tests had the following main results:

3.1 The influence of batch test reaction time

The reaction time of biological treatment tests was in the range of 1-7 hours, biological sludge dose 80 mg/L (3990 mg/L d.w.) and TCE initial content 1.62 mg/L. Table 1 shows the evolution of TCE residual content vs. reaction time.

Table 1

The influence of reaction time for TCE removal efficiency

Sample	Contact time hours	TCE initial mg/L	TCE residual mg/L	η TCE aeration %	η TCE aeration + biological treatment %	η TCE biological treatment %
RT1	1	1.62	1.295	5.9	20.1	14.2
RT2	2		0.906	8.2	44.1	35.9
RT3	3		0.730	21.3	54.9	33.6
RT4	4		0.601	34.5	62.9	28.4
RT5	5		0.526	44.5	67.5	23.0
RT6	6		0.393	53.9	75.7	21.8
RT7	7		0.257	62.6	84.1	21.5

The increasing of reaction time leads to the following observations:

- residual TCE concentrations had a linear decreasing from 1.29 mg/L (1h reaction time) to 0.26 mg/L (7 h reaction time);
- total TCE removal efficiency had the same linear decreasing, max. 84% (sample RT7/7 h reaction time);
- the efficiency of biological treatment step didn't have a linear decreasing 36% being maximum value (sample RT2, 2 h reaction time); minimum 14.2% after one hour;
- the aeration efficiency had a constant increasing and after 4 h reaction time was over the biological TCE removal efficiency (sample RT4 34.5%>28.4%); this effect has a negative impact both to economic impact because longer reaction times mean higher energy consumption and environment because of TCE transfer into atmosphere.

3.2. The influence of biological sludge dose

The amount of granular biological sludge which were used in batch tests was 25.2-378 mg/L as dry substance (dry substance d.w. = 6305 mg/l). Table 1 shows the evolution of TCE residual content vs. biological sludge dose for the same reaction time (2 h).

Table 2

The influence of biological sludge dose for TCE removal efficiency

Sample	Biological sludge dose mg/L	TCE initial mg/L	TCE residual mg/L	η TCE aeration %	η TCE aeration + biological treatment %	η TCE biological treatment %
BS1	25.2	5.95	2.772	29.8	53.4	23.6
BS2	75.6		2.761		53.6	23.8
BS3	126		2.549		57.2	27.4
BS4	252		2.441		59.0	29.2
BS5	378		2.115		64.5	34.7

The main experimental observation regarding the influence of biological sludge amount are:

- the variation domain of residual TCE concentrations were 2.11-2.77 mg/L, very tight, and maximum removal efficiency was 64,5% with 34.7% efficiency of biological treatment;
- there is no need to use over 380 mg/L biological sludge because of organic and microbiological load increasing with advantage (additional costs and technological complications for filtration and ultrasonication steps).

3.3 The influence of TCE concentration

The influence of TCE initial concentration was studied keeping constantly reaction time = 2 h and biological sludge dose = 353 mg/L.

Table 3

The influence of TCE concentration

Sample	TCE initial mg/L	TCE residual mg/L	η TCE aeration %	η TCE aeration + biological treatment %	η TCE biological treatment %
C1	0.73	0.40	19.1	45.2	26.1
C2	2.00	1.02	29.5	49.0	19.5
C3	3.62	1.83	30.4	49.4	19.0
C4	6.78	3.70	28.0	45.4	17.4

The removal efficiencies of TCE were in the range of 45-49% for initial concentration of 0.73-6.78 mg TCE/L. Lower TCE initial concentration leads to a higher biological treatment efficiency (26.1%, sample C1). An additional sample similar with C1 with 4 h reaction time was tested but removal efficiency of TCE was the same. This behavior shows the limits of biological treatment because of at least two reasons: biological degradation is slower than chemical reactions and the aeration which is able to transfer this volatile chlorinated compound into

atmosphere. From these reasons, some studies are focused on anaerobically biological degradation of TCE with specific advantages and disadvantages.

Ultrasonic treatment step is able to combine de performances of biological and physical-chemical processes in order to have better TCE removal efficiencies for higher and lower initial concentrations.

3.4 Combined biological and ultrasonic processes for TCE removal

The optimal experimental conditions were selected based on laboratory tests presented above (biological treatment phase) and previously experimental work in case of ultrasonic treatment: pH~7, biological sludge dose 360 mg/L, biological treatment time 2 h, 5 minutes ultrasonic treatment (20 kHz, 5000 kJ energy). Table 4 and figure 1 show the TCE removal in these experimental conditions for three initial TCE content.

Table 4

The efficiency of TCE removal for the selected treatment flow

Sample	Process	TCE initial mg/L	TCE residual mg/L	η TCE treatment %	η TCE total %
T1	Aeration (blank)	4.607	3.223	30	88
	Biological		1.287	30	
	Sonolysis		0.553	57	
T2	Aeration (blank)	0.530	0.395	25	87
	Biological		0.254	35	
	Sonolysis		0.064	75	
T3	Aeration (blank)	0.262	0.209	20	97
	Biological		0.120	43	
	Sonolysis		0.007	94	

The main experimental observations were as following:

- pH of the final effluent 6.7-7 in the admitted domain
- the minimal residual TCE concentration was 0.007 mg/L (sample T3) below admitted limit (0.010 mg/L); removal TCE efficiencies were: total 97%, biological treatment phase 43%, ultrasonic treatment step 94%;
- the efficiency of ultrasonic treatment increases with decreasing of initial TCE concentration but in case of biological phase 75% was the higher value in case of sample T1 (4.6 mg TCE/L initial concentration);
- the microbiological load was over the admitted limits after biological treatment (total coliforms 9400 FCU/100 mL,

enterococcus 8 FCU/100 mL) but decreases after ultrasonic treatment (total coliforms 7200 FCU/100 mL, enterococcus 4 FCU/100 mL).

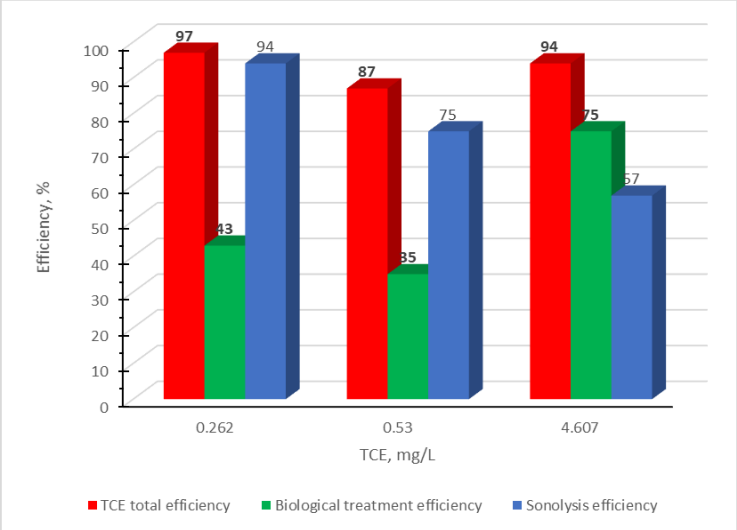

Fig. 1. TCE removal efficiency: total, biological treatment - phase 1, ultrasonic treatment - phase 3

The overtaken of microbiological concentration limits can be corrected by additional ultrasonication or by chlorination (recommended because of residual chlorine in drinking water).

4. Conclusions

This paper shows the experimental results of TCE removal from aqueous solution by biological treatment with granular activated sludge and sonolyse in order to find a new hybrid treatment of groundwater polluted with this micropollutant.

The main conclusions of experimental tests were;

– the best TCE removal efficiencies in the phase of biological treatment were the followings: biological sludge dose 380 mg/L, two hours reaction time, 6 mg TCE/L initial concentration;
– the removal efficiencies of TCE along the treatment flow (biological treatment, phase separation, sonolysis) had the optimal conditions: biological sludge dose 360 mg/L, two hours reaction time, five minutes of ultrasonication (20 kHz, 5000 kJ energy) 0.262 mg TCE/L initial concentration;
– these operational conditions can remove TCE from water below admitted limit for drinking water but request a new additional treatment step:

chlorination in order to remove residual microorganisms content and to assure residual chlorine amount according with drinking water quality regulations.

R E F E R E N C E S

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