

## Pre-nitrification by encapsulated nitrifiers – a possibility for self-sufficient energy operation of domestic WWTPs

M. Sievers\*, K.D. Vorlop\*\*, J. Hahne\*\*, M. Schlieker\*\* and S. Schäfer\*

\* CUTEC-Institut GmbH (Clausthal Environment Technology Institute), Leibnizstr. 21+23, 38678 Clausthal-Zellerfeld, Germany (E-mail: [michael.sievers@cutec.de](mailto:michael.sievers@cutec.de))

\*\* Institute for Technology and Biosystems Engineering, Federal Agricultural Research Centre (FAL), Bundesallee 50, 38116 Braunschweig, Germany

**Abstract** The overall energy consumption of domestic wastewater treatment plants (WWTPs) increases with treatment efficiency. Approximately 30 to 45 kWh per person equivalent and year is mostly necessary for advanced nitrogen and phosphorus removal, while the aeration contains the main part of approximately 60%. A new process using encapsulated nitrifiers on gel lens beads is introduced to overcome the high energy consumption of aeration. A more selective nitrification process was found at a nitrification rate of between 50 and 60 mg nitrogen per hour and litre reaction volume corresponding to a hydraulic retention time (HRT) of about 30 to 60 minutes while the soluble Chemical Oxygen Demand (COD) removal could be less than 30% depending on operational conditions of the bio-reactor. The latter enables internal use of wastewater's COD for a post denitrification. For the new process the energy consumption as well as total volume of bio-reactor are much less (approximately 30 to 50% for both) than conventional processes due to the low sludge age for COD and nitrate removal and the avoidance of internal wastewater recycle. Therefore, self-sufficient energy operation of domestic WWTPs operating with advanced treatment efficiency could become possible, if energy recovery by anaerobic sludge digestion is included.

**Keywords** Denitrification; encapsulation; energy consumption; immobilisation; nitrification; nitrogen removal

### Introduction

Nitrogen removal has become a standard technology in wastewater treatment. Nevertheless, interest has recently increased in cost-effective processes for ammonia and nitrate removal due to the fact that they still require significant resources, i.e. energy and chemicals. The consumption of energy and chemicals still increase with treatment efficiency. To make wastewater treatment more sustainable, nitrogen removal processes have to be improved by implementation of new processes and/or changes in treatment technologies.

Presently, several novel biological treatment processes and technologies are under development. The objectives are commonly to avoid disadvantages of a) energy consumed by aeration, b) organic chemicals needed for denitrification of nitrified water and c) water capacity for pH adjustment. Examples for a more sustainable treatment of wastewater are ammonia oxidation processes, which in particular produce directly molecular nitrogen, reducing the BOD needed for denitrification. The treatment of wastewater containing high ammonia concentration could become suitable by the anaerobic ammonia oxidation process (combined Sharon/ANAMMOX, Jetten *et al.*, 1999; van Dongen *et al.*, 2001), by additional feed of nitrogen dioxide NO<sub>2</sub> (Schmidt and Bock, 1997), by the autotrophic denitrification process (Twachtmann and Metzger, 1998). Moreover, process intensification by immobilisation are often used additionally (Twachtmann and Metzger, 1998; Jetten *et al.*, 1999; van Dongen *et al.*, 2001; Tanaka *et al.*, 1996).

This paper aims at an alternative cost-effective nitrogen and COD elimination process dealing with a conventional two step nitrification process by encapsulated nitrifying

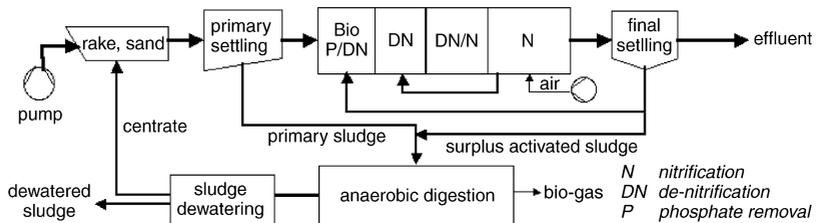
micro-organisms, which seems to be more suitable for low ammonia concentration levels such as in sewage.

### Energy and cost distribution of domestic WWTPs

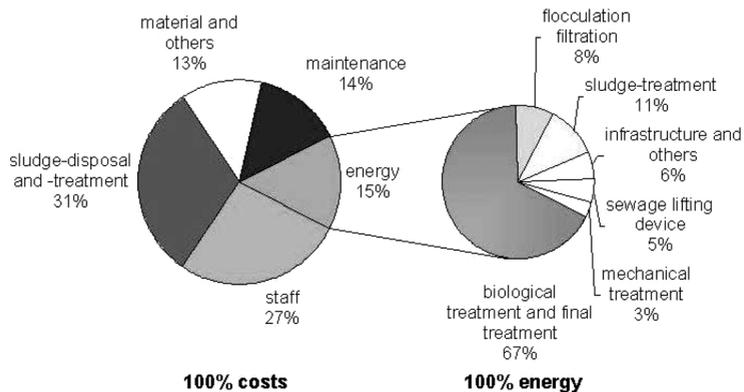
A flow diagram of a typical sewage treatment for 100,000 people equivalent is shown in Figure 1. The average daily flow rate is about 24,500 m<sup>3</sup> and the overall volume of the biological basin is about approximately 25,000 m<sup>3</sup> (aeration zone 11,500 m<sup>3</sup>) i.e. considered by software ARA-BER for advanced nitrogen and phosphorus removal (Müller *et al.*, 1999). Additionally, primary sludge and surplus activated sludge could be treated by anaerobic digestion followed by thickening or dewatering producing stabilised dewatered sludge for utilisation as manure or further treatment by thermal processes and land-filling/disposal.

An example of distribution of costs and energy for such sewage treatment is based on a German benchmarking study (Müller *et al.*, 1999) shown in Figure 2. Referring to this figure, costs of energy have a substantial contribution of 15% of total costs, whereas 10% of total costs is for aeration of biological processes.

The present electrical energy consumption is mostly between 30 and 40 kWh per people equivalent (p.e.) which could be reduced to 25 kWh per p.e. and year by using the best available techniques with minimal energy requirement (Müller *et al.*, 1999). For WWTPs including anaerobic sludge digestion, reduction of total energy consumption would take place by energy recovery from biogas. The biogas production depends on many different factors; commonly, the average energy content of biogas is about 6.4 kWh per m<sup>3</sup> gas under standard conditions containing 65 vol.-% of methane. Therefore, a yearly average of 13 kWh per people equivalent and year is suitable for electrical energy recovery by a combined power station (Müller *et al.*, 1999). Under these assumptions, a total minimal electrical energy requirement of 12 kWh/p.e. a for advanced sewage treatment is necessary.



**Figure 1** Flow diagram for advanced sewage treatment



**Figure 2** Distribution of costs and energy for advanced sewage treatment (Müller *et al.*, 1999)

### Specific oxygen consumption

Based on the low growth rate of nitrifying bacteria, the sludge age has to be adapted to the suitable nitrification rate and avoidance of wash out of nitrifying bacteria by surplus activated sludge. (Note: sludge age is defined as Solid Retention Time (SRT) of sludge mass in biological basin related to the daily mass flow rate of surplus activated sludge.) A high SRT of more than 10 days is common in single sludge systems with nitrification and pre-, post- or simultaneous de-nitrification and it often has to be increased to 20 days or more, especially at lower temperatures. Reference to Table 1 shows that the specific oxygen consumption related to the Biological Oxygen Demand (BOD) to be oxidised increases significantly by improving the SRT. This data calculated from measurements (Schmitt *et al.*, 1998) show that a potential of 30 to 40% for energy reduction through less specific oxygen consumption is still possible by reduction of sludge age from 20 to less than 4 days. Two effects are mainly reasonable for this different oxygen consumption: a) the oxygen consumption for endogenous respiration would be decreased for lower sludge ages and b) the BOD of settled sewage would be less oxidised into carbon dioxide but more into additional biomass. This change in biomass yield coefficient would lead to a more energy efficient technology as it gives a significant potential for higher biogas production during anaerobic digestion of sludge. A more sustainable wastewater treatment could then be realised, because more of organic compounds in sewage would be used for energy production via sludge digestion. Table 1 gives also an overview of biogas production depending on the SRT (Müller *et al.*, 1999). It is obvious that an increase of 10% in biogas production would happen by reducing SRT from 25 to 13 days. Moreover, novel technologies for enhanced biogas production by cost-effective sludge pre-treatment processes are under demonstration (Onyeche, 1999; Onyeche *et al.*, 2002). It seems to be possible that the biogas production could be increased further by an additional 20% corresponding to the carbonaceous compounds of well stabilised sludge being reduced by an additional 50%.

The total amount of oxygen required for oxidation of ammonia and COD depends on their mass flow rate in settled sewage. In general, the minimal specific oxygen consumption for conventional nitrification-denitrification process is theoretically reached if the COD of nitrified sewage contains enough BOD<sub>5</sub> for denitrification process with respect to the minimum endogenous respiration and maximum yield for heterotrophic micro-organisms. This would principally lead to a selective pre-nitrification process followed by high loaded de-nitrification.

### Methods

The methods that have been used in development of a more sustainable advanced sewage treatment with less energy consumption are: a) cheap immobilisation of nitrifying bacteria on gel lens beads (LentiKats®), b) process configuration such as “pre-nitrification”, c) high rate nitrification of settled sewage to produce an effluent having sufficient BOD<sub>5</sub>, d)

**Table 1** Specific oxygen consumption and biogas production in single sludge systems for different sludge ages (Müller *et al.*, 1999; Schmitt *et al.*, 1998)

SRT in days	Specific oxygen consumption in kgO <sub>2</sub> per kgBOD <sub>5</sub>	Specific bio-gas production in m <sup>3</sup> per people equivalent and year			comment
		Pre-settling	Pre-settling	Pre-settling	
		HRT 0.5 h	HRT 1 h	HRT 2 h	
4 to 5	0.81 (10°C)–0.97 (20°C)	8.4–9.5	9.1–10.6	9.9–11.3	C-Removal
10	1.02 (10°C)–1.17 (20°C)	–	–	–	C+N-Removal
13	–	6.9–8.0	8.0–9.1	9.1–10.6	C+N-Removal
25	1.21 (10°C)–1.31 (20°C)	6.6–7.7	7.3–8.4	8.4–9.5	C+N-Removal

comparison of treatment efficiency and energy consumption with conventional treatment plants capable of producing similar effluents.

### Immobilisation

A new method, which allows the gelation of PVA-solutions at room temperature by means of controlled partial drying, has been developed. Due to their characteristic lenticular shape the resulting particles are named LentiKats<sup>®</sup>. The particles formed by this procedure combine the advantages of both large and small beads. On the one hand side their size is about 3 to 4 mm in diameter and can be retained i.e. by sieve technology or by classifying in a combined fluidised-bed/settling system. On the other hand they are only 200 to 400  $\mu\text{m}$  thick, thus causing hardly any limitations due to diffusion to the enclosed biocatalysts.

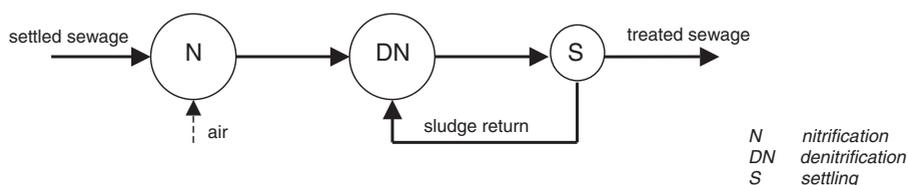
Since LentiKats<sup>®</sup> are based on polyvinyl alcohol they have the same properties as PVA cryogels. But based on the very mild encapsulation conditions, very high survival rates can be obtained. The complete immobilisation procedure takes place in less than one hour and also the stress of the partial drying is tolerable for most organisms.

The benefiting influence of avoiding extreme temperature conditions in contrast to the cryogelated PVA-hydrogels was shown by immobilising a mixed culture of sensitive nitrifying bacteria (*Nitrosomonas europaea* and *Nitrobacter winogradskyi*), raising the initial activity of the biomass from below 1% (entrapped by freezing-thawing method,  $-20^{\circ}\text{C}$ ) to 75% for immobilisation in LentiKats<sup>®</sup> compared to suspended cells of nitrifiers. Production devices for lab- and technical scale were developed to produce identical droplets simultaneously using special printers and a conveyor belt system.

### Nitrogen elimination process with LentiKats<sup>®</sup>

The simplified flow diagram for the LentiKats<sup>®</sup> nitrogen elimination process is shown in Figure 3. The first stage contains the encapsulated nitrifying micro-organisms. No heterotrophs would be encapsulated and a more selective nitrification is possible in cases of low biofilm thickness of heterotrophs attached on lens bead surfaces. This could be ensured i.e. by more turbulence in reactor. Minimisation of BOD-removal in this stage by low concentration of heterotrophic micro-organisms is needed to ensure complete de-nitrification at the second stage by using the original BOD as an electron-donor. To minimise BOD removal in the first stage, no thickened activated sludge would be cycled back from settling tank to this stage. Therefore, this process would like called “pre-nitrification” instead of post-denitrification.

The main advantages of this process are: a) the high SRT, which is necessary in single sludge systems to ensure complete nitrification due to the slow growth of nitrifying micro-organisms, would be reduced. Therefore, the additional aeration, which increases with the SRT, could be reduced substantially, b) The internal recycle of nitrified wastewater as commonly established for pre-denitrification process is not commonly necessary with respect to waste waters concentration of carbon ions and capacity to adjust pH. Additional reduction of energy consumption is possible.



**Figure 3** Flow diagram for “pre-nitrification” process

## Experimental

Continuously driven lab scale experiments have been carried out with settled sewage, which was collected twice a week from a domestic WWTP. The settled sewage was fed to the system via a continuously stirred tank. The nitrification and denitrification reactor were operated as stirred vessels with reaction volume of 2.8 L (aerated) and 9 L (not aerated) respectively.

The daily flow rate of settled sewage has been changed to between 40 and 150 L. Correct system operation have been checked by online measurement of

- pH, temperature and stirring in both reactors,
- flow rate of settled sewage, air and return sludge and
- dissolved oxygen in nitrification reactor.

Additionally, samples were collected before and after each treatment process once a day and five days per week for laboratory analysis of COD both homogenised and filtrated, ammonia, nitrite, nitrate, etc.

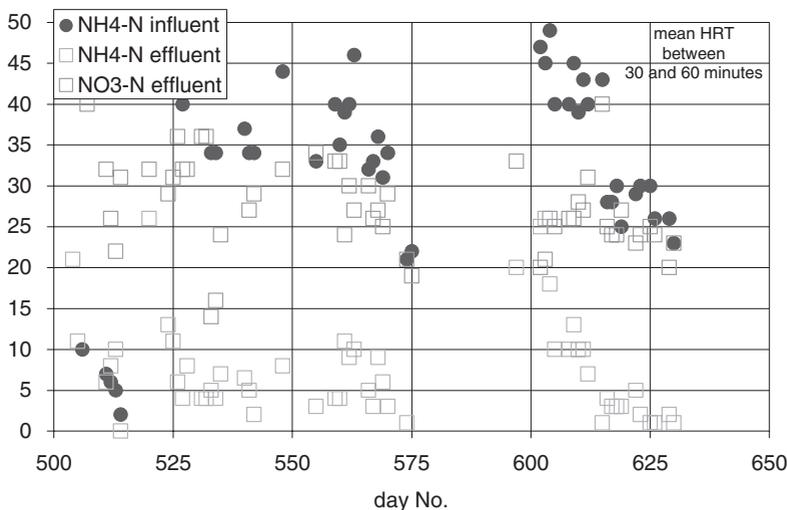
## Results and discussion

### Nitrification

As shown in Figure 4, a nearly complete nitrification of ammonia to nitrate has been established with an average volumetric reaction rate of about 45 mg nitrogen per litre reaction volume and hour. The hydraulic retention time was always at approximately 30 or 60 minutes. This is approximately 10 times lower compared to the conventional adjusted hydraulic retention times in aeration zones of sewage treatment plants. This figure also implies that nitrification could be always stable within a period of two years at present. Many disturbances including interruption of treatment process have been happened, but never destroy the LentiKats<sup>®</sup>. Moreover, full treatment efficiency was mostly reached at the next sampling period one day later. The high ammonia effluent at period of day No. 600 to 610 show that maximum nitrification rate was reached.

### De-nitrification and carbonaceous oxidation

The results listed in Table 2 show that the process of denitrification after nitrification with LentiKats<sup>®</sup> would be possible by internal use of wastewater carbon sources for denitrification by a nitrate removal efficiency up to 50–60%. However, the denitrification



**Figure 4** Nitrification of ammonia

**Table 2** Results during stable denitrification operation period

Run	T [°C]	Nitrification			de-nitrification					
		$\Delta\text{COD}^a$ [%]	r COD <sup>a</sup> [mg/(l·h)]	$\Delta\text{NH}_4\text{-N}$ [%]	r NH <sub>4</sub> -N [mg/(l·h)]	$\Delta\text{NO}_3\text{-N}$ [%]	r NO <sub>3</sub> -N [mg/(l·h)]	$\Delta\text{COD}^a$ [%]	r COD <sup>a</sup> [mg/(l·h)]	$\Delta\text{COD}^a$ overall [%]
1	16.6	50.0	182.4	53.1	25.8	61.9	2.6	36.9	7.6	68.4
2	17.0	50.3	188.4	53.8	25.2	26.1	1.2	28.4	5.0	64.4
3	17.4	51.2	169.2	64.6	31.8	26.3	1.0	26.8	4.4	64.3
4	17.1	43.1	124.2	98.2	67.2	12.8	1.2	24.3	5.0	56.9
5	17.9	60.1	243.6	94.9	45.0	24.4	2.0	12.6	2.4	65.1
6	18.4	41.2	175.2	95.3	60.6	17.9	1.4	40.0	6.4	64.7
7	18.2	65.4	105.6	97.0	58.8	31.1	2.8	40.4	3.8	79.4
8	18.0	85.2	474.0	95.9	56.4	50.0	2.8	27.9	3.8	89.3
9	18.4	77.7	636.0	90.9	48.0	31.0	1.8	27.8	4.0	83.9
10	17.9	89.3	700.8	95.6	51.6	33.3	2.2	7.0	0.8	90.0
11	18.7	84.7	753.6	93.2	49.2	20.6	1.4	31.0	4.4	89.5
12	18.3	64.1	199.2	92.2	28.2	29.7	2.2	33.3	5.0	76.1
13	18.3	2.0	162.0	94.3	49.2	22.2	1.6	73.7	35.8	74.2

<sup>a</sup> COD from filtered sample

process has to be optimised concerning kinetic study and avoidance of sludge flotation caused by denitrification in final clarifier. Note that the sludge concentration in the denitrification reactor was always very low at about 0.5 to 2 g/L. The reaction rate for denitrification is therefore around 1.5 mg per gram biomass and hour, which is a little lower than the reaction rate in pre-denitrification processes.

The oxidation of carbonaceous compounds in the nitrification reactor, measured in terms of COD in filtrated samples, has mostly between about 40 to 60% of the settled sewage. In spite of this a substantial denitrification has taken place. The reason of this could be a conversion of dissolved COD into particulate COD (heterotrophics) in the nitrification reactor. Therefore this particulate COD would act as an additional internal carbon source.

The overall COD elimination reaches 60 to 90% of the influent COD (filtrated). This corresponds to an effluent between 48 and 80 mg/L, which has to be stabilised and reduced by optimisation of the denitrification process to meet strong regulations.

### Energy analysis

The dissolved oxygen in the nitrification has been controlled around 4 mg/L at a volumetric aeration rate of 35 m<sup>3</sup> air per m<sup>3</sup> reactor volume and hour. This is equal to 17.5 m<sup>3</sup> air per m<sup>3</sup> settled sewage. Based on the energy consumption of the compressor of about 5 Wh per m<sup>3</sup> air this would result in 87.5 Wh per m<sup>3</sup> sewage corresponding to 7.8 kWh per p.e. and year. Unaerated reactors for the denitrification need about 2.4 kWh per p.e. and year (see Müller *et al.*, 1999) including sludge return. Therefore, the overall energy consumption for biological sewage treatment could be reduced by 38% down to 10.2 kWh per p.e. and year. Overall energy consumption inclusive filtration (2 kWh/p.e.\*a), anaerobic sludge stabilisation and dewatering (2.7 kWh/p.e.\*a), pre-treatment inclusive delivery lift of 3 metres (1.9 kWh/p.e.\*a), infrastructure (1.6 kWh/p.e.\*a) would result in 18.4 kWh/p.e.\*a.

Under consideration of energy recovery through conventional biogas production of about 13 kWh/p.e.\*a, the overall energy consumption would be 5 kWh/p.e.\*a. Note that additional biomass production by reduction of SRT down to less than 3 days has not been taken into consideration.

### Conclusions

Continuously lab scale experiments with encapsulated nitrifying micro-organisms have shown a complete nitrification at a hydraulic retention time of 30 to 60 minutes for more

than 650 days. This is approximately 10 times lower compared to conventional sewage treatment plants. The volumetric reaction rates vary mainly between 50 to 60 mg/l\*h.

The proposed pre-nitrification process followed by high loaded denitrification seems to be possible by internal use of wastewater carbon sources for denitrification by a nitrate removal efficiency up to 50–60% due to low COD removal in the nitrification reactor. However, the denitrification process has to be optimised concerning kinetic study and avoidance of sludge flotation in final settler.

The energy consumption for aeration could be reduced substantially by 38% based on the measurements. This would lead to an overall energy consumption for advanced sewage treatment (complete phosphorus and nitrogen removal) of 5 kWh per p.e and year. With respect to the possibility of additional production of biogas through reduced SRT and less oxidation of COD into carbon dioxide, a self sufficient operation of advanced sewage treatment plants seems reachable by further optimisation of both the aerobic sewage treatment and the anaerobic sludge treatment.

The results have to be confirmed at pilot scale under realistic sewage treatment conditions onsite. It is expected that the efficiency of the process would decrease, but on the other hand, there is a high potential for optimisation of more selective LentiKats<sup>®</sup> with higher reaction rates and better aeration conditions.

## Acknowledgements

The authors wish to express thanks to Mrs Petra Maslowicz, Mrs Carmen Kiefer and Mr Gerd Böhmert for their analytical assistance with the variability tests.

## References

- Brandenberger, H. and Widmer, F. (1997). A new multinozzle encapsulation/immobilization system, in Godia, F., Poncelet, D. (eds.) *Proceedings of the International Workshop Bioencapsulation VI*, Barcelona, poster 9.
- Jetten, M.S.M., Strous, M., Van de Pas-Schoonen, K.T., Schalk, J., Van Dongen, U.G.J.M., Van de Graf, A.A., Logeman, S., Muyzer, G., Van Loosdrecht, M.C.M. and Kuenen, J. (1999). The anaerobic oxidation of ammonium, *Microbiol. Rev.*, **22**, 421–437.
- Leenen, E.J.T.M., Dos Santos, V.A.P.M., Tramper, J. and Wijffels, R.H. (1996). Characteristics and selection criteria of support materials for immobilization of nitrifying bacteria, in *Immobilized Cells: Basics and Applications*, Wijffels et al. (eds.) Elsevier Sciences B.V., 205–212.
- Lozinsky, V.I. (1998). Cryotropic gelation of poly(vinyl alcohol), *Russian Chemical Reviews, English Edition*, **67–7**, 573–586.
- Lozinsky, V.I. and Plieva, F.M. (1998). Poly(vinyl alcohol) crygels employed as matrices for cell immobilization. D. Overview of recent research and developments, *Enzyme Microb. Technol.*, **23**, 227–242.
- Müller, E.A., Kobel, B., Künti, T., Pinnekamp, J., Seibert-Erling, G. and Böcker, K. (1999). *Energy in wastewater treatment plants* (in German), Handbook, MURL, Düsseldorf.
- Muscat, A., Prübe, U. and Vorlop, K.-D. (1996). Stable support materials for the immobilization of viable cells, in *Immobilized Cells: Basics and Applications*, Wijffels, R.H., Buitelaar, R.M., Bucke, C., Tramper, J. (eds.) Elsevier Sciences B.V., 55–61.
- Onyche, T.I. (1999). Mechanical disruption and anaerobic digestion of conventionally stabilised sewage sludge, PhD thesis, TU Clausthal, Germany.
- Onyche, T.I., Schlaefler, O. and Sievers, M. (2002). Improved energy recovery from waste sludge, Accepted, ENVIRO 2002, Waste conference, Melbourne, Australia.
- Prübe, U., Bruske, F., Breford, J. and Vorlop, K.-D. (1998). Improvement of the Jet Cutting method for the production of spherical particles from viscous polymer solutions, *Chem. Eng. Technol.*, **21**, 153–157.
- Schmidt, I. and Bock, E. (1997). Anaerobic ammonia oxidation with nitrogen dioxide by *Nitrosomonas europaea*, *Arch. Microbiol.*, **167**, 106–111.
- Schmitt, F., Klauwer, E. and Feckler, H. (1998). Examples for reduction of energy consumption on wastewater treatment plants (in German), *WAR serial report 108*, Darmstadt, Germany.

- Sievers, M., Vorlop, K.-D., Hahne, J., Schlieker, M. and Schäfer, S. (2002). Advanced nitrogen elimination by encapsulated nitrifiers (accepted), Environmental biotechnology 2002 conference, Palmerston, New Zealand.
- Tanaka, K., Sumino, T., Nakamura, H., Ogasawara, T. and Emori, H. (1996). Application of nitrification by cells immobilized in polyethylene glycol. *Immobilized Cells: Basics and Applications*, Wijffels, R.H. et al. (eds.) Elsevier Sciences, 622–632.
- Twachtmann, U. and Metzger, J.W. (1998). A novel concept for the treatment of the effluent from anaerobic sludge digestion with trickling filters. Proceedings of European conference on new advances in biological nitrogen and phosphorous removal for municipal or industrial wastewaters, INRA, Narbonne, France, 373–377.
- Vorlop, K.-D. and Klein, J. (1983). New developments in the field of cell immobilization: formation of biocatalysts by ionotropic gelation, in Lafferty, R.M. (editor) *Enzyme Technology*, Springer Berlin, 219–235.
- Vorlop, K.-D. and Breford, J. (1994). German Patent DE 4424998.
- Vorlop, K.-D. and Klein, J. (1985). Immobilization techniques, in Moo-Young (editor) *Cells in Comprehensive Biotechnology 2*, Pergamon Press, 203–224.