

# Challenges of a feasible route towards sustainability in environmental protection

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**Abstract** Anaerobic processes for treatment of low and high strength wastewaters and solid wastes constitute the core method in the natural biological mineralization (NBM) treatment concept. When adequately combined with the complementary NBM-systems and modern clean water saving practices in wastewater collection and transport, they represent a feasible route to sustainable environmental protection (EP<sub>sus</sub>), in essence even towards a more sustainable society. Despite the development and implementation of modern high rate Anaerobic Wastewater Treatment (AnWT-) systems and complementary innovative NBM-processes, the considerable progress made since the seventies in fundamental insights in microbiology, biochemistry and process technology, still numerous challenging improvements in the NBM-field can be realized. This contribution is mainly based on the insights attained from wide ranging literature evaluations and the results of experimental research conducted by numerous PhD students who participated in our group over the last four decades. An attempt is made here to identify major facets on which an improved insight can, and consequently should, be obtained in order to accomplish more optimal operation and design of various types of Anaerobic Degradation (AnDeg-) processes.

**Keywords** sustainability, environmental protection, anaerobic treatment, micro-aerobic treatment, natural biological mineralization concept, traces elements, macro-nutrients

## 1 Introduction

It would be fortunate if mankind could succeed in accomplishing a fully sustainable protection of the environment (EP<sub>sus</sub>) on a global scale. The habitats for flora, fauna and humans need to be protected through the

lowest possible input of resources, labor, and capital by using technologically and economically affordable, plain and robust concepts and technologies. Since practically all the tools required for the implementation of this vision lie ready ‘on the shelf’, it is an imperative to implement these EP<sub>sus</sub> concepts. There are certainly reasons for optimism. However, at the same time there are doubts as well. The principal question is “Are we capable of implementing them in time?”. And in the case that we are, then we need to ask “Is the extent of environmental deterioration already not too dramatic?”, and “can we get the mechanisms under control, which lead to excessive world population growth?”, and “what actions need to be put in place to mitigate extreme urbanization and the dramatic deterioration of living conditions, in, for example, rural areas?”.

More than 20 years ago the Brundtland Committee [1] emphasized the need to realize drastic changes in society. Severe threats in society need to be eliminated, such as continued prevailing extreme poverty, dramatic environmental deterioration, and the excessive growth of world population. ‘Sustainability in Society’ stands for “Absence of man-made scarcity with respect to all primary needs and prevention of any form of wasting”. Figure 1 depicts the high ambitions of what Brundtland meant by ‘Sustainability’. In all respects Brundtland goes much further than the superficial interpretations of the vast majority of contemporary political leaders, multinational enterprises and many scientists. In essence Brundtland’s message is that scientific, technological and cultural achievements of mankind should be made available for all world citizens without any restrictions.

Is it an almost hopeless mission to achieve sustainability in society? Not entirely. History teaches that drastic beneficial shifts occasionally occur when conditions in society suddenly become mature for change. This may happen as a result of any ‘success’ on the route towards greater sustainability, as for instance in the field of EP. Once citizens understand that such a progress indeed serves the well-being of all people, they will become eager

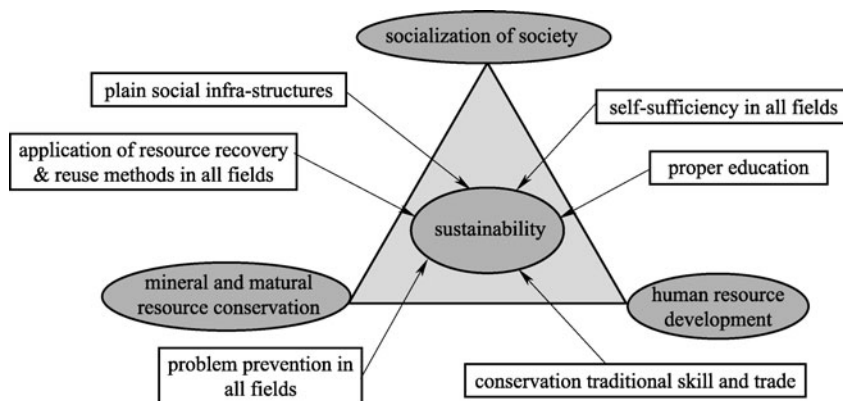


Fig. 1 Three 'pillars of sustainability' in society and some tools to achieve it

to force political leaders to initiate drastic changes. And one of the more crucial needs for the quality of life of citizens undoubtedly comprises a clean and secure living environment for all people, i.e.,  $EP_{sus}$ . In order to make progress in achieving the high goals of  $EP_{sus}$  the focus needs to be put on i) 'Pollution Problem Prevention ( $P_3$ )' and ii) the achievement of the recognition of a maximum of valuation of domestic residues ('wastes') and optimal reuse of effluents (treated wastewater) from treatment systems. As a result of preventing the waste of clean water in the collection and transport of waste (water), and in order to achieve the valuation issue, conventional treatment systems need to be substituted by systems based on the Natural Biological Mineralization (NBM) route [2,3].

Anaerobic Wastewater Treatment (AnWT) processes comprise the core method in NBM-treatment concepts, because i) they offer the unique advantage of realising a maximum recovery of resources from wastes (e.g., energy and nutrients), while ii) at the same time they lead to substantial savings of fossil energy, compared with conventional waste treatment technologies. Consequently, in order to achieve progress toward  $EP_{sus}$  we need to substitute conventional 'Centralized Sanitation' concepts (CENSA) for modern "Decentralized Sanitation and Resource Recovery & Reuse" concepts, utilizing the very promising DESAR<sub>3</sub>.

## 2 Features of a sustainable environmental protection ( $EP_{sus}$ )

The  $P_3$  and  $R_3$  pillars of  $EP_{sus}$ , imply the focus of the  $EP$ -measures to be taken in society merely on tackling pollution problems, which originate from the inevitable natural disasters, e.g., earthquakes, floods, storms, future drastic changes in climate, etc. In combating environmental pollution, exclusive use should be made of robust concepts and technologies; history teaches that disastrous environmental damage generally originates from inevitable natural disasters and/or from all kinds of man-made

catastrophes, frequently due to the collapse of far too vulnerable  $EP$ -facilities or their destruction on purpose, e.g., in military events. Therefore, robust methods of environmental protection are required, which neither rely on advanced and expensive transport and treatment technologies, nor on complex infrastructural provisions, such as the power supply and highly specialized technicians or institutions.

In nature, wastes do not exist; almost everything is part of the eternal life cycle. It would be wise to imitate nature, particularly in the protection of the environment, for example by closing water and material loops through using the economically most affordable and sustainable technological and conceptual means, all directed on waste valorization. When realized at on-site and/or at regional level, it will enable communities, towns, villages, cities and regions, and even countries, to accomplish a maximum extent of self-sufficiency in  $EP$ ; this then will have a very positive spin-off in other fields. Such an  $EP_{sus}$ -tackle obviously requires a holistic attack of the 'real' problems. In order to achieve that, all presently available relevant knowledge needs to be transferred free of charge to potential users in order to enable them the implementation of the required systems/concepts properly. Then, concomitantly, all kinds of challenging opportunities will evolve in other fields, for instance in local (urban) agricultural practices. In this way, step by step a more sustainable society will be attained, enabling all citizens to employ presently available technical, scientific and cultural achievements. As a result humanity can ultimately release itself from serious threats from the obscure past such as its immense social insecurity, a disease inherited from the past! However, all kinds of established groups, institutions, etc., generally are extremely reluctant to abandon their privileged positions; this seriously frustrates the smooth implementation of the  $EP_{sus}$ -concepts. The latter is particularly true for the public sanitation sector. The main bottlenecks for implementation in this sector can be found in the lack of 'political' willingness and/or courage, absence of proper 'market incentives' and/or lack of a

‘well established logistic, educational and decision making infrastructures’. Moreover, despite the enormous potentials of the envisaged  $EP_{\text{sus}}$ -tackle, the industrialized world tends to look for highly advanced, generally expensive solutions, which leads to more rather than less dependency on specialists and vulnerable infrastructures like that of supply of energy; this virtually is the case for almost any sector in society. These developments are enhanced by a dramatic lack in social and entrepreneuring security in society. The question is how to escape from this threat.

### 3 NBM-route based waste (water) treatment tackles in EP<sub>sus</sub>

It is well known that under natural conditions the recuperation of polluted surface waters proceeds via a sequential anaerobic, micro-aerobic and fully aerobic biodegradation of the pollutants, on the basis of processes proceeding according to the biological C-,N and S-cycle, together with associated chemical and physical processes. As clearly demonstrated in ‘aging experiments’ with sewage samples [4] this Natural Biological Mineralization (NBM) route of organic matter, depicted in Fig. 2, ultimately leads to the complete recuperation of the polluted water. However, at the same time this technologically undemanding  $R_3$ -directed treatment concept for wastes and wastewaters enables recovery of useful by-products, provided that those responsible are capable of deploying them wisely. In essence the various NBM-treatment systems are available for use, although their microbiology and (bio) chemistry may be designated as

‘complex’, and certainly not fully understood yet. Perhaps some of them never will be understood fully either, but working with a partial ‘black box’ does not represent a real bottleneck for optimal applications. The NBM-treatment route meets practically all criteria for  $EP_{\text{sus}}$  [2].

Apart from the fact that NBM-treatment systems enable an almost complete removal of organic pollutants from the wastewater(s), they provide ideal conditions for conserving/recovering resources in the form of fertilizers, soil conditioners and renewable energy. This specific feature can be attributed particularly to the renowned potential of Anaerobic Degradation (AnDegr) steps, consisting of:

i) Anaerobic digestion (AnDi) processes, which convert biodegradable organic compounds into a mixture of methane and carbon dioxide,

ii) Sulfate reduction (SuRe) processes, which take care of the conversion of oxidized forms of sulfur into  $H_2S$ ,

AnDegr-processes constitute the ideal mineralization tackle for organic matter; they therefore should constitute the first treatment step. AnDi-systems for stabilizing very high strength wastewater(s) and/or slurries can consist of conventional low or high rate digesters, or innovative high rate digesters complemented with specific physical-chemical pre-treatment systems. For the treatment of medium and (very) low strength wastewaters, modern high rate anaerobic wastewater treatment (AnWT) systems have become available since the ninety-seventies; so far, generally they have found application mainly in ‘one-step’ reactor configurations. Nowadays, the most popular AnWT-systems comprise stationary upward or downward flow Anaerobic Filter (AF) systems, (re)introduced in the sixties in the USA [3,5], and Upflow Anaerobic Sludge

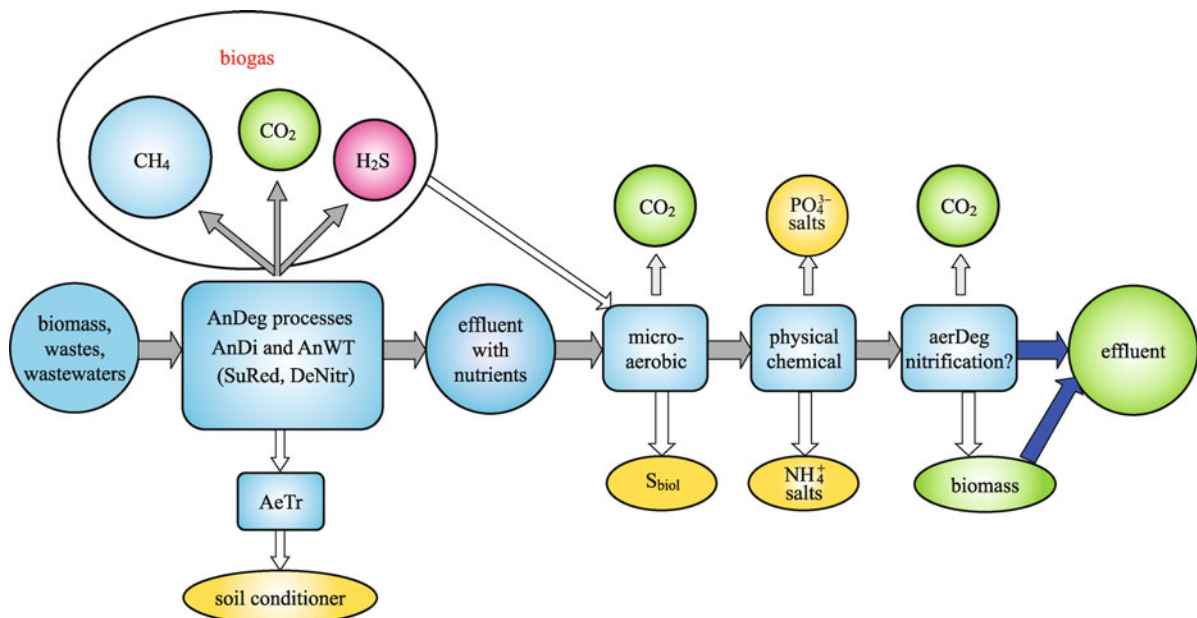


Fig. 2 The natural biological mineralization (NBM) route as treatment concept for waste and wastewater valorization

Bed reactors (UASB), developed in the early seventies in the Netherlands [6,7,8]. The very promising next developments are the Expanded Granular Sludge Bed (EGSB), developed in the Netherlands in the early eighties, and the Attached Fixed Film Expanded Bed (AFFEB) system, developed in the USA [9]. The Sequential Anaerobic Batch Reactors (SABR) and Baffled Anaerobic reactor systems [10,11], and various types of hybrid reactors may also undoubtedly hold out some promise.

These systems, particularly the UASB and EGSB-process [8], have found a wide application for a large variety of industrial effluents, although so far mainly under optimal mesophilic conditions. Since the late nineties these systems have been applied to the pre-treatment of raw sewage in a number of tropical countries. In the last decades insight into the reactor and process technology has improved significantly.

Compared to UASB-systems, EGSB-systems possess significantly better potential for removing toxic biodegradable compounds like lauric and capric acids [12–14], higher fatty acids [15–17], formaldehyde [18,19] and/or complex compounds present in paper pulping effluent [20]. This feature of the EGSB-reactor system which is very attractive for practice, can be attributed to i) the significantly better contact attained in EGSB-systems as a result of the high upward liquid velocities and ii) the low concentration of these compounds maintained in the reactor liquid phase as a result of the imposed high effluent (so-called ‘upfront dilution’) recycle factors. In view of the prevailing high substrate transport rates in EGSB-systems, they even are well suited for the treatment of very low strength wastewaters, i.e., down to about 200 mg COD/L, both under mesophilic and psychrophilic conditions [21–27]. Clear experimental evidence was obtained that the methanogenic activity of the retained biomass still increased under these extreme conditions, indicating that the wash-out of viable methanogenic organisms due to erosion of granules remained exceptionally low! The potentials of the EGSB-concepts therefore look exceptionally promising, provided the stability and the size of the sludge grains can be maintained in the optimal region. The latter condition certainly cannot always be met. Therefore, in specific cases, for example effluents containing triglyceride emulsions, a modified, more sophisticated, Gas-Solids Separator device needs to be installed in order prevent problems due to floating granular sludge [13]. Nevertheless, undoubtedly challenging developments for EGSB-reactor systems lie in front of us.

Although modern versions of high rate AnWT-systems only became popular starting in the seventies, the AF-system already was introduced at the Massachusetts experimental station almost 1.5 centuries ago [5]. The first full-scale AF-system, consisting of a bed of sand, was put in operation at that experimental station in 1887 for sewage treatment. In one of the AF-reactors, up to 89 %

removal of the organic impurities was obtained at a pore space detention time of about 8 days; in a second AF-system, using 0.5–2 inch diameter broken stone, 85% organic matter removal from domestic wastewater was achieved at a loading rate of about 2 m/d. However, contrary to ‘ancient’ AnWT/AnDi-systems of the septic tank and Imhoff tank type and despite the promising results obtained, the AF-system apparently could not conquer the required popularity. Although in essence all these ‘ancient’ AnWT and AnDi systems at that time were “black box reactor systems”, the vast majority of them probably performed reasonably satisfactorily, even despite the presence of volatile obnoxious compounds in the effluent. But undoubtedly many of them suffered from severe overloading. Consequently, they became unwelcome in the neighborhood. Nevertheless, despite their ‘bad smell’ and complete lack of understanding of the microbiology, and of technological aspects from the side of the sanitary engineering world, their use fortunately has never been abandoned. In many countries and regions these systems still have a very important role in environmental protection. Moreover, it is very likely that promising innovative versions will soon become available, because the insight into the microbiology, biochemistry and reactor technology of the anaerobic digestion process which has been improved very substantially over the last decades. The revival of these systems in modern DESAR<sub>3</sub>-concepts in the public sanitation sector lies ahead of us. For billions of citizens suffering from lack of any adequate sanitation this is a matter of the utmost importance. But undoubtedly this also will become true for prosperous citizens since everybody ultimately wants a robust and sustainable type of EP.

The major role of the micro-Aerobic Wastewater Treatment ( $A_{e_{micro}}WT$ ) systems is to accomplish the required first post-treatment, comprising particularly the removal of volatile mal-odorous compounds. These innovative methods constitute the ideal complementary treatment step to AnWT. The idea underlying  $A_{e_{micro}}WT$  is that by supplying minor amounts of oxygen in the anaerobic effluent, a ‘perfect’ environment is created for a variety of specific micro-aerobic organisms. The organisms will take care of i) the high-rate conversion of reduced (highly odiferous and toxic) S-compounds into elementary sulfur, ii) the removal of part of the remaining, easily biodegradable organic pollutants; iii) the oxidation of specific reduced inorganic compounds (e.g., FeII) and iv) the removal of colloidal matter (including dispersed pathogenic organisms, i.e., via a coagulation and/or bio-film entrapment process. Moreover, since the content of biodegradable matter in the effluent of the anaerobic (pre-) treatment step is low, the  $A_{e_{micro}}WT$  step pairs minor energy demands with a very low excess sludge production. According to the findings of Tawlik et al. [28–30] a one- and/or two-step micro-aerophilic Rotating Biodisk Contactactor (RBC-reactor) system is capable of removing

dissolved mal-odorous compounds and reduce inorganic compounds efficiently from an anaerobically pre-treated sewage at hydraulic retention time (HRT) values of only 15 min. However, the elimination of soluble biodegradable ( $\text{COD}_{\text{biodeg}}$ ) and colloidal matter, including indicator organisms for pathogens, needs more time, for example in the range of 30–150 min, depending on the quality of the effluent.

Undoubtedly, the major innovative development of the  $\text{Ae}_{\text{micro}}$ WT-system lies in its capacity to convert reduced S-compounds into elementary sulfur [31–37]; the system enables a high rate and low-cost bio-conversion of volatile S-compounds like  $\text{H}_2\text{S}$  and methanethiol from polluted gases, e.g., from Liquefied petroleum gas (LPG) [38–41] and from natural gas and/or biogas. The system is effective up to removal capacities of 50 tons  $\text{H}_2\text{S}$ /day; for higher loads tentatively the well-proven amine/Claus process still looks more attractive [40].

This manipulated oxidative section of the  $\text{S}_{\text{bio}}$ -cycle comprises, when properly combined with its reductive part, an attractive piece of equipment for the remediation of Zn-contaminated soils (already applied at full-scale) and soils likely to be contaminated with other heavy metals. The  $\text{S}_{\text{bio}}$ -cycle is also effective for the treatment of aqueous solutions (acid mine drain water) contaminated with heavy metals and for the treatment of  $\text{SO}_2$ -polluted air. The elementary biological-sulfur ultimately *recovered* in the process, and comprising one of the many allotropes of elementary sulfur, possesses peculiar features; it is hydrophilic and dispersible in water, can be used as fertilizer, fungicide and raw material in the industry; consequently, it will become an attractive by-product.

The remaining function for aerobic treatment ( $\text{AeWT}$ ) lies mainly in i) the polishing of the effluent of the  $\text{Ae}_{\text{micro}}$ WT step, i.e., the removal of the (very) small amounts of remaining biodegradable matter, and ii) as the nitrification step of dissolved ammonia, in case N-removal should be needed. In the latter case a complementary denitrification step also needs to be incorporated, or possibly better, the innovative systems Anammox process [42]. However, in case when sufficient land is available for agricultural purposes, it will be a much more valuable use of the dissolved nutrients to apply them as fertilizer, e.g. for cultivating biomass in algae or duckweed generating ponds [43,44].

As in the case of the treatment of wastewaters,  $\text{AeWT}$ -systems should be abandoned as the primary step for the ‘stabilization’ of solid organic wastes/residues; conventional composting systems do not lead to best use of solid organic residues. However, subsequent to a first AnDi-step, they can be quite useful as composting step. Regarding the merits of the NBM-concept it is obvious that old-fashioned sanitary engineering practices like land-filling need to be abandoned, due to potential pollution problems.

In order to achieve an optimal value for the pollutants, NBM-systems can be supplemented/complemented with proper physical-chemical (PC) removal/recovery steps, e.g., stripping-absorption processes for ammonia (and phosphate) recovery, precipitation processes for insoluble phosphate and ammonia-salts (struvite), membrane processes and UV-radiation in order to produce potable or process water.

The NBM-treatment concept, at some time in the future, will undoubtedly bring a definite end to episode of the ‘advanced’  $\text{AeWT}$  secondary treatment tackle and the ‘clean water wasting’ centralized sanitation (CENSA) approaches in the public sanitation (PuSan) sector. These conventional systems simply suffer from too numerous serious drawbacks [2], they are far too complex and too expensive, and for those reasons are often abandoned in developing countries soon after having been installed, with all the dramatic consequences for the environment! NBM-systems are simply by far superior (see Table 1–Advantages).

The established ‘advanced  $\text{AeWT}$  system’ is a heritage from the late nineteenth and early twentieth centuries; the first activated plants were put into operation in 1917 in Manchester and in Houston [11], and the first trickling filter was already established in 1893 in the UK; the system became particularly popular in USA. The  $\text{AeWT}$ -system superseded the promising house and community on-site AnWT pre-treatment approach introduced around 1900, possibly because of their complete ‘black box’ character.

**Table 1** Advantages of the NBM-WT route relative to conventional contemporary WT-practices

No.	advantages
1	Instead of being energy demanding, they generate energy.
2	Leading to ‘waste’ and water recovery, i.e., waste as a valuable resource, urban agricultural practices.
3	Very low production of—well stabilized and concentrated—excess sludge.
4	Very low space requirements.
5	Low in investment, operational, and maintenance costs.
6	Simple in operation and maintenance, consequently hardly depending on specialists.
7	Use of technically simple and generally locally manufacturable equipment.
8	Long life-time of installations and auxiliary equipment.
9	Applicable at almost any scale and at almost any location.
10	Very robust, e.g., absence of any need for any complex infrastructures, e.g., for power supply.
11	Almost absence of any mal-odor nuisance problems.
12	Formation of recalcitrant organic compounds (e.g., humic acids) can be avoided.
13	Efficient in degrading various resistant compounds like azo-dyes, poly-aromatic compounds (PAC’s), and nitro-aromatics.

#### 4 Challenging expected future operational developments in NBM-based treatment

Obviously, the first priority for achieving a more sustainable EP is to accomplish the required conceptual innovations (paradigm shifts) in the sector. However, this comprises a task of immense difficulty, because the interests of the established sanitary engineering world are generally opposite, i.e., particularly directed to pursuing further technological innovations within the well established CENSA-concepts and the further world-wide implementation of these systems. However, regarding the urgent need to realize more sustainability in society, all possible emphasis needs to be put on the implementation, optimal application and the further improvement of NBM-based systems. Despite the very significant progress already made, still much more can be achieved by improving the insights in the microbiology, ecology and (bio)chemistry and reactor- and process technology of the various NBM systems. As far as AnWT/AnDi systems are concerned, numerous challenging innovations can be accomplished. This certainly is not limited to modern high rate AnWT-systems for wastewater treatment and conventional AnDi-systems for slurry stabilisation and energy production [2,45], but also for so-called “outdated” systems, like septic tanks and latrines. And as far as AnDeg-processes in the wider context are concerned, particularly the  $S_{\text{biol}}$ -cycle based systems are open for creative innovations. Regarding NBM-systems, interesting innovation undoubtedly can be expected for both high rate  $Ae_{\text{micro}}$ WT-based post-treatment systems and  $Ae$ WT-based systems as well. The requirement for applying ‘post-treatment of anaerobic effluents’ is of a recent date. Furthermore, it is clear that there will evolve an increasing opportunity for nutrient recovery systems applicable for wastes, wastewaters and energy crops.

##### Process and operational technological improvements in AnDi and AnWT

Many improvements undoubtedly can be achieved by conducting a thorough evaluation of the enormous amount of relevant information available in literature. Unfortunately, literature search frequently receives far less priority than experimental research; it looks less prestigious. Nevertheless, still numerous issues only can be elucidated on the basis of comprehensive experimental investigations, and some of them, regarding their enormous complexity, even need a very well coordinated multidisciplinary attack over prolonged periods of time.

Below process and technological improvements in AnDi and AnWT will be discussed, viz. including operational temperature, high operational pressures, mixing conditions, occurrence of chemical precipitation, extreme salt concentrations, trace elements and macronutrients, use of electron and redox mediators, and biological sulphate reduction.

##### Operational temperature

The potentials of both psychrophilic and thermophilic AnDi and AnWT-systems are significant. It should be emphasized here that these systems offer the greatest potential when applied in their optimal physiological temperature ranges. However, it should be emphasized here that their application become especially useful under temperature conditions far below the optimal physiological operating temperature and, of greater importance, under varying temperature conditions as well. In this connection it should be understood that the required process-stability of AnWT-systems, when applied under whatever operational conditions, including sub-optimal, always needs careful adjustment of the imposed organic load.

The feasibility of high-rate psychrophilic EGSB-systems has been demonstrated for the treatment of volatile fatty acids containing wastewaters in bench scale experiments at temperatures down to 4°C[46]. Contrary to acidified substrates, direct application of high-rate AnWT-systems is not feasible for soluble carbohydrate wastewaters under psychrophilic conditions; a complementary acidogenic pre-treatment process step then needs to be incorporated in order to achieve the required pre-acidification [26]. However, such a complementary acidogenic reactor can probably be omitted when treating soluble protein containing wastewaters, because the yield of acidogenic sludge in that case is significantly lower than for carbohydrate substrates.

Direct application of a high rate AnWT-system under psychrophilic conditions also is not feasible for the stabilisation of insoluble organic matter; the very low rate of the hydrolysis step becomes restrictive at temperatures below 15°C–17°C. High-rate AnWT-systems for treating such types of complex types of wastewaters under low ambient temperature conditions are viable only by incorporating a conventional digester, operated under optimal mesophilic temperatures, and in parallel with the high-rate reactor. The function of this complementary digester is to achieve i) a sufficient stabilization of the insoluble organic matter entrapped in the high rate reactor and ii) to enable maintenance of a satisfactory methanogenic activity in that reactor, which is accomplished by returning part of the stabilized sludge from the digester into it. This integrated UASB-AnDi-reactor concept offers promise for sewage pre-treatment under winter time conditions in moderate climate regions.

##### High operational pressures

So far, very little relevant information is available concerning the practical feasibility of AnDeg-systems at pressures exceeding 100 bars. However, regarding the existence of anaerobic organisms in the deep oceans, the development and implementation of ‘pressurized’ AnDi systems might offer promise, e.g., for energy generation purposes, particularly when they can be optimized by adjusting the operational temperature.

### Mixing conditions

Because anaerobic organisms have a strong preference to form 'balanced' micro-ecosystems, heavy mixing in anaerobic reactors should be avoided. A gentle mode of mixing comprises a basic condition underlying the UASB-system, also of other modern high rate AnWT-reactors. However, in various earlier versions of the 'Anaerobic Contact' process, applied around the nineteen fifties and sixties for treatment of high strength industrial effluents, this condition was insufficiently met. It is probable that this also was/is not the case in many conventional sludge digesters.

Moreover, apart from enhancing process performance, the minimization of mechanical agitation will reduce the energy requirements, and thus also the investment costs.

**Occurrence of chemical precipitation** (e.g.,  $\text{CaCO}_3$ ,  $\text{MgNH}_4\text{PO}_4$ , and metal sulphides)

Chemical precipitates in and/or around immobilized films, sludge aggregates affect the performance of an AnWT quite detrimentally when during the operation of the system i) the active biomass will become scaled-in a tight  $\text{CaCO}_3$ -layer, ii) the sludge aggregates become far too heavy and/or iii) when it leads to 'cementing' of the sludge bed [47–51]. On the other hand, in case it is possible to control the precipitation process adequately, e.g., by maintaining the  $\text{PO}_4^{3-}$ -concentration at about 5 mg/L or by using small granules, a heavy (high ash-content) granular sludge will retain a high activity. This was demonstrated during the start-up of an 800 m<sup>3</sup> full scale UASB-reactor treating corn starch wastewater (COD: 1.5–11 g/L,  $\text{Ca}^{2+}$  up to 800 mg/L). It was the second full scale reactor implemented in the Netherlands, designed and commissioned according to criteria developed at our department [52]. A typical needle shaped, 60% ash-content, fine granular sludge evolved.

The benefit of a high ash-content granular sludge is that it enables the application of very high superficial velocities, e.g. as applied in EGSB-reactors [53]. It is interesting to note that this specific feature is not unique for high rate AnWT-processes, it also offers great potential for high rate upflow granular sludge  $\text{Ae}_{\text{micro}}$ -reactors for post-treatment purposes [54], very likely for high-rate granular sludge bed nitrification systems, even for granular sludge bed  $\text{AeWT}$ -systems for post-treatment, but possible even for direct treatment of sewage.

### Extreme salt concentrations

Regarding the high salt content of many industrial effluents, especially in situations where water loops have been closed to a large extent, an increasing need evolves for AnWT-systems that can cope with high salt concentrations. Particularly, a need arises for AnWT-systems that can cope with high sulphate concentrations, i.e., systems capable of reducing sulphate under conditions of high salt concentrations. According to findings by Vallero et al. [55–59] the potentials of such systems are substantial.

### Trace elements and macro-nutrients

Macro-nutrients (N and P) and numerous trace elements (Co, Mn, Ni, etc) are essential growth factors [60–64]. They therefore represent a unique tool for minimizing or maximizing bacterial growth, that is to say the fractional conversion of biodegradable substrate into either new biomass or methane. However, before this kind of 'steering' really can become practically fully applicable, still a lot of supplementary research needs to be done. Aspects to be elucidated with respect to the impact of trace element concern assessment of the effect of concentration, composition, mode and rate of supply, and their bio-availability, both chemical and physical. With respect to their bio-availability, the presence/absence of various types of chelating agents is of great importance.

Despite the extreme complexity of the issue of trace elements, nevertheless relevant information for practice has already been obtained nevertheless from laboratory investigations with respect to questions like 'Which are the really essential elements?' and 'In what amounts and how to supply them?' Based on the insights obtained, to some extent the excessive spreading of trace elements over the environment can be prevented, while concomitantly the operational cost of AnWT can be reduced. The studies conducted in our department so far particularly have been addressed to the assessment of the effect of trace elements on the anaerobic conversion of i) substrates/wastewaters originating from potato, and ii) of methanol, in the latter case both under mesophilic [65–67] and thermophilic conditions [68]. Comprehensive investigations of Florencio et al. [65–67,69] with methanol, lead to the insight that at low methanol and bicarbonate concentrations, and exceptionally low cobalt-concentration ( $< 0.0001$  mg/L) methanogens predominate over acidogens in the methanol conversion. Methanol is converted into methane directly, not via the intermediate formation of Volatile Fatty Acid (VFA). However, acidogenesis on methanol can predominate over methanogenesis at a relatively high concentration of methanol (i.e., overloading, poor treatment efficiency), when bicarbonate is supplied to the system, when Co is available and methanogens are inhibited due to the presence of undissociated VFA (for example) or an excessively low pH in the system. Through these findings, by now a stable performance of AnWT on methanolic solutions is possible.

The extraordinary complex character of the trace element issue has been more recently elaborated in comprehensive multidisciplinary investigations by Gonzalez-Gil et al. [19]. They investigated the effect of the mode of supply of trace element cocktails and found that a continuous supply is significantly more efficient than a slug-delivered supply. Comprehensive studies of Zandvoort et al. [70–74], conducted in close cooperation with the departments of Physical & Colloid Chemistry (e.g., Jansen, Van Leeuwen, et al., [75]), revealed the detrimental

effect of reduced bio-availability of the trace elements, e.g., due to their precipitation as sulphides. As remedial action the supply of chelating agents such as nitrilotriacetic acid (NTA), ethylenediaminetetraacetic acid (EDTA) and citrate were found promising; these compounds can restrict metal-sulphide precipitation, so that sufficient trace elements remain bio-available. However, it is again a very complex matter, because the chelating compound should not be easily biodegradable nor be a too strong chelating compound. Moreover, big differences in behaviour were observed between crushed and uncrushed granular sludge samples.

A category of chelating agents of eminent importance undoubtedly comprise ‘humic acids’. They represent a class of natural—chemically very complex—aromatic compounds, non-biodegradable and with strong chelating characteristics. They are of extraordinary importance for life. The first time we were faced with their presence/absence in AnWT- and AnDi-experiments was with potato derived substrates/wastewaters. In digestion experiments with solid matter of potato (but without peelings) and with real potato starch wastewater conducted in UASB-pilot plants, we observed an almost 100% conversion of the substrate-COD into methane-COD. Apparently, any bacterial growth in these experiments did not occur. Bacterial growth only could be realized by supplying a trace element cocktail to the system! These observations, made in the late seventies, ultimately made the board of the Potato Starch Company decide to implement the UASB-system for the treatment of the effluents of their factories, which were for decades heavily polluting the environment. The implementation was a great success. However, looking back, it has been a delicate matter, in the sense that the success indeed depended highly on the (rough) insight obtained on the effect of presence/supply of trace elements during the previous years of research. The point in this connection is that in the preliminary laboratory experiments conducted in the early seventies with freshly prepared potato juice (i.e., in fact comprising the effluent of the potato starch industry) in AF-systems, we did not observe any need to supply a trace element cocktail, nor was any retarded bacterial growth observed.

This surprising discrepancy in behaviour between freshly prepared potato juice solutions and real potato starch wastewater may in our view be attributed to the presence of humic acids in the real wastewater and absence in freshly prepared potato juice solutions. According to Field et al. [76] humic type compounds are easily formed from the highly reduced phenolic compounds via condensation reactions under the influence of free oxygen. These phenolic compounds are present in most plant juices, especially in potato juice. In potato starch production, these compounds become exposed to air once released to the wastewater. Since they are strongly chelating agents, and are present in relatively high concentrations in many wastewaters, e.g., potato starch

wastewater, they undoubtedly seriously can reduce the bio-availability of essential trace elements.

The above evidence illustrates sufficiently that clarification of the mysteries of the ‘trace element matter’ goes far beyond human capabilities. On the other hand, as pointed out, it is a matter of crucial importance at least to attempt to make relevant information available, even though it is very superficial.

A similar story applies for macro-nutrients such as phosphate or ammonia. The absence (or serious lack) of these macro-nutrients [77,78] will result in a severely retarded growth, although without affecting the treatment efficiency detrimentally for prolonged periods of time. However, for this category of essential nutrients few important aspects have been conclusively researched, leaving gaps in our knowledge. Compared to the issue of trace elements, it is certainly less complex.

Summarizing the above, it is evident that improving the insight into the effect of trace elements and macro-nutrients represents a matter of enormous practical impact for i) the quality of the environment, ii) for achieving an optimal performance of many biotechnological processes, and it is a matter of great scientific importance, e.g., for improving insight into natural life in general. Regarding the extraordinary complex character of this matter it implies that a lot of long-term, very well coordinated multidisciplinary research activities need to be initiated.

#### **Use of electron and redox mediators**

Compounds like humic acids, activated-carbon, quinones, e.g., anthraquinone disulfonate (AQDS), anthraquinone-2-sulphonate (AQS) catalyze the anaerobic degradation reactions of numerous recalcitrant compounds; they facilitate electron transport in the degradation of, e.g., azo dyes [79–87], and reductive de-halogenation [88]. However, as with the issue of ‘trace elements’ in this particular field, a better understanding is required which will lead to a wider and more optimal application of AnWT.

#### **Biological sulphate reduction (SubioRed)**

Numerous efforts have been made in the past and continue to be made in the sanitation world to suppress the occurrence of sulfate reduction (SuRed), viz. rather than attempting to take all possible benefits from it, which would be much more worthwhile. Since the late eighties numerous useful applications have already been discovered and implemented [89–92], under both mesophilic [93–97] and thermophilic conditions [58,98–101]. In several of these studies, issues such as immobilization, substrate competition between SuRed-organisms and methanogens, and exposure to high salt concentrations have been addressed.

Fascinating applications can be envisaged by combining the reductive and oxidative parts of the  $S_{\text{biol}}$  cycle, which to some extent already have been developed and implemented. Tempting examples are the removal of sulphate from wastewater, of  $SO_2$  from exhaust gases, of



the removal and recovery of heavy metals from contaminated soils or from acid mine drain water. But still substantial optimizations can be attained, e.g. with respect to the type of electron donors to be used, applicable process conditions and the immobilization of the involved organisms.

Immobilization of anaerobic microbial consortia in granules and films comprises a phenomenon of extraordinary importance and certainly not only for high rate AnWTs. Numerous biological processes can benefit from it. Since the first observation of the sludge aggregation phenomenon in AF-experiments [102] and in UASB-experiments in the early seventies and understanding its great importance for the UASB-process [6], many researchers have attempted to elucidate the mechanism of the phenomenon, e.g., Hulshoff Pol et al. [89,103,104], McHugh et al. [5,8], Fang et al. [105], resulting in a number of 'theories'. However, to date insufficient attention has been afforded to the effect of kinetic factors such as death, decay and growth rate of organisms participating in the immobilization process. Also, a number of other important questions so far have been relatively poorly addressed, such as that of i) the mechanism of the granular sludge augmentation (increase of the volume) in AnWT-reactors, ii) how to incorporate specific organisms in an existing granular sludge in order to enable the degradation of various kinds of complex compounds, and iii) the cultivation of specific types of granular sludge by using pure cultures. Numerous challenging questions remain to be resolved; it will need well coordinated interdisciplinary research.

## 5 Conclusions

1) In order to attain the required sustainable protection of the environment and consequently a more sustainable society, all already available means needed for that should be disseminated without any restriction to all world citizens.

2) With the various types of available NBM-based treatment systems and complementary clean water saving methods for waste & wastewater collection and transport, the major means to attain  $EP_{\text{sus}}$  lie ready 'on the shelf'.

3) In order to accomplish the optimal application of NBM-based treatment systems there is still a lot of challenging work to be done. The efforts to be made to generate that knowledge should not remain restricted to experimental laboratory work, but particularly also should be directed to thorough evaluations of the significant amount of relevant information already available in the literature.

4) With respect to the required continued experimental research on various complex issues a much better coordinated multidisciplinary tackle should be realized over prolonged periods of time.

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Upflow Anaerobic Sludge Bed Reactor system (UASB). He and his colleagues expanded the application of the system to a great variety of industrial effluents, including quite complex and even toxic wastewaters. The technology has been widely used by both industry and municipalities, since Prof. Lettinga has chosen not to patent this invention and has also shared his knowledge with young water engineers and professionals all over the world. The 2007 Tyler Prize for Environmental Achievement is awarded to him in recognition of his research and development of the environmentally sound novel process for the treatment of polluted wastewater and its implementation worldwide, especially in developing countries.