Six years' practical experience with aerobic/anoxic deammonification in biofilm systems

A. Hippen, C. Helmer, S. Kunst, K.-H. Rosenwinkel and C.F. Seyfried

Institute for Water Quality and Waste Management, University of Hannover, Welfengarten 1, D-30167 Hannover, Germany

Abstract Nitrogen elimination through autotrophic micro-organisms is currently in the focus of research projects on the treatment of wastewater with high nitrogen contents, for instance to be able to develop and fix dimensioning parameters for purposeful application. In fact, several industrial plants have already shown for some years that the steady operation of nitrogen elimination without carbon demand is possible. Due to the low growth rates of the participating micro-organisms, these processes can be found in particular in biofilm systems, which also allow for the simultaneous running of the two basic processes.

In the following, we will discuss on the basis of the operation results of industrial and pilot-scale plants the operation stability of the aerobic/anoxic deammonification, and explain which experiences are available in particular for conversion in biofilm systems.

Keywords Deammonification; biofilm; nitritation; anaerobic ammonium oxidation

Introduction

Biological methods for the treatment of wastewater with high nitrogen loads are facing the problem that the law requires extensive or even complete elimination of the nitrogen in regard to the denitrification, due to the fact that these wastewaters often are riddled by an extremely unfavourable COD/N ratio. COD/N ratios below 4 increase the denitrification volume by a factor of 1.5 to 1.7; with a ratio below 2.5, sufficient denitrification cannot be achieved at all without the use of external carbon sources. This is where new technological developments come into their prime which on the one hand reduce the reaction volume through compact technology or which on the other hand shorten the way to complete degradation of the nitrification/denitrification. For instance, already at the beginning of the 90s Abeling and Seyfried (1992) pointed out that the inhibition of the nitrification up to the nitrite stage can be very useful because the denitrification can begin with the nitrite, which allows us to dispense with one step for either oxidation or reduction. In this way, up to 25% of the oxygen and 40% of the carbon demand can be saved.

There, the basis was well known metabolic processes of micro-organisms used for wastewater treatment. Now, however, the development of "new" ways comes more and more in the focus of scientific interest. In regard to the treatment of wastewater with high nitrogen loads, methods like ANAMMOX or deammonification are discussed ever more frequently, because there autotrophic micro-organisms are relevant, which accordingly makes for even higher carbon saving potentials. If the method operations and control are selected with dexterity, it is also possible to reduce the oxygen demand. Against this background, biofilm methods, which contribute to an economically efficient wastewater treatment anyway through their compact technology, are the method of choice (Seyfried *et al*., 2001). Several different practical examples show the advantages and disadvantages of the "new" conversion ways and the frame conditions which must be considered in the single cases.

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Basics: aerobic/anoxic deammonification in biofilms

In the reference literature, there are plenty of reports on so-called nitrogen losses, that is, high nitrogen elimination rates due to insufficient carbon supply (Binswanger *et al*., 1997, Helmer and Kunst, 1998, Hippen *et al*., 1997, 1998). Hitherto, however, with these analyses of mixed biocoenoses in practically relevant wastewater treatment plants the question remained which reactions are responsible for the observed nitrogen deficit and which micro-organisms take part in the process. Different possibilities were researched and initially considered under the term "(aerobic) deammonification" (Hippen *et al*., 1998). In the meantime, however, more exact results have become available, so that the term aerobic/anoxic deammonification which is used now can be defined as a combination of preceding nitritation and succeeding oxidation of the remaining ammonium with nitrite as electron recipient (Helmer *et al*., 1999, 2001). The second stage of this reaction sequence was postulated for the first time by Broda (1977) on the basis of thermo-dynamic considerations; it has since then been proved as the so-called ANAMMOX process. (Mulder *et al*., 1995, van de Graaf *et al*., 1997). The unequivocal relevance for wastewater treatment in contrast to other options of nitrogen metabolism has been documented by van Loosdrecht and Jetten (1998).

For the utilisation of the ANAMMOX process in wastewater technology, Jetten *et al*. (1997) propose a preliminary nitritation stage in suspended biomass (SHARON; Mulder and van Kempen, 1997), followed by the ANAMMOX conversion in a fluidised bed system. Yet, the industrial and semi-industrial results presented here and in more detailed analyses (Helmer *et al*., 1999, 2001) show that at low oxygen concentrations an ammonium elimination can be achieved even in only one reactor when biofilm technology is used. It can be stated that at low dissolved oxygen (DO) concentrations ammonium will be nitrified into nitrite in the outer aerobic layers, which thus produces the second reaction partner for an anoxic ammonium oxidation.

Operational results and discussion

Full-scale (leachate treatment in RBC-systems)

The first example of aerobic/anoxic deammonification having been run for seven years without any operational problems is the biological pre-treatment unit of the leachate purification plant at Mechernich/Germany, where a multi-stage operation concept is realised (Seyfried and Weber, 1987, Baumgarten and Seyfried, 1996, Hippen *et al*., 1999). Two other full-scale examples of – more or less – stable nitrogen losses are the leachate treatment plants in Kölliken/Switzerland (Binswanger *et al*., 1997, Siegrist *et al*., 1998) and Pitsea/UK (Knox, 1991). The flow sheets of the biological steps are shown in Figure 1 to Figure 3.

The plant at Mechernich has been running completely since 1994; in the meantime, it has been extended by a secondary denitrification unit and a moving bed plant parallel to the existing rotating biological contactor plant in order to effect nitrification. This last construction stage was started at the end of 1998, with nitrogen losses being observed after a short time here as well (Hippen *et al*., 1999). For the following comparison with the other industrial plants, however, we will mainly consider the results of the rotating biological contactor plant. For the entire leachate treatment concept, please see Hippen *et al*. (1999).

The plant at Kölliken is a leachate purification plant belonging to a dumping ground for hazardous waste. In contrast to the plant at Mechernich, where a styrofoam contactor system of the Stengelin company is used, in this plant the rotating contactors (Hipur) of the Mecana company are operated. The flow sheet (Figure 2) immediately makes apparent that in the nitrification stage only insufficient COD for a possible nitrogen elimination can be made available, due to the fact that this stage follows a COD elimination (BC1) and

activated carbon filtration. This plant was initiated in 1995; in the following, however, we will consider only the operation period since 1997 because then the nitrification stage (BC2) was equipped with new rotating contactors.

The leachate purification plant at Pitsea was built in 1985 and initially operated with three parallel rotating contactors (corrugated segments made of polypropylene, which are assembled on an axis to form discs). The extension by two further units with 15,000 m^2 each was done in 1991 (data basis here: since 1992). Here, the leachate is first conveyed through a planted ditch system (this was previously a swamp area), where the first conversion of the nitrogen parameters take place. The purification target of the entire concept is the complete nitrification. In contrast to the nitrification stages of the two other plants (Mechernich: 3 identical cascades each in 4 parallel lines; Kölliken: 2 cascades with 5,225 and 2,090 m^2, respectively), here the leachate flows parallel through all units.

Table 1 shows the comparison of the operation parameters and the leachate data (influent to the nitrification stage) of the single plants.

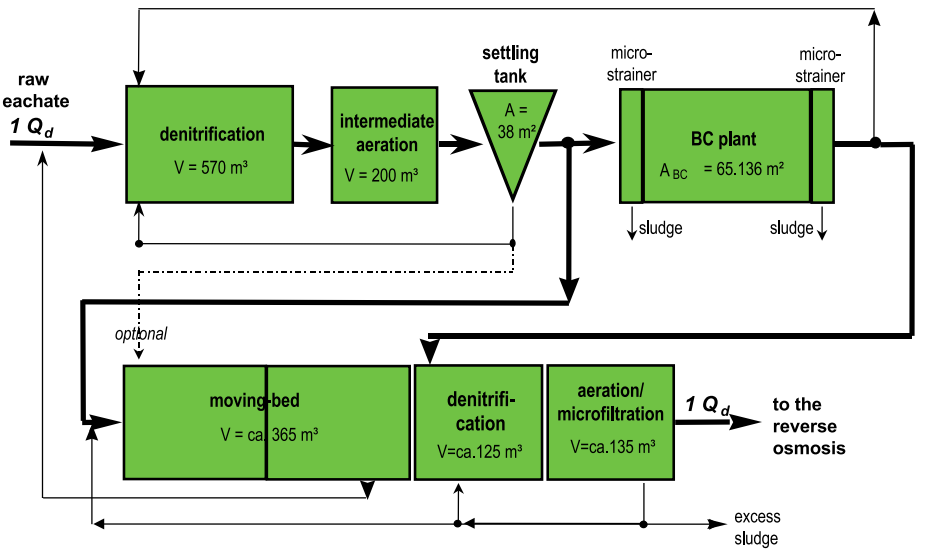

Figure 1 Flow sheet of the biological pre-treatment plant in Mechernich/Germany

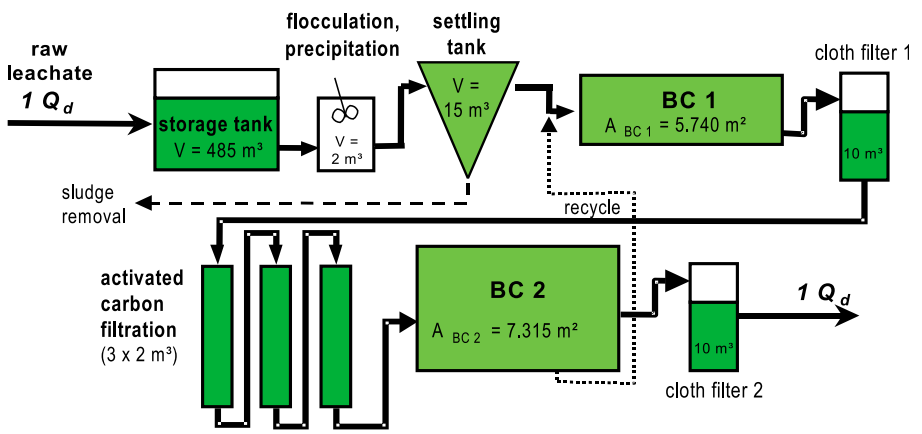

Figure 2 Flow sheet of the biological treatment plant in Kölliken/Switzerland

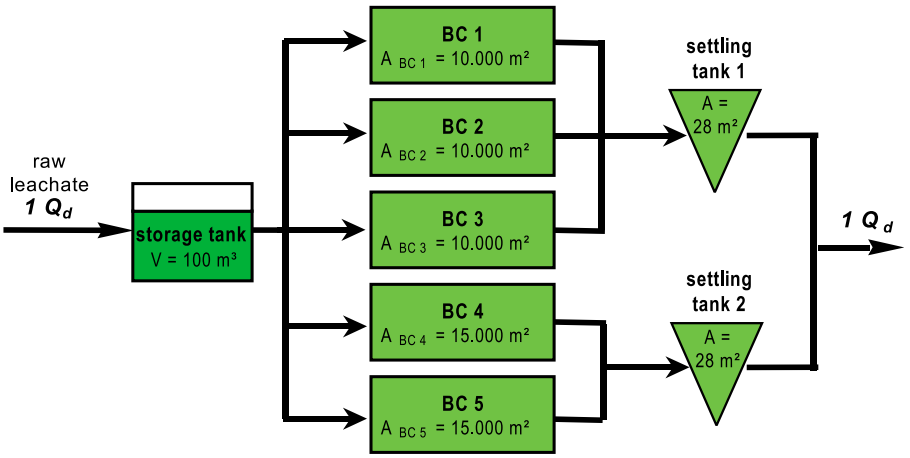

Figure 3 Flow sheet of the biological treatment plant in Pitsea/UK

Table 1 Operation parameters and characteristics of leachate in the influent of the nitrification step

		Mechernich	Kölliken	Pitsea
water quantity[1]	m^3/d	102 (0–182)	81 (12–197)	457 (0–915)
Temperature	°C	28 (27–30)[2]	16 (13–20)	14 (10–28)
pH	–	8.3 (7.4–8.7)	7.3[3]	8.1 (7.2–8.8)
DO[4]	mg O_2/l	0.7–1	1–2	0.8–1.2
NH_4–N	mg N/l	209 (32 – 681)	206 (18–494)[5]	423 (147–780)
CSB	mg O_2/l	1,645 (442–2,900)	–	1,000 (748–1,593)
TOC	mg C/l	310 (216–412)[6]	10 (5–20)	400 (253–651)
$B_{A,NH4-N}$	g N/m^2d	1.5 (0.5–2.6)	2.7 (0.7–7)	3.3 (0.3–6.3)
$DB_{A,NH4-N}$	g N/m^2d	1.5 (0.5–2.6)	2.5 (0.5–5.4)	3.2 (0.3–6.2)

[1] flow without recycle
[2] almost constant through usage of heat exchangers
[3] constant through the usage of Na_2CO_3
[4] values of the cascades with deammonification
[5] target range: 100–150 mg/l through purposeful recycle guidance
[6] data of the first half of 1994

The balances of the nitrogen compounds in the nitrification stages of all rotating biological contactor (RBC) plants show that apart from an almost complete nitrification there has for several years been a considerable nitrogen elimination, which for the single plants moves in a range between 40 and 70% in regard to the influent load. A possible ammonia stripping or N_2O production are not the reason for these amounts of eliminated nitrogen, and the ratio of the nitrogen load incorporated in the biomass is also not the explanation for these nitrogen losses. Moreover, in all cases there was only a negligible degree of carbon elimination; thus, it is apparent that mainly autotrophic micro-organisms must be responsible for these conversions (Helmer *et al*., 1998, Hippen *et al*., 1998). Using extensive experiments run as batch, ^{15}N-marking, and FISH-(fluorescence-*in-situ*-hybridisation) experiments, Helmer *et al*. (1999, 2001) have meanwhile succeeded to define the aerobic/anoxic deammonification as the reaction mechanism responsible for the conversions found at Mechernich (as well as in the pilot-scale plant described in the following).

In Figure 4 to Figure 6, the respective nitrogen loads eliminated in the nitrification stages are related to the nitrogen loads in the influents (considering the respective recycle streams). It becomes apparent that – especially at Mechernich – major parts of the nitrogen are eliminated via the aerobic/anoxic deammonification in a very steady manner and following the variations of the influent load. In contrast, at Kölliken the nitrogen losses

are at times very low or varying considerably. When looking for significant differences between the operation conditions at Mechernich (or Pitsea) and Kölliken, Table 1 clearly shows that there were differences in regard to the measured O_2-concentrations and the pH-values.

In regard to the oxygen ratios, it must be mentioned that differences of the measured concentrations in the surrounding medium (which for rotating contactors is also the air supplier during the surfacing phase!) do not allow for unequivocal statements, because for the biofilm systems discussed here only the oxygen diffusion depth is of any relevance. This depth depends on the DO concentration in the surrounding medium and on the biofilm thickness. Any exact comparison of the oxygen ratios would thus only be possible for the examination of the oxygen concentration profiles in the respective biofilm. Such a comparison, however, is only possible with much effort or only to a limited degree, anyway – in the present case, the statement that in all plants there are limited O_2-conditions must suffice as explanation. It is important is that in all three plants there are both aerobic conditions in the outer biofilm layers and anoxic conditions at the core of the biofilm. Batch tests have shown that deammonification consisting of nitritation and anoxic ammonium elimination takes place in all plants, so that it is safe to assume that the described conditions are actually valid.

An explanation for the differences in regard to a stable deammonification performance seems to be found rather through the examination of the pH-conditions. Whereas at Kölliken purposeful setting keeps the pH-value at about 7.3, in the other two plants

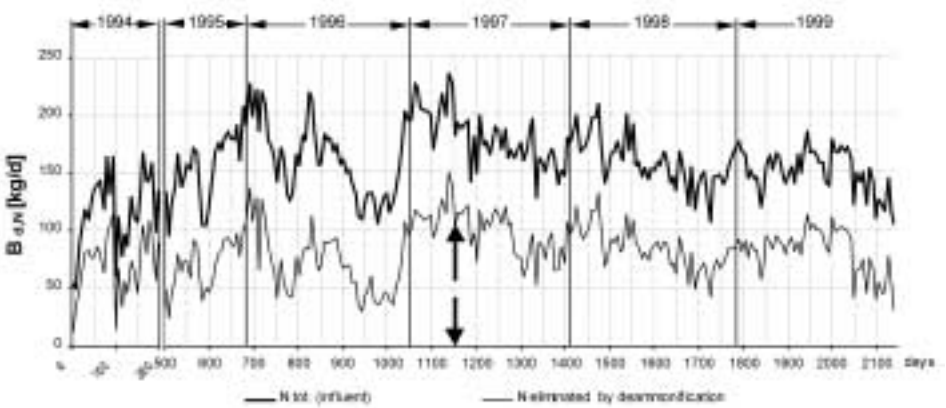

Figure 4 Presentation of influent and eliminated nitrogen loads in the RBC plant of Mechernich

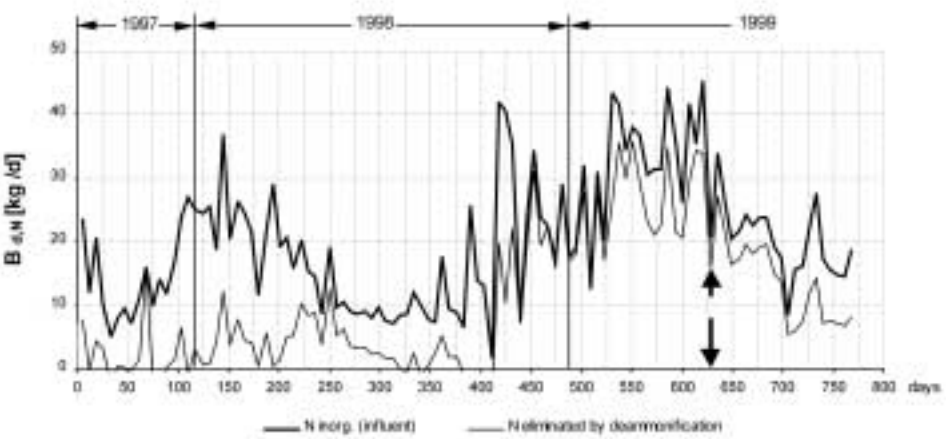

Figure 5 Presentation of influent and eliminated nitrogen loads in the RBC plant of Kölliken

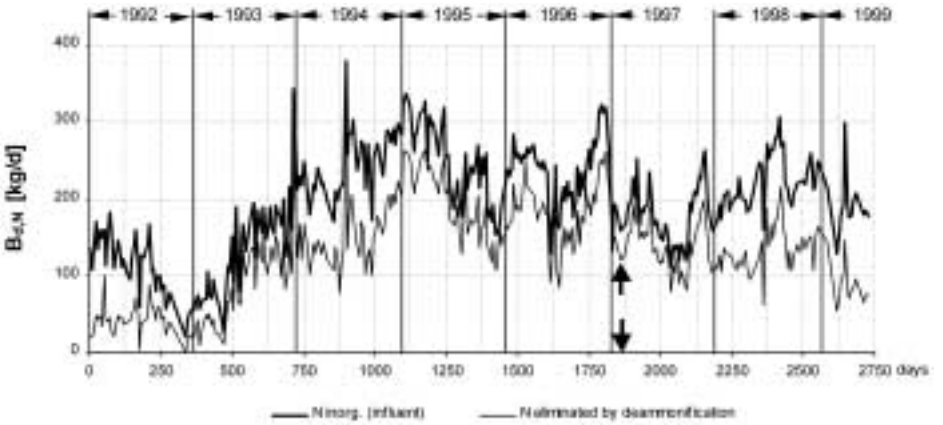

Figure 6 Presentation of influent and eliminated nitrogen loads in the RBC plant of Pitsea

pH-values > 8 are the rule. When we consider in addition that the first step of deammonification must be the nitritation, it is safe to assume that the ammonium flow and the nitratation inhibition resulting thereof are of particular importance. And indeed, this relation can be proved by examining the ammonia concentrations and the nitrogen losses measured simultaneously (given as deammonification rate, that is in relation to the ammonium nitrogen converted in the RBC plant). This is exemplified by selected operation periods for Mechernich, one unit (BC5) in Pitsea and Kölliken (see Figure 7 to Figure 9).

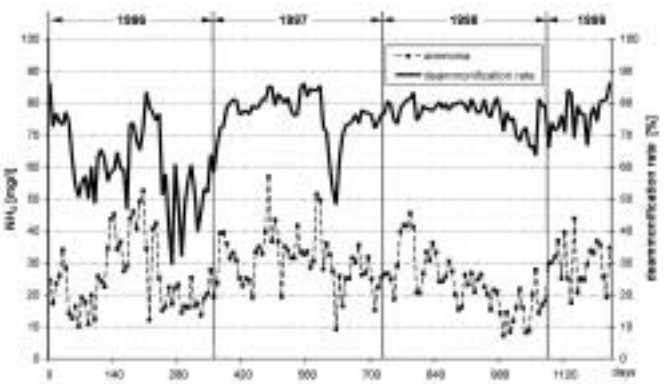

Figure 7 NH_3 concentration and deammo-nification rate in Mechernich

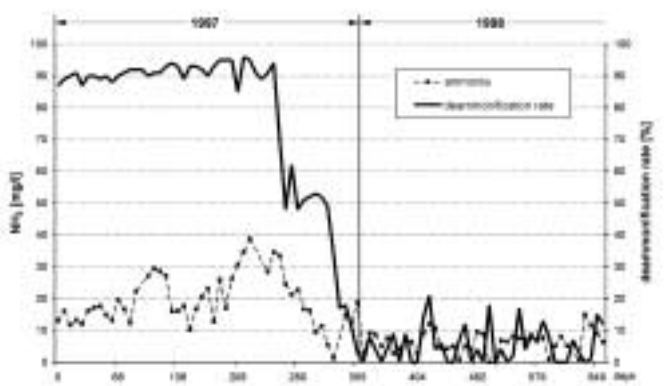

Figure 8 NH_3 concentration and deammo-nification rate of BC5 in Pitsea

Figure 9 NH_3 concentration and deammonification rate in Kölliken

Generally, it can be stated – as could be expected from the pH-conditions – that the ammonia concentrations at Mechernich and Pitsea move on a much higher level than at Kölliken: in these two plants, the concentrations range from 10 to (respectively) 40 or 60 mg NH_3/l; at Kölliken, however, the range is from 0 to 2.5 mg NH_3/l. The biofilm at Mechernich (and Pitsea) shows a very high adaptation to high ammonia contents, but also a noticeable connection between inhibition or high ammonia concentrations and nitrogen loss: at concentrations >20 NH_3/l mg, the inhibition is to be maintained; if the concentrations drop to 10–15 mg NH_3/l, however, there also occur drops of the deammonification performance. The same is the case at Pitsea with a limit at 10 mg NH_3/l for constant deammonification. At Kölliken, however, there is only a low ammonia inhibition due to the pH-control, so that in this respect also the mixed biocoenosis must be defined as non-adapted or as unstable in regard to the deammonification performance. Accordingly, the deammonification rate is subject to high variations.

Pilot-scale (sludge liquor treatment in a moving-bed system)

In pilot-scale tests on sludge liquor treatment, the aerobic/anoxic deammonification in a moving-bed system has been examined. A two-line laboratory plant was operated, with the two lines being constructed almost identically, but operated differently. At the core of both lines, there were two in-line reactors which were each filled to 20% with Kaldnes growth carriers. At reactor filling volumes of 40 l each and a specific surface of the growth carriers of $500 m^2/m^3$, the material filling degree of 20% leads to a growth surface of $4 m^2$ per reactor. For the detailed test construction and crucial test results, please see Hippen *et al.* (1999).

The basis of the following evaluations are the operation results of the so-called nitritation line, where through purposeful setting of the operation parameters it was possible to establish the deammonification (Hippen *et al.*, 1999). This was reached on the basis of a nitratation which was examined in detail in the first phase (period 1). The ensuing operation stages (periods 2 and 3) were to serve for the evaluation of the operation stability and the efficiency of the deammonification in the moving-bed method dependent on the respective operation control. The central question was to what degree a thin biofilm, as is normally found on moving-bed growth carriers, allows for the "single-stage" realisation of both processes. The crucial parameter here is the oxygen concentration, the variation of which can be used for a directed control of the biological conversion of the nitritation and the deammonification.

Table 2 gives a survey over the operation conditions during the single test stages. Period 1 is the period of nitritation, that is, the period when at sufficient oxygen supply in

Table 2 Operation parameters (average values) of the pilot moving-bed plant

		period 1 (9/97–5/98)	period 2 (6/98–1/99)	period 3 (3/99–9/99)
Temperature	°C	25.1	28	29.1
pH (reactor 1)	–	8.1	8	8.1
$NH_{4-}N_{infl}$	mg N/l	196	188	192
$B_{A,NH4-N}$	g N/m^{2}d	4.9	4.7	4.8
DO reactor 1	mg O_2/l	5.9	0.8	2
reactor 2	mg O_2/l	6.7	0.8	0.8

both reactors the step of nitratation is inhibited through pH-value regulation. As there occurred at times nitrogen losses during this period, it was then attempted to reduce the oxygen concentration in both reactors, to stabilise these losses and to maximise them (period 2). During this test stage, however, the ammonia conversion rates dropped considerably, and a further test stage was run with the target to separate the two processes which participate in the deammonification. In the first reactor, the oxygen concentration was increased, but then set to a value of approximately 2 mg/l in order to support the nitratation inhibition.

When we compare the conversion performances during these phases and differentiate according to which ratio of the ammonium load could be nitrified or deammonified, respectively, the numbers in Table 3 result.

The conversion performances during the single test stages make it obvious that a minimum oxygen concentration must be available so that ammonium can be subjected to a conversion or oxidation to a significant degree in the first place. For instance, the conversion rates in Period 1 still range around approximately 60%; at low contents, however, they drop during Period 2 to below 30%. It is possible in this phase to stabilise and maximise the nitrogen losses, but the separation of the processes (nitritation and anoxic ammonium oxidation) to two reactors is more efficient, as can be seen from the conversion performances during Period 3. The biofilm thickness degrees found on the growth carriers – between approximately 100 and 500 µm – are on the one hand not sufficient to serve as habitat for both the ammonium oxidisers and the so-called ANAMMOX organisms; on the other hand, at the selected oxygen concentration of 0.8 mg/l the diffusion depth is too shallow to realise (in the first step) any significant oxidation processes.

Conclusions

In regard to the treatment of wastewater with high nitrogen loads, the aerobic/anoxic deammonification is a highly promising method. In the meantime, much information has been compiled about the actually running processes, but there are still quite a lot of open questions about realising the optimal operation control, in particular the questions whether separated or simultaneous process guidance of nitritation and ensuing ammonium

Table 3 Efficiency and ways of nitrogen removal during each period of the pilot moving-bed plant

		period 1(9/97–5/98)	period 2 (6/98–1/99)	period 3 (3/99–9/99)
$hNH_{4-}N$	%	59	27	63
$\Delta B_{A,NH4-N}$	g N/m^{2}d	3	1.6	3.1
Nitritation	%	53	19	10
	g N/m^{2}d	*1.6*	*0.3*	*0.3*
Deammonification	%	10	75	71
	g N/m^{2}d	*0.3*	*1.2*	*2.2*
Nitrification	%	37	6	19
	g N/m^{2}d	*1.1*	*0.1*	*0.6*

oxidation is more eligible, which biofilm method is the most suitable, or which method for purposeful nitrite enrichment should be chosen.

These questions can be answered by purposefully run pilot-scale test series, but also experiences on an industrial scale. In this report, three industrial leachate purification plants have been presented, all of which have for several years shown that the autotrophic nitrogen elimination happening simultaneously in the biofilm can be realised both in rotating biological contactor plants and moving-bed systems.

The results from the industrial plants make it apparent that in particular the rotating contactor method is very suitable for the single-stage deammonification in the biofilm. The biofilm thickness of 1 and 2 cm produced in cases of such high loads allows for the necessary separation into outer aerobic and inner anoxic zones, so that there is sufficient habitat for both the ammonium oxidisers and the organisms for anoxic ammonium oxidation. Deammonification rates of 60–80% in relation to the amount of converted ammonium are normally the precondition to realise the stable nitritation or high ammonia contents. Thus, what is favourable are high loads, high pH-values, and approximately limited oxygen concentrations.

The optimal realisation of deammonification in the moving-bed method depends on several factors, with the biofilm thickness gaining particular importance. In contrast to the rotating contactor method, there are considerably higher shearing powers on an extremely small area, so that depending on the carrier material maximum biofilm thickness of 500 µm could be achieved. There are layerings into aerobic and anoxic zones at lower thickness degrees (Schramm *et al*., 1997), but here one has to additionally consider the impact of the oxygen concentration. In the moving-bed method, this parameter must be set purposefully, with the optimal value being defined through the oxygen diffusion depth and thus through the thickness of the biofilm. The purposeful control of the biofilm is necessary, in contrast to the rotating contactor method, because a thin biofilm will not necessarily offer sufficient habitat for two different kinds of organism. In this case, the separation of the processes of nitritation and anoxic ammonium elimination seems to be definitely more efficient, as can be seen from the pilot-scale results presented here.

The concluding evaluation shows that the target of an economically efficient application of the autotrophic nitrogen elimination necessitates the availability of more exact knowledge of the biofilm processes, but even more the exact observation of wastewater treatment plants which do already run deammonification. This combination of scientific basics research and application-related examination of operation-technology connections will eventually facilitate the purposeful application of the aerobic/anoxic deammonification.

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