Co-reduction of nitrate and perchlorate in a pressurized hydrogenotrophic reactor with complete H₂ utilization

Razi Epsztein *, Chaitanyakumar Desitti, Michael Beliavski, Sheldon Tarre, Michal Green

Faculty of Civil and Environmental Engineering, Technion – Israel Institute of Technology, Haifa 32000, Israel

**HIGHLIGHTS**

- A two-stage hydrogenotrophic process to remove NO₃⁻ and ClO₄⁻ is presented.
- NO₃⁻ is removed in a first unsaturated-flow pressurized reactor stage.
- The residual H₂ is coupled to ClO₄⁻ reduction in a second polishing stage.
- Large presence of Dechloromonas was detected before and after ClO₄⁻ addition.
- Effluent ClO₄⁻ concentration of 2 μg/L and ~100% H₂ utilization were achieved.

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**ABSTRACT**

A novel pressurized hydrogenotrophic reactor operating at high rates was recently developed specifically for the removal of nitrate (NO₃⁻) from drinking water. The reactor is characterized by safe and economical operation since hydrogen (H₂) purging intrinsic to conventional H₂-based denitrifying systems is not required and H₂ loss occurs only through the effluent, resulting in H₂ utilization efficiency above 90%.

In this research, a new treatment scheme to remove NO₃⁻ and perchlorate (ClO₄⁻) combining the pressurized reactor with a following open-to-atmosphere polishing unit is presented. In the pressurized reactor, NO₃⁻ and ClO₄⁻ are simultaneously removed. In the polishing unit, the residual dissolved H₂ from the pressurized reactor serves to further reduce ClO₄⁻ to trace concentrations below recommended levels.

First, ClO₄⁻ reduction together with denitrification was demonstrated in the pressurized reactor without special inoculation and a maximal ClO₄⁻ volumetric removal rate of 1.83 g/(L·d) was achieved. Microbial population analyses before and after the addition of ClO₄⁻ were similar with a large fraction of the genus Dechloromonas. Results show that the combined treatment scheme consisting of the pressurized reactor and the polishing unit allowed for the reduction of ClO₄⁻ concentration down to a minimal value of 2 μg/L with a simultaneous increase of the H₂ utilization efficiency from 95% up to almost 100%.

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**1. Introduction**

Intensive use of nitrogen-based fertilizers and wastes from rocket facilities are the main sources for groundwater contamination by nitrate (NO₃⁻) and perchlorate (ClO₄⁻), respectively [1,2]. Despite the different pollution source, co-occurrence of both ions is common, especially in groundwater close to military bases that house rockets [2]. In some cases, the high ClO₄⁻ concentration in the discharge point can lead to migration of ClO₄⁻ in groundwater far away from the focus of pollution and mixing with NO₃⁻-contaminated groundwater, as reported in the Ramat Hasharon area in Israel [3]. High ClO₄⁻ levels were also detected in groundwater throughout the U.S., mainly in California, Nevada, Utah, Arizona and other states where rocket and missile production occurs [4]. Also, ammonium perchlorate (NH₃ClO₄) occurs naturally in NO₃⁻ deposits that are used in some fertilizers [5]. In California, for example, drinking water sources that contain ClO₄⁻ was found to have much higher concentrations of NO₃⁻ than wells with no measurable ClO₄⁻ [6].

The World Health Organization (WHO) standard for NO₃⁻-N is 11.3 mg/L (as nitrogen) [7]. As for ClO₄⁻ standards, are more variable and location-dependent. The Environmental Protection Agency (EPA), for example, established an advisory standard of 15 μg/L, but numerous states in the U.S. promulgated enforceable standards for ClO₄⁻ in drinking water of only 2 μg/L [2]. Therefore, a comprehensive solution for meeting the drinking water standards determined by the health organizations is required. In the

* Corresponding author.
E-mail address: raziepsztein@yale.edu (R. Epsztein).

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case of ClO₄⁻, reduction to trace concentrations is needed and therefore much harder to achieve by any treatment technique [8]. Biological denitrification and biological ClO₄⁻ reduction are two processes proved to efficiently reduce NO₃⁻ and ClO₄⁻ concentrations to the permitted thresholds without the production of waste brine [7,9]. Moreover, many perchlorate-reducing bacteria can grow also on NO₃⁻ and therefore a biological denitrification system may be (but not necessarily) effective for ClO₄⁻ reduction [10]. Using H₂ gas as the electron donor for bacterial growth in both processes is advantageous over the common organic donors (mostly methanol, ethanol and acetate) for drinking water treatment, mainly due to the lower cell yield of autotrophic bacteria reducing methanol, ethanol and acetate) for drinking water treatment, to prevent a possible regrowth of biomass in the distribution system and ensure a safe drinking product. In the following research work, the simultaneous removal of ClO₄⁻ and NO₃⁻ was investigated in a modified version of the above reactor, i.e., a combined treatment scheme. The combined treatment scheme (Fig. 1) combines the unsaturated-flow pressurized reactor with an up-flow submerged open-to-atmosphere polishing unit. The polishing unit aims to increase H₂ utilization (~100%) by the consumption of the residual dissolved H₂ from the pressurized reactor and further reduction of ClO₄⁻ to trace concentrations below recommended levels (between 1 and 15 μg/L).

2. Materials and methods

2.1. Experimental setup

A schematic diagram of the combined treatment scheme is illustrated in Fig. 1. The combined treatment scheme included the unsaturated-flow pressurized reactor, i.e., the main reactor unit, combined with a submerged open-to-atmosphere polishing unit to reduce ClO₄⁻ by the residual dissolved H₂ in the effluent of the main reactor unit. A detailed description of the main reactor unit was given in an earlier publication [24]. Briefly, it comprised of a clear PVC cylindrical reactor 70 cm in height and 10.5 cm in diameter divided into three unequal parts. The top part of the reactor (height 20 cm) served as an empty headspace, the middle part (height 30 cm) was filled with plastic biofilm carriers (total surface area of 900 m²/m³, Aqwise) and separated by a metal screen from the bottom part (height 20 cm) of the reactor where recirculating
water collected. Hydrogen gas was supplied continuously from H<sub>2</sub> cylinder. The reactor was connected to a feed pump (Diaphragm pump model 7090-42, Cole-Palmer, USA), recirculating pump (FL-2403, ProPumps, China) and pH controlling unit (standard pH electrode, pH controller – Alpha 190, Eutech, Singapore; hydrochloric acid tank and acid pump – gamma/L, ProMinent, Germany). The main reactor unit was operated as a trickling filter with water recirculation. It was continuously fed with simulated NO<sub>3</sub> and ClO<sub>4</sub> contaminated groundwater. An automatic drain valve discharged accumulated water to the polishing unit.

The polishing unit comprised of a PVC cylindrical polishing unit 25 cm in height and 10.5 cm in diameter, filled with the same plastic biofilm carriers as in the main reactor unit. The effluent water from the main reactor unit was introduced at the bottom of the polishing unit and released at the top part. The polishing unit was operated under a saturated-flow mode (i.e. submerged unit) and its discharge was open to the atmosphere.

Reactor start-up and initial investigation of ClO<sub>4</sub> reduction (Sections 3.1) were performed in the main reactor unit only, using the same biomass carriers from previous denitrification experiments [24]. Start-up of the polishing unit in the following trials was performed by filling the polishing unit with additional clean carriers mixed with biomass carriers from the main reactor unit (the biomass carriers taken from the main reactor unit were replaced with new carriers). Tap water enriched with NaNO<sub>3</sub>, NaClO<sub>4</sub> and KH<sub>2</sub>PO<sub>4</sub> (influent concentration of 1 mg P/L) was used as feed solution for all experiments. Carbon source for bacterial growth was not added and based on the inherent carbon content of the water (alkalinity of ∼140 mg/L as CaCO<sub>3</sub> at pH 7.5–8). The recirculation flow rate was 6600 mL/min in the first experiment when only the main reactor unit was used (Section 3.1). In all other trials, the recirculation flow rate in the main reactor unit was 3800 mL/min. Water temperature was kept at 30 ± 1 °C. The pH in the main reactor unit was maintained at 7–7.1 to by dosing hydrochloric acid. The relatively low pH was aimed to prevent an extreme pH increase within the biofilm, which leads to NO<sub>2</sub>-N accumulation [26,27]. Samples of influent, effluent from the main reactor unit and effluent from the polishing unit were collected for further water analyses.

Rate calculations in this work were based on the packing volume of the carriers in the main reactor unit (2.5 L) and the polishing unit (1.9 L). In all experiments, excess biomass growth was removed every few days by washing of carriers, column and pipes with tap water (the polishing unit never had to be cleaned).

### 2.2. Water and gas analyses

Nitrate, perchlorate and sulfate were determined using a Metrohm 761 ion chromatograph (IC) equipped with a 150 mm Metrosep A Supp 5 column with column guard and suppressor using a CO<sub>3</sub><sup>2-</sup>/HCO<sub>3</sub> eluent. Nitrite-N and alkalinity were measured according to Standard Methods (Method 4500 and Method 2320, respectively). Total Organic Carbon (TOC) concentration was determined by a TOC-VCPH analyzer (Shimadzu, Kyoto, Japan). DOC concentration was determined by performing TOC analysis on samples filtered through 0.22 mm syringe filter. Hydrogen concentration in gas phase was measured by gas chromatography (TCD detector; column: HP-PLOT-Q 30 m; 0.53 mm. 40u, Agilent 7890A). Gas samples were injected directly from the reactor headspace into a 20 mL sealed serum bottle for 1 min with gas flow rate of 250 mL/min to ensure exchange of the entire gas volume in the bottle. Dissolved H<sub>2</sub> concentration was measured by headspace analysis of effluent samples injected to a sealed serum bottle using the same gas chromatograph.

### 2.3. Microbial population analysis using high-throughput sequencing and PCR-DGGE

Biofilm samples for microbial population analysis were taken from the pressurized hydrogenotrophic denitrifying reactor (i.e. the main reactor unit) before (t = 0) and after (t = 25 days) the addition of ClO<sub>4</sub>. Total genomic DNA was extracted using FastDNA SPIN Kit for Soil (MP Biomedicals) following the manufacturer’s protocol. Pellets of 0.5 mL from suspensions of the reactor’s biofilm were used as samples. The DNA concentrations of the extracts were measured with the NanoDrop 1000 Spectrophotometer (Thermo Scientific), adjusted for polymerase chain reaction (PCR) amplification and stored at −20 °C until further use.

High throughput sequencing analysis was performed by using Illumina Miseq (Hy laboratories Ltd, Israel). Samples of DNA were subjected to two rounds of PCR to prepare the libraries for sequencing. The first PCR reaction was performed to amplify the V4 region of the 16 s rDNA gene, with primers that included the CS1 and CS2 sequences from Fluidigm. The second PCR was performed using the Access Array Barcode Library for Illumina Sequencers from Fluidigm. The sample data were analyzed using the 16 s metagenomic application on BaseSpace (Illumina). The high quality reads that passed quality filtration were used for the identification of microbial population. Only predominant microbial populations are given; the remaining microbial population is shown as ‘Others’.

For DGGE, approximately 600 base pairs (bp) of the 5’ end of the variable region V3–V5 of the bacterial 16s rDNA were amplified using the primer pair consisting of 341F (5’-CCTACGGGAGGCAGCAG-3’) with a GC clamp (5’-CCCCCGCCGCCCCCGCTCCCGCCGCCCCCG-3’) and 907 R (5’-CCCTCAATTCTACCTAGTG-3’) [28]. The PCR reactions were performed in a thermocycler (TProfessional Basic Gradient Thermocycler; Biometra) with Apex RED Taq Master Mix (Genesee Scientific Corp.). The PCR conditions, denaturing gradient gel electrophoresis (DGGE) and sequencing procedures were used as mentioned by Desitti et al. [29]. Four dominant bands from DGGE were sequenced and compared with the partial sequence of 16s rDNA bacterial names obtained by high throughput sequencing method. Sequence alignment and phylogenetic tree were performed using MEGA6 [30].

### 3. Results and discussion

#### 3.1. Perchlorate removal in the pressurized hydrogenotrophic denitrifying reactor

An initial investigation of ClO<sub>4</sub> removal in the pressurized hydrogenotrophic denitrifying reactor using biofilm carriers from former denitrification experiments was first carried out for 25 days. During the experimental period, the inlet NO<sub>3</sub>-N and ClO<sub>4</sub> concentrations were 15 and 20 mg/L, respectively. The flow rate was increased gradually over time from 20 to 200 mL/min. The reactor’s total pressure was 2 bar and the recirculation flow rate was 6600 mL/min. The results for volumetric ClO<sub>4</sub> removal rate over time are shown in Fig. 2. Fig. 2 shows that ClO<sub>4</sub> reduction started immediately after ClO<sub>4</sub> addition, i.e., during the first day of operation. The immediate acclimation of bacteria from the former denitrification reactor to reduce ClO<sub>4</sub> demonstrates that no specialized inoculation was required. A maximal ClO<sub>4</sub> volumetric removal rate of 1.83 g/(L reactor·d) was observed after 25 days of operation. For comparison, Logan et al. reported a slightly lower removal rate of 1.16 g/(L reactor·d) in a non-pressurized unsaturated-flow hydrogenotrophic reactor at a lower temperature of 23 °C, similar pH (7) and influent ClO<sub>4</sub> concentration (18 mg/L) without NO<sub>3</sub> [5]. Sharp fluctuations in the ClO<sub>4</sub> removal...
immediately upon its addition (Section 3.1). Two main phyla, Flavobacteriia and Bacteroidetes, accompanied with loss of biomass and change of conditions. Effluents (e.g. day 19 and 24) can be attributed to reactor cleaning due to the large presence of Dechloromonas, as confirmed by high throughput sequencing and PCR-DGGE analysis. Strains of Dechloromonas have been shown to grow on ClO4 and NO3, while its ability to use H2 as an electron donor has also been shown [32]. Significant changes were not observed in the microbial population after 25 days of concurrent ClO4 and NO3 reduction, primarily due to the much greater electron accepting capacity (EAC) of NO3 (15 mg/L or 5.4 mequiv EAC) as opposed to ClO4 (20 mg/L or 1.6 mequiv EAC) during the experimental period.

Fig. 3 shows the relative amounts of the dominant genera before and after the addition of ClO4. Zoogloea was the dominant genus in the reactor both before and after the addition of ClO4, accounting for 38.3% and 31.0%, respectively, followed by Dechloromonas (28.7% before, 23.4% after), Flavobacterium (13.7% before, 20.3% after), Chryseobacterium (10.5% before, 11.0% after), and Vogesella (2.4% before, 2.9% after). Less significant genera are listed as ‘Others’ (6.4% before, 11.0% after). All five genera have species that can carry out denitrification, but only Dechloromonas has been associated with denitrification and ClO4 reduction [31].

PCR-DGGE analysis of the pressurized hydrogenotrophic denitrifying reactor gave similar results with seven nearly identical bands observed before and after the addition of ClO4 (Fig. 4). Four of the bands from the DGGE were sequenced (Fig. 4, Lane 1) and compared to the closest phylogenetic relatives found in the NCBI gene bank of the predominant bacteria recovered from high throughput sequencing (Zoogloea ramigera, Zoogloea resiniophila, Dechloromonas hortensis, Dechloromonas agitata, Vogesella perlucida, Flavobacterium cheniæ, and Chryseobacterium soli). The aligned sequences from high throughput sequencing and PCR-DGGE are presented in Fig. 5.

The phylogenetic tree shows a divergence of only 5%, with DGGE bands-1,2 showing a near 100% similarity to Dechloromonas sp., while DGGE bands-3,4 have a close similarity with Flavobacterium and Chryseobacterium. The reactor’s ability to metabolize ClO4 almost immediately from the outset of ClO4 addition was due to the large presence of Dechloromonas as confirmed by high throughput sequencing and PCR-DGGE analysis. Strains of Dechloromonas have been shown to grow on ClO4 and NO3, while its ability to use H2 as an electron donor has also been shown [32]. Significant changes were not observed in the microbial population after 25 days of concurrent ClO4 and NO3 reduction, primarily due to the much greater electron accepting capacity (EAC) of NO3 (15 mg/L or 5.4 mequiv EAC) as opposed to ClO4 (20 mg/L or 1.6 mequiv EAC) during the experimental period.

3.3. Reduction of different electron acceptors in the combined treatment scheme

Following the initial investigation of ClO4 reduction using only the pressurized hydrogenotrophic reactor, the removal of different electron acceptors (NO3-N, NO2-N, ClO4 and SO42-S) was studied in the combined treatment scheme at different flow rates for two months. The inlet concentrations of NO3-N and ClO4 were adjusted to 25 and 10 mg/L, respectively, while SO42-S concentrations in tap water ranged between 7 and 9 mg/L. The results are summarized in
Fig. 6. All measurements in Fig. 6 were repeated five times, each in a different day.

As expected, higher NO$_3$-N removal was observed in the main reactor unit at lower flow rates due to the higher retention time. Significant ClO$_4$ removal was observed in the main reactor unit (from 10 to 2 mg/L) only when the lowest flow rate was applied; probably due to the higher retention time with the correspondent lower NO$_3$-N concentration (CSTR conditions, i.e., NO$_3$-N concentration of about 1 mg/L). At higher flow rates, the effluent NO$_3$ concentration in the main reactor increased and the average ClO$_4$ removal rates calculated in the main reactor unit decreased (0.73, 0.3 and 0.26 g/(L$_{reactor}$ d) for the operation with 150, 225 and 300 mL/min, respectively), suggesting that simultaneous removal of NO$_3$ and ClO$_4$ occurred with inhibition of ClO$_4$ reduction due to the competition for electrons by NO$_3$ [33].

In the polishing unit denitrification always occurred, while significant ClO$_4$ reduction (>1 mg/L) occurred only in the presence of very low NO$_3$-N concentrations. Sulfate reduction was observed only in the presence of very low NO$_3$-N and ClO$_4$ concentrations. These results can be explained by the combination of low concentration of dissolved H$_2$ in the polishing unit (Section 3.4), higher denitrifying population than perchlorate-reducing population in the system, NO$_3$ reduction by some perchlorate-reducing bacteria (Section 3.2) and the thermodynamics-based priority of NO$_3$ and ClO$_4$ reduction over SO$_4^{2-}$ reduction. Therefore, further reduction of ClO$_4$ to concentration close to zero occurred in the polishing unit only under the lowest flow rate when the NO$_3$-N concentration was already very low. The average ClO$_4$ removal rates calculated in the polishing unit were much lower compared to the main reactor unit (0.12, 0.02 and 0 g/(L$_{reactor}$ d) for the operation with 150, 225 and 300 mL/min, respectively). This observation
can be attributed to the lower ClO₄⁻ concentration in the polishing unit. Also, the lower NO₃⁻ concentration in the polishing unit may not support a significant growth of ClO₄⁻ reducing bacteria as in the main reactor unit [8]. In the case where NO₃ or ClO₄ are further reduced in the polishing unit, an improved H₂ utilization is achieved. Sulfate reduction is not one of the treatment goals and therefore does not improve H₂ utilization in terms of financial aspects. However, it minimizes the amount of H₂ released to atmosphere and therefore may contribute to the safety of the process. Detailed calculations and measurements for H₂ utilization efficiencies are described in the next section. DOC analysis showed a minor increase of 0.15 mg/L after the polishing unit as compared to the inlet of the polishing unit.

3.4. Hydrogen utilization and effluent quality using the combined system for the treatment of typical polluted groundwater

Following the experiments with relatively high influent ClO₄⁻ concentrations, the removal of a lower inlet ClO₄⁻ concentration of 1.5 mg/L (the NO₃⁻ concentration remained 25 mg/L) was studied in order to simulate typical conditions and to check the ability of the polishing unit to decrease ClO₄⁻ levels to trace concentrations, below 15 mg/L. The flow rate in this experiment was adjusted to 155 mL/min.

The removal of ClO₄⁻, together with that of NO₃⁻ and NO₂⁻, over the different treatment stages at steady state are shown in Fig. 7. Fig. 7 shows that ClO₄⁻ concentration was reduced to an average trace level of lower than 7 mg/L in the polishing unit. The lowest value observed during steady state was 2 mg/L. Together with reduction of NO₃ and NO₂ concentrations to below 0.1 mg/L, without any accumulation of chlorate (ClO₃⁻) and chlorite (ClO₂⁻), and with minimal increase in DOC concentration after the biological process (maximum DOC measured in effluent water was ~2 mg/L compared to 0.6 mg/L in feed water), the combined treatment scheme is suitable for drinking water production. The plug-flow character of the polishing unit is advantageous for reducing ClO₄⁻ concentrations to such low trace levels for two main reasons: (1) in CSTRs, reaching such low trace concentrations is harder due to mixing with the inlet stream having much higher concentrations; (2) better performance of ClO₄⁻ reduction can be achieved downstream after depletion of NO₃⁻.

The submerged-flow regime in the polishing unit minimizes H₂ discharge to the atmosphere and allows for its further consumption. In the polishing unit where ClO₄⁻ concentration is very low, NO₃ can also support growth of ClO₄⁻ reducing bacteria and thus maintain this bacterial population [8]. Fig. 7 also shows that no
SO\(_4^{2-}\) reduction was observed in the polishing unit due to the low concentrations of H\(_2\) (Table 1).

Table 1 summarizes the main results and calculations at steady state, including GC analyses for H\(_2\) concentration in gas and liquid phase. Table 1 shows the high denitrification rates obtained in the pressurized reactor as compared to other technologies, even at low effluent NO\(_3^-\)-N concentrations. The rate can be further increased by applying higher recirculation rates [25]. A good correlation was found between measured and theoretical H\(_2\) pressure in the closed-headspace reactor, indicating steady-state conditions. As expected, the measured dissolved H\(_2\) concentration was a bit lower than its value at saturation due to H\(_2\) consumption by biomass. The H\(_2\) utilization efficiencies of the main pressurized reactor unit or the combined treatment scheme were calculated by Eq. (1).

\[
\text{H}_2 \text{ utilization efficiency} = \frac{H_c}{H_c + H_r} \times 100\%
\]  

where H\(_c\) is the measured dissolved H\(_2\) in the effluent of the main reactor unit or the polishing unit; and H\(_r\) is the H\(_2\) consumption (in units of mg/l] in the main reactor unit or overall process. In order to calculate the H\(_2\) consumption, a previously suggested metabolic stoichiometry for hydrogenotrophic denitrification [34] (Eq. (2)) and SO\(_4^{2-}\) reduction [35] were used. For ClO\(_4^−\) reduction, the metabolic stoichiometry (Eq. (3)) was built applying the same yield coefficient used for hydrogenotrophic denitrification due to the similar thermodynamics of the processes [10].

\[
\text{NO}_3^- + 3\text{H}_2 + \text{H}^+ + 0.22\text{CO}_2 \rightarrow \text{0.48N}_2 + 3.35\text{H}_2\text{O} + 0.04\text{C}_3\text{H}_7\text{O}_2\text{N} \tag{2}
\]

\[
\text{ClO}_4^- + 4.61\text{H}_2 + 0.31\text{CO}_2 \rightarrow \text{Cl}^- + 4.48\text{H}_2\text{O} + 0.06\text{C}_3\text{H}_7\text{O}_2\text{N} \tag{3}
\]

The H\(_2\) utilization efficiency calculated after the main reactor unit was similar to the previous findings in the pressurized reactor [24]. The theoretical consumption of H\(_2\) in the polishing unit was based on the assumption that all three electron acceptors were reduced by H\(_2\) consuming bacteria. The result (0.56 mg/L) was very close to the measured dissolved H\(_2\) after the main reactor unit (0.52 mg/L), albeit a bit higher. The difference can be attributed to minor heterotrophic activity. The almost zero residual of H\(_2\) in the polishing unit effluent correlates well with the fact that SO\(_4^{2-}\) was not reduced in the second unit due to lack of H\(_2\). The results of the combined treatment scheme show almost complete H\(_2\) utilization with a total consumption of 10.9 mg H\(_2\) per liter of water treated. To the best of our knowledge, our results of 100% utilization of H\(_2\) gas together with reduction of perchlorate concentration to low trace concentrations of ~2 μg/L were previously reported only for the MBR [10].

**4. Conclusion**

A new treatment scheme for removal of NO\(_3^-\) and ClO\(_4^-\) from drinking water, based on an unsaturated-flow pressurized hydrogeotrophic reactor combined with an up-flow submerged-bed open-to-atmosphere polishing unit was investigated. Degradation of ClO\(_4^-\) started immediately after the addition of ClO\(_4^-\) to the pressurized denitrification reactor, indicating that no special inoculum was needed for adjusting the reactor for ClO\(_4^-\) reduction. This finding was supported by the large presence of the genus *Dechloromonas* in the reactor prior to the introduction of ClO\(_4^-\). Co-reduction of NO\(_3^-\) and ClO\(_4^-\) was observed in the pressurized reactor with significant inhibition in the ClO\(_4^-\) reduction rate at higher NO\(_3^-\) concentrations. The combination of submerged and plug-flow conditions in the polishing unit minimizes the discharge to atmosphere of the residual dissolved H\(_2\) from the pressurized reactor and allows for the decrease of ClO\(_4^-\) concentration to trace levels of 2 μg/L. The further consumption of H\(_2\) in the polishing unit resulted in an increase in H\(_2\) utilization efficiency from 95% to almost 100%.

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