ABSTRACT OF THE PH.D. THESIS

UNIVERSITY OF SZEGED PH.D. SCHOOL OF ENVIRONMENTAL SCIENCES PROGRAMME OF ENVIRONMENTAL BIOCHEMISTRY AND BIOTECHNOLOGY

BIO-ELECTROCHEMICAL SYSTEMS IN DRINKING WATER TREATMENT

BY ISTVÁN KISS

SUPERVISOR: Dr. MIKLÓS KÁLMÁN

BAY ZOLTÁN FOUNDATION FOR APPLIED RESEARCH INSTITUTE FOR BIOTECHNOLOGY

Szeged, 2002

INTRODUCTION

One of the most important current problems is the contamination of environment contamination by agricultural and industrial activity. Accordingly, tasks of the environmental sciences include surveys of the state of the environment and the prevention of further contamination.

One of the central topics of research projects relating to environmental protection is drinking water and its quality control. Nitrate, a prominent inorganic contaminant, does not have direct harmful effects on human health, but its derivative nitrite is very toxic, particularly for infants. The extent of contamination of groundwater with nitrate is so high that its human utilization for drinking purposes is not recommended or is even prohibited, because of public health regulations.

A number of methods are available for drinking water denitrification, including physicochemical or biological methods and their combination.

However, only biological processes can solve the elimination of nitrate completely. This follows from the fact that some microorganisms possess a complete enzyme system with wich to reduce nitrate to inert nitrogen gas in the presence of appropriate electron donors.

Depending on the nature of the electron donors, biological denitrification can be divided into two groups, such as heterotrophic (demanding organic compounds) and autotrophic (not demanding organic compounds). Some difficulties are encountered in the application of heterotrophic procedures for drinking water denitrification, considering the use of organic compounds and difficult downstream processes.

The practical application of autotrophic denitrification is hindered by the poor operability of hydrogen gas as an optimal electron donor. A well-organized, and regulated hydrogen source, such as the electrolysis of the water, would facilitate the widespread technology based on autotrophy. The biological denitrification systems referred in the literature are characterized by low activity levels.

The Jacob Blaustein Institute for Desert Research (Israel), the Biological Research Center of the Hungarian Academy of Sciences (Szeged) and the Bay Zoltan Foundation for Applied Research (Szeged) set out to perform joint research with a view to the development of a high activity biological water treatment system based on electrochemical water hydrolysis as a source of hydrogen.

AIMS

The objective of this study was to develop an efficient, cost-effective microbial process for the removal of nitrate from polluted groundwater.

From a consideration of the advantages and disadvantages of autotrophic and heterotrophic denitrification, it was decided to develop an autotrophic system: on adoption of this technology, presence of organic compounds can be avoided which makes the downstream process simpler. On the application of such methods, the expected lower denitrification activities will be satisfactory when groundwater containing medium or low nitrate levels is treated.

The following criteria were defined:

• The microbes applied are characterized by autotrophic denitrification on the use of hydrogen as an energy source.

• The hydrogen required by the biological processes is available from the electrolysis of water.

• The reactor set-up should operate in continuous mode.

METHODS

To achieve our aims, appropriate microbes were isolated and characterized, and used to develop a bioactive, denitrifying biofilm on the surface of granulated activated carbon as a carrier, via a heterotrophic, autotrophic and partial adhesion method.

The biofilms on the surface of the carrier were characterized according to their polysaccharide and protein contents, and their denitrification activities were determined.

Practical testing of the carriers covered by biofilm was carried out under anaerobic, autotrophic circumstances, hydrogen being produced as the electron donor for the biological processes by the electrolysis of water.

SUMMARY OF NEW SCIENTIFIC RESULTS

An autotrophic, biological denitrification system consuming hydrogen was set up in our work, and its development, functioning, activity and other characteristics were analysed.

CONCLUSIONS:

• Biofilm development based on the adhesion characteristics of the applied carrier is much easier than common biofilm development methods, and allows the preparation of stable biofilms of high activity using isolated bacteria.

• The applied bacteria strains afforded high denitrification activity under autotrophic circumstances, consuming hydrogen as an electron source.

• Granulated activated carbon is suitable for purposes of drinking water treatment, its application facilitates biofilm development, and the main malfunction of sand bead reactors due to gas inclusions can be avoided.

• The stable operation of the reactors indicates that electrochemical cells are suitable for practical purposes, and their maintenance is simple.

- The bioactive carrier placed into the cathodic chamber of the ECBR reactor (*ElectroChemical BioReactor*) set-up functioned with short hydraulic retention times (HRT) and exhibited denitrification activity.
- It was found that carriers covered by biofilm prepared from the same bacterial strains displayed different HRT values and activities, the values being significantly higher for the ECBR arrangement than for the EC+BR arrangement. It was proved that in such a case not only biological, but also electrochemical reactions play a role in the performance of denitrification activities.
- As concerns the microbial processes inside the reactor, the stages of denitrification were separated, the reduction of nitrate taking place in the initial part of the set-up, followed by nitrite reduction.

- Changes of the initial microbial composition (BMIX strain mixture) were studied. It was found that, in spite of non-sterile operating circumstances, only the initial microorganisms strains could multiply; the concentrations of other, non-initial microbes were negligible.
- The maintenance and running cost of the reactors are very low, which is promising as regards their future practical application. Scaling-up is not difficult, and on use of the same electrochemical set-up, the costs would not increase proportionally.
- The results calculated from the reactor running data are in the same range as other results in literature, but our results are better than those of other systems based on water electrolysis.

LIST OF PUBLICATIONS

Publications related to the thesis

Papers

- Kiss I., Szekeres Sz., Bejerano T.T. és Soares M.I.M. (2000). Hydrogen-dependent denitrification: preliminary assessment of two bio-electrochemical systems. *Water Science and Technology* 42:373-379. Impact factor (2000): 0.495
- Szekeres Sz., Kiss I., Bejerano T.T. és Soares. M.I.M. (2001). Hydrogen-dependent denitrification in a two-reactor bio-electrochemical system. *Water Research* 35:715-719. Impact factor (2000): 1.285
- Szekeres Sz., **Kiss I.,** Kalman M., Soares M.I.M. (2002). Microbial population in an hydrogen-dependent denitrification reactor. *Water Research* (accepted). Impact factor (2000): 1.285

POSTERS

Hansel M., Kesserû P., Tatar-Kis T., Szemes M., Kiss I., Polyak B. (1996) New prospects in aerobic water denitrification. *Third International Symposium and Exhibition on Enviromental Contamination in Central and Eastern Europe*, Warsaw, Poland 1996, szeptember 10-13.

OTHER PUBLICATIONS

Papers and posters

- Kiss I., Kesserű P., Fehér B., Bihari Z. és Polyák B. (2002) Co-immobilization of symbiotic green algae and Saccharomyces unispora. *Symbiosis* (in press). Impact factor (2000): 0.895
- Kesserű P., Kiss I. Bihari Z. Polyák B. (2002) Investigation of the denitrification activity of immobilized *Pseudomonas butanovora* cells in the presence of different organic substrates. *Water Research* (in press). Impact factor (2000): 1.285
- Kesserű P., Kiss I. Bihari Z. Polyák B. (2001) Roles of nitrate ions. *Technika Műszaki szemle* 44.(9): 37-39.
- Perei K., Rákhely G., Kiss I., Polyák B. és Kovács. K.L. (2001) Biodegradation of sulfanilic acid by Pseudomonas paucimobilis *Appl. Microbiol. Biotechnol.* 55:101-107. Impact factor (2000): 1.505
- Bálint T., Ferenczy J., Kátai F., Kiss I., Kufcsák O., Láng G., Polyhos Cs., Szabó I., Szegletes T. és Nemcsók J. (1999) The contribution of a pyrethroid insecticide to the massive eel (Anguilla anguilla) devastation, in Lake Balaton, in 1995. *Acta Biologica Hungarica* 50(1-3): 161-173. Impact factor (2000): 0.297.
- Perei K., Bodrossy L., Rákhely G., Bagyinka Cs., Bihari Z., Kesserű P., Kiss I., Polyák P. és Kovács K.L. (1999) *Biodegradation of selected hazardous waste*. Proc. COST Action 831.
- Vértesi A., Simon L.M, Kiss I. és Szajáni B. (1999) Preparation, characterization and application of immobilized carboxypeptidase A. *Enzyme and Microbial Technology* 25: 73-79. Impact factor (2000): 1.411
- Bálint T., Ferenczy J., Kátai F., Kiss I., Kráczer L., Kufcsák O., Láng G., Polyhos Cs., Szabó
 I., Szegletes T. és Nemcsók J. (1997) Similiraties and differences between the massive eel (Anguilla anguilla L.) devastations that occurred in Lake Balaton in 1991 and 1995. *Ecotoxicology and Environmental Safety* 37: 17-23. Impact factor (2000): 1.060

- Bácsi Z, Kiss I, Polyák B. 3. (1997) Artificial symbioses in biotechnology. Environmental Meeting and exhibition, Veszprém.
- Dr. Polyák B, Perei K., Bihari Z, Eördegh G., Gárgyán A., Hansel M., Kesserű P., Kiss I., Tatár-K. T., Tóth M. (1997) "Sárkány ellen sárkányfű" *Technika Műszaki Szemle*, XL. évf. 4, 19-21.
- Kiss I. Polyák B., Hansel M., Perei K. (1995) Artificial symbioses in biotechnology Xth International Photosynthesis Congress. Montpellier, france, 20-25 August 1995
- Hörcsik T.Zs., Kiss I., Zsíros O., Polyák B., Balogh Á. (1995) Properties of fructose 1,6bisphosphatase from Chlorella pyrenoidosa and Chlorella zofingiensis ssp. International Conference of the Hungarian Biochemical Society Szeged.