TREATING INDUSTRIAL WASTEWATER: Anaerobic Digestion Comes of Age

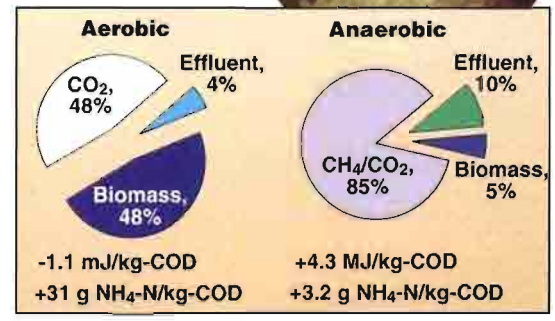

FIGURE 1. This comparison shows the respective fate of organic materials that are biodegraded under aerobic versus anaerobic conditions. Aerobic treatment requires energy input for aeration, whereas a net energy surplus is generated during anaerobic treatment, in the form of methane-bearing biogas that can be used to power utility boilers onsite

Anaerobic treatment systems offer important advantages over conventionally applied aerobic processes for removing organic pollutants from water-based streams

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In the absence of molecular oxygen, natural environments depend on the activity of anaerobic microorganisms for the biological degradation of organic substrates. In anaerobic environments where, other than carbon dioxide, no inorganic electron acceptors are present, the final degradation of organic compounds is achieved by their conversion to gaseous methane and carbon dioxide. As discussed here, wastewater-treatment systems that rely on the anaerobic digestion process for the removal of organic pollutants in industrial wastewater streams offer important advantages over conventionally applied aerobic processes.

Discussed in detail below, and summarized in Figure 1, are several of the most compelling operational advantages and sources of cost savings associated with using anaerobic biological-treatment systems.

Despite these demonstrable performance and cost-saving advantages — and the fact that nearly 1,600 commercial installations are currently in operation around the world — anaerobic wastewater treatment still remains something of a mystery to many chemical process operators. Many operators still harbor misconceptions and prejudices about the poor biodegradability and presumed toxicity of chemical and petrochemical wastewater constituents on anaerobic microbes. As a result, it is the opinion of these authors that such systems are not currently being applied as widely as they could be within the global chemical process industries (CPI).

This article aims to address some of the lingering misconceptions associated with anaerobic wastewater treatment. It provides a description of the microbiological basis of anaerobic biodegradation, and defines the boundary conditions required to enable anaerobic treatment.

A brief discussion of the pros and cons of today's leading anaerobic bioreactor designs is also provided, along with some simple calculations and basic guidelines for selecting the best bioreactor design for the waste stream at hand. Finally, the article provides a list of organic compounds that have proven to be degradable using anaerobic digestion, and includes several tables that summarize the current status of commercial-scale, anaerobic-treatment systems throughout the global CPI.

Primary advantages

Listed here are some of the leading operational advantages and sources of cost savings associated with using anaerobic biological-treatment systems:

1. Anaerobic treatment is in principle an energy-generating process through the production of byproduct methane-rich biogas. The use of the methane for energy generation elsewhere at the plant allows for conservation of more than 90% of the caloric value of the organic substrates being treated. In addition, modern anaerobic bioreactors do not require large energy input for mechanical mixing (which is required to maintain adequate aeration during aerobic treatment).

By comparison, during aerobic treatment, most of the caloric value of the organic substrates is dissipated as non-recoverable heat. And, aerobic bioreactors require significant amounts of energy for aeration.

2. Typically, anaerobic digestion produces only one-fifth to one-tenth as much biomass per unit of organic substrate converted as comparable aerobic processes. Since the disposal or treatment of waste sludge may account for 50% or more of the total waste-treatment costs, the ability to reduce sludge production is a major advantage of anaerobic treatment.

3. All microorganisms require nutrients, such as nitrogen, phosphorus and sulfur, for growth. However, many chemical and petrochemical wastewater streams contain relatively low levels of these nutrients.

If insufficient nutrient concentrations are present, additional nutrients must be dosed into the wastewater to

sustain the process. Since the amount of nutrients to be dosed is directly related to the amount of biomass formed, the nutrient loads required for anaerobic treatment systems are significantly lower than for aerobic systems.

4. Despite the relatively low biomass production that typifies most anaerobic digesters, higher biomass concentrations can be achieved in today's anaerobic bioreactors. In general, higher biomass concentrations enable higher volumetric treatment capacities, thereby minimizing the reactor volume and footprint required.

Heading for a breakdown

Thanks to the voracious appetites of so many types of aerobic microorganisms, and the robustness of the traditional aerobic activated-sludge process, aerobic wastewater treatment has long reigned as a leading method for managing organics-laden industrial wastewater streams. The activated-sludge process takes place in open basins, equipped with surface or submerged aeration facilities for transferring molecular oxygen from air to the liquid phase.

During respiration, aerobic microorganisms use molecular oxygen to convert complex organic compounds into carbon dioxide, water, and large quantities of residual biomass. Whereas roughly 50% of the organic substrate is respired, the remaining 50% is converted to biomass. The low levels of residual organic compounds that are typical in aerobically treated effluent often allow for direct discharge of the treated stream to surface waters.

By comparison, anaerobic microorganisms convert organic compounds into biogas consisting of methane and carbon dioxide. Only 5–10% of the organic compounds are converted to biomass. Despite the lower volume of biomass produced per unit of substrate converted, higher biomass concentrations (20–35 kg of dried solids per cubic meter) can be achieved in so-called "high-rate" anaerobic bioreactors.

Since the volumetric treatment capacity is directly related to the biomass concentration achieved, such high-rate bioreactors result in higher volumetric treatment capacities [on the order of 5 to 40 kg of chemical oxygen demand (COD) per cubic-meter reactor per day; see the box on p. 59 for more on COD]. Higher treatment capacities also allow for the design of compact reactors, with small footprints.

TABLE 1. ANAEROBIC SCORECARD: NUMBER OF SYSTEMS, BY REACTOR TYPE

Number of commercial scale reactors built in the world by January 2003 for treating different kind of industrial wastewaters and sewage.*

Type of wastewater	Type of reactor (see Figure 4 for reactor types)						
	Low-rate	AC	Fixed-bed	Moving-bed	UASB	EGSB	Total number
Food & related Industries							
Brewery & malt	2	—	6	4	185	88	285
Distillery & ethanol	25	31	40	—	76	9	181
Other beverage	—	3	11	2	88	15	119
Sugar production	—	49	7	1	32	3	92
Potato processing	14	4	2	—	46	10	76
Dairy, ice-cream & cheese	12	10	10	2	27	6	67
Starch production	2	9	10	2	34	7	64
Yeast production	7	8	6	—	25	8	54
Candy & confectionery	4	—	3	—	15	2	24
Citric acid production	2	3	1	1	3	5	15
Coffee processing	—	—	7	—	4	1	12
Wine processing	—	—	6	1	3	1	11
Fish & seafood processing	1	4	—	—	2	1	8
Miscellaneous	10	22	40	5	112	25	214
Non-food industries							
Pulp-&-paper	1	16	5	3	75	37	137
(Petro)chemical	3	4	43	1	20	20	91
Leachates	—	—	6	—	18	—	24
Pharmaceutical	4	1	2	—	6	3	16
Pig, cow manure & poultry	5	3	6	—	1	—	15
Textile	—	—	1	—	4	2	7
Natural rubber	—	—	3	—	3	—	6
Sludge & sludge liquor	1	—	2	1	1	—	5
Tobacco manufacture	—	—	—	—	4	—	4
Tannery	—	—	—	—	3	—	3
Fluegas desulfurization	—	—	—	—	—	1	1
Electronic components	—	—	—	—	1	—	1
Sewage	—	—	2	1	64	—	67
Number of reactors per type	**93**	**167**	**219**	**24**	**852**	**244**	**1,599**

*NOTES: Low-rate reactors include CSTR, lagoons and BVF reactors from the firm ADI. 80% of the plants reported correspond to BVF, since ADI offers one of the few low-rate systems commercialized as a turnkey package, and for which statistics exist. AC = Anaerobic Contact; UASB = Upflow Anaerobic Sludge Bed; EGSB = Expanded Granular Sludge Bed. The fixed-bed systems reported correspond for 44% to upflow anaerobic filters (UAF); 26% to downflow filters (DAF); and 30% to hybrid reactors. UASB reactors include reverse Dorr-Oliver clarigesters. Both upflow fluidized-bed reactors and the AnAerobics Mobilized Film Technology are classified as moving-bed reactors. They represent, respectively, 79% and 21% of this group of reactors. The Biothane Biobed EGSB (43% of the units) and Paques IC (57% of the units) systems conform to the expanded sludge bed class (EGSB) of reactors. The data presented here have been compiled from the January 2003 reference list of ADI (Canada), Biotec (Colombia), Biotecs (Brazil), Biothane (USA/Netherlands), Entec (Austria), Global Water Engineering (ex-Enviroasia, Germany), Paques (Netherlands), Proserpol (France), Purac (Sweden), Sinko Pantec (Japan) and older reference lists of AnAerobics (2001, USA), Applied Technologies (1999, USA), Badger (1994, USA), Biotim (2000, Belgium), Grontmij (1998, The Netherlands) and Ondeo-Degrémont (1999, France), as well as data from the literature and the authors' files. Care has been taken not to double count units reported in different reference lists due to commercial agreement between companies (Shinko Pantec with Badger and Grontmij; Biotecs with Biotim and Global Water Engineering). The data reported here correspond to treatment plants. This means that various units on one site have been considered as one sole reactor, unless the technology used changed at the occasion of a treatment expansion. As for the low-rate reactors, the number of plants treating sewage is probably much more important than the given one. Actually, most of these plants are designed by a large variety of local engineering companies difficult to identify, which is particularly true for Latin America.

Anaerobic limitations

While the cost and performance advantages of anaerobic digestion, as noted above, can be compelling, this approach also has some limitations. For instance, the lower biomass production per unit of substrate removed is an advantage when considering the amount of surplus sludge to be disposed, but becomes a disadvantage when considering the time needed for startup of an anaerobic treatment system. If no specifically suitable biomass is available in sufficient quantities, startup of the system may require up to several months. This

TABLE 2. A SAMPLING OF ANAEROBIC SYSTEMS BROUGHT ONLINE 1989–EARLY 2003

Commercial-scale anaerobic wastewater treatment plants built in the chemical & petrochemical industries (for a detailed list of plants built from 1981 to 1999, see Ref. (10)).*

Reactor number	Year of construction	Company and location	Industrial production generating the wastewater	Type of reactor	Organic Load Ton COD/d	COD removal %	Constructor, references
1	1989	K. Chemical, Hyogo, Japan	Dyestuff raw material	UAF	4.8	80 (BOD_5)	Shinko Pantec
2	1992	Undisclosed, Taiwan	PET	UASB	6	——	Global Water Engineering
3	——	Changzho Worldbest Radici, China	PET	Hybrid	1.7	——	Biotim
4	——	BP Amoco, Cooper River, SC, USA	PTA	DAF	——	80-85 (TOC)	BP Amoco
5	1994	Baek Hwa Co. Korea	Alcohols	EGSB	2.5	75**	Biothane
6	1996	APR, Kamalapuram, India	pH liquor rayon pulp	UASB	40	65** 85 (BOD_5)	Paques
7	1996	Copenor Brazil	Formaldehyde, formic acid, sodium formate, polyols, Hexamethyleneamine	EGSB	2.2	60**	Biothane
8	1997	Eastman Chem. Co. Singapore	Oxo chemicals	EGSB	12.5	92	Biothane
9	1997	Hyosung Industries Korea	Plastics	EGSB	0.9	75**	Biothane
10	1997	Procter & Gamble Indonesia***	Soaps, detergents	Hybrid	3	70**	ADI
11	1999	M. Gas Chemical Okayama, Japan	PTA	UASB	6 (BOD_5)	75 (BOD_5)	Shinko Pantec
12	2000	BP-Amoco Chem., Kuantan, Malaysia	PTA	IC	——	——	Paques
13	2000	SKW Trotsberg Germany	Metallurgical	EGSB	6.7	80**	Biothane
14	2000	Borsudchem Co. Hungary	PU, PVC	EGSB	10.7	80**	Biothane
15	2000	Reliance Industries, Maharastra, India	PTA	EGSB	25.2	70**	Biothane
16	2000	Sam Nam Petrochemicals, Korea	PTA	Hybrid	6	75**	ADI
17	2000	PT Bakrie Kasei Corp., Indonesia	PTA	Hybrid	18.8	75**	ADI
18	2001	Getec, São Gonçalo, Brazil	Sorbitol, manitol, food taste additives	IC	7.5	80	Paques
19	2001	Yizheng Chem. Fiber China	PTA	UAF	81.6	80**	Global Water Engineering
20	2001	Interquisa, Canada	PTA	EGSB	26.3	75**	Biothane
21	2002	Toray Industries Tokai, Japan	PTA	IC	16.8	75	Paques
22	2002	SUT IWWT Singapore	Various (13 chem. plants)	EGSB	20.8	60**	Biothane
23	2002	Procter & Gamble Thailand	Soaps, detergents	Hybrid	2.7	75**	ADI
24	2003	Celanese, Cangrejera Mexico	Alcohols, derivatives of acetic acid, amines, ketones and acrylates	Hybrid	9.6	> 90**	Celanese

* NOTES: The abbreviations are the same as in Table 1; PET = polyethylene terephthalate; PTA = purified terephthalic acid; PU = polyurethane; PVC = polyvinyl chloride; TOC = total organic carbon. Plant #4 must be operated similarly to the other DAF designed by BP Amoco at three of its PTA production facilities. The TOC removal is estimated from the one previously given for these units [10]. This plant is planned to be replaced by a UASB system. Plants number 16 & 17 correspond to expansions of the treatment capacity of already existing anaerobic plants. The information reported in this table comes from the same source as in Table 1, and from Yan et al. [13]. ** Design values. *** Factory closed down.

effect may become even more pronounced because most specific anaerobic microorganisms are capable of degrading a limited range of substrates. This means that seeding an anaerobic bioreactor for the treatment of a specific type of wastewater with biomass obtained from a reactor treating another type of wastewater may still take up to one month. Analogously, the time required to recover from a process upset is generally longer for anaerobic systems than for aerobic ones.

Another point to be considered is that the treated effluent from an anaerobic bioreactor typically has (slightly) higher effluent concentrations of organic materials, compared to that treated aerobically (0.1 to 0.5, versus 0.05 to 0.2, kg of COD per cubic meter of treated wastewater). In general, this makes anaerobic digestion less suitable for the treatment of very dilute wastestreams (those with less than 0.5 kg of COD per cubic meter of wastewater).

Higher concentrations of organic compounds in anaerobically treated effluent also implies that some form of post-treatment — such as aerobic biological treatment, or various types of physical-chemical treatment — will be required before discharge to surface waters. In general, anaerobic diges-

tion is better-suited for removing the bulk of organic materials from relatively concentrated wastestreams (those containing more than 4 kg of COD per cubic meter of wastewater).

Still, the evident advantages outweigh these potential shortcomings of anaerobic wastewater treatment, as reflected by the nearly 1,600 full-scale reactors that are now in commercial service worldwide (Table 1).

To date, anaerobic treatment has evolved to become the dominant treatment method for brewery, distillery, and numerous food-processing wastewaters, and has demonstrated great potential for treating more-complex waste streams. Examples include sewage, and wastewater produced by the chemical, petrochemical and pulp-and-paper industries.

Still, since less than 15% of all anaerobic wastewater-treatment systems are currently in use by such CPI facilities, a large potential expansion seems possible in the future. A summary of anaerobic bioreactors constructed between 1989 and early 2003 in the CPI is shown in Table 2. This table is an update of a longer compilation [*10*].

The implementation of anaerobic wastewater treatment in chemical and petrochemical applications has, for decades, been hampered by the presumed limited biodegradability of numerous substrates, and the presumed susceptibility of primarily methanogenic *Archaea* toward potential toxicants in petrochemical wastewaters. For both limitations, commercial experience has demonstrated that this is true only to a very limited extent; this is discussed further below.

Target compounds

The range of compounds that have been found to be degradable in methanogenic environments has increased enormously during the past three decades (Table 3). In general, most aliphatic and homocyclic aromatic compounds can be degraded in methanogenic environments, as long as they have at least one oxygen-containing functional group. Even some aromatic hydrocarbons, such as toluene and *o*-xylene, are completely fermented to methane and carbon dioxide in an anaerobic biodegradation system [*3*].

CHEMICAL OXYGEN DEMAND (COD)

The COD concept is a lumped concept that is used to represent the concentration and the oxidation state of organic materials that are present in a water-based stream [*1*]. The COD concentration corresponds to the concentration of oxygen that is required for the full oxidation of all organic carbon into carbon dioxide and water.

The units that are typically used to describe the COD concentration of a wastewater are kg of COD per cubic meter of wastewater, or kg of O_2 per cubic meter of wastewater. For solutions of a known composition, the COD-equivalent concentration can be calculated from the stoichiometry of the combustion reaction of the organic compounds. Combustion of ethanol for example can be described as:

$$C_2H_5OH + 3O_2 \rightarrow 2CO_2 + 3H_2O \qquad (1)$$

Consequently, to obtain COD-equivalent concentrations the conversion factors of 3 mol O_2 per mol ethanol, or 2.09 g O_2 per g ethanol, have to be applied. In general terms, the combustion reaction of an organic compound containing the atoms C, H, O, and N (with the oxidation state of ammonia being –III) can be written as:

$$C_xH_yO_zN_v^u + \frac{4x+y-2z-3v-u}{4}\cdot O_2 \rightarrow x\cdot CO_2 + \frac{y-3v-u}{2}\cdot H_2O + v\cdot NH_4^+ + (u-v)\cdot H^+ \qquad (2)$$

and the corresponding conversion factors can be calculated. □

It should be noted that lag periods prior to the degradation of some anthropogenic organic by anaerobic inocula from natural environments can be very long. The natural inocula contain a very limited number of microorganisms that can degrade these substrates, and they often need several months of growth before measurable conversion rates are achieved.

For example, for the anthropogenic phthalic acid isomers, it has been shown that the lag periods prior to quantifiable degradation were in the range of 20 to 100 days, depending on the origin of the inoculum and the orientation of the carboxylic groups on the benzene ring [*7*].

Meanwhile, compounds that were found to be persistent in anaerobic environments include several molecules containing tertiary-substituted carbon or ether bonds [i.e., methyl *tert*-butyl ether (MTBE), *tert*-amyl methyl ether (TAME), ethyl *tert*-butyl ether (ETBE)]. It should be noted, however, that several of these compounds are similarly hard to degrade in aerobic environments.

It should also be noted that some compounds that are persistent in aerobic environments are susceptible to partial conversion or even complete degradation in methanogenic environments. Examples include polyols (such as pentaerythritol, trimethylolpropane, azo-dyes, nitroaromatics and polychlorinated aromatic and aliphatic compounds, including tri-, tetra-, pentachlorophenol and tetrachloroethylene). In some cases, a sequential treatment scheme, which involves the use of both anaerobic and aerobic digestion, is required to achieve full degradation of these substrates.

Toxicity

Numerous organic and inorganic chemicals may have either a reversible or irreversible toxic effect on microorganisms. Irreversible toxic properties are generally related to the presence of highly reactive functional groups, such as aldehyde, or nitro-groups. Temporary exposure to elevated concentrations of some cations, organic acids or unfavorable pH values generally has a reversible toxic effect on microorganisms.

Anaerobic processes have a notorious reputation for being very susceptible to process upsets as a result of inhibition by toxicants. However — contrary to a still-common belief — anaerobic microorganisms, particularly methanogenic *Archaea*, are not more susceptible to toxic compounds than aerobic bacteria, except in the case of chlorinated aliphatic hydrocarbons [*2*]. Still, wastewaters containing elevated concentrations of potential toxicants will require specific pretreatments before either anaerobic or aerobic biological treatment. In some cases, it was found to be possible to treat toxic wastewater by acclimation of the biomass or by application of specific reactor designs.

Environmental factors

Petrochemical and chemical wastewaters typically have a temperature of 30 to 70°C. Anaerobic digestion has successfully been applied within two temperature ranges: 30–40°C and 50–65°C. In both zones, specialized microorganisms develop, enabling the application of the process in the full range of wastewater temperatures that are typical of CPI operations.

By comparison, at elevated temperatures, aerobic treatment systems can

suffer, due to decreased oxygen solubility (partly compensated by higher oxygen mass-transfer rates), higher heat production, and the large amount of water that will be evaporated.

Optimal conversion of organic substrates in anaerobic environments occurs at around neutral pH values. Since acidic carbon dioxide is one of the end products of the anaerobic digestion process, a basic compound needs either to be present in the wastewater or it needs to be dosed in, typically in the form of sodium hydroxide or carbonate.

In presence of sufficient neutralizing capacity (alkalinity), both gaseous carbon dioxide and bicarbonate will be formed, resulting in a strongly buffered system with a pH around 7. Depending on the oxidation state of the organic pollutants, and the strength of the wastewater, the biogas produced will typically contain about 30% carbon dioxide. In order to achieve a pH around 7 at a carbon dioxide concentration in the biogas of 30%, roughly 40 equivalents of bicarbonate per cubic meter of wastewater must be present in the reactor.

It should be noted that the actual pH inside the anaerobic reactor can be significantly different from the influent pH. Wastewaters containing high concentrations of biodegradable organic acids (>100 eq. per cubic meter of wastewater) and a low pH (around 4), for example, can be treated in a well-mixed reactor. Treatment is possible as long as at least about 40 eq. of the acids per cubic meter of wastewater are neutralized. Assuming that all organic acids are degraded in the reactor, this will result in the formation of 40 eq. bicarbonate per cubic meter of wastewater and a pH around 7.

TABLE 3. ORGANIC COMPOUNDS THAT ARE CANDIDATES FOR ANAEROBIC DIGESTION

Partial list of organic compounds known to be biodegradable under methanogenic conditions and susceptible to be present in the effluents of chemical and petrochemical industries.*

Aromatic compounds (homocyclic and heterocyclic)

- Benzene
  - methylbenzene (toluene)
  - 1,2-dimethylbenzene (o-xylene)
- Carboxybenzene (benzoate)
  - *o, m, p*-amino, chloro, iodo, methyl and methoxybenzoate
  - *o*-nitrobenzoate
  - mono, di, tri-hydroxybenzoate
  - 3,5-dichlorobenzoate
  - 3-chloro,4-hydroxybenzoate
  - 2-acetylbenzoate (acetylsalicylate)
  - 3,4,5-trimethoxybenzoate
  - 4-hydroxy-3- methoxybenzoate
  - 4-hydroxy-3,5-dimethoxybenzoate
- *o, m, p*-dicarboxybenzene (phthalates)
  - dimethyl *o*-phthalate and *p*-phthalate
  - diethyl *o*-phthalate
  - di-n-butyl *o*-phthalate
  - Butylbenzyl *o*-phthalate
- Formylbenzene (benzaldehyde)
  - 4-hydroxy-3,5-dimethoxy-benzaldehyde
- Formylbenzene (benzaldehyde) (cont.)
  - 4-hydroxy-3-methoxy-benzaldehyde
- Hydroxybenzene (phenol)
  - *o*-aminophenol
  - *o, m, p*-chloro, hydroxy, methoxy and nitrophenol
  - 2,4 -dichloro, 3,4-dichloro and 3,5-dichlorophenol
  - pentachlorophenol
  - trihydroxybenzene
  - *m, p*-methylphenol (*m, p*-cresol)
  - 2,6-dimethoxyphenol
- Nitrobenzene
  - 3-nitrobenzene sulfonate
- phenylacetate
- phenylpropenoate (cinnamate)
- phenylpropionate (hydrocinnamate)
- 3-methoxy-4-hydroxy cinnamate
- 4-hydroxyphenylalanine (tyrosine)
- Benzyl alcohol
  - 4-hydroxy benzyl alcohol
- 2-furaldehyde (furfural)

Aliphatic compounds

- Hydrogen cyanide
- ***Acids***
  - C1 to C18 *n*-carboxyalkanes
  - 4-aminobutyric
  - glyoxalic
  - 2-hydroxypropanoic (lactic)
  - 3-hydroxybutanoic and propanoic
  - *i*-butyric and *i*-valeric
  - acrylic, β-methylacrylic (crotonic) and 2-propenylacrylic (sorbic)
  - C2 to C6 *n*-dicarboxyalkanes
  - 4-aminoadipic
  - *cis* and *trans*-1,2-ethylenedicarboxylic (maleic and fumaric)
  - 2-hydroxy-1,2,3-propanetricarboxylic (citric)
- ***Aldehydes***
  - C1 to C4 *n*-formylalkanes
  - 2-butenal (crotonaldehyde)
  - 2-propenal (acrolein)
- ***Amines***
  - *sec*-butylamine
  - methyl, dimethyl, ethyldimethyl and trimethylamine
  - triethanolamine
- ***Alcohols***
  - C1 to C8 *n*-hydroxyalkanes
  - *i*-butanol
  - 1-amino-2-propanol
  - 3-methylbutanol
  - butylene, ethylene and propylene glycol (diols)
  - di, tri and polyethylene glycol (up to MW* 20,000)
  - glycerol (triol)
- ***Esters***
  - ethyl, methyl and vinyl acetate
  - butyl, ethyl and methyl acrylate
  - methyl *n*-, *i*-butyrate and propionate
- ***Ethers***
  - ethylene glycol monomethyl ether (2-methoxyethanol)
  - ethylene glycol monoethyl ether (ethoxyethanol)
  - methyl butyl ether
- ***Ketones***
  - acetone
  - methyl ethyl ketone

* MW: molecular weight.

Engineering aspects

Figure 4 provides a comparison of six widely used bioreactor designs, and provides optimal values for design boundary conditions that need to be observed during design. Brief descriptions are provided here.

Continuously stirred tank reactors (CSTR) were the first anaerobic bioreactors to be built, and are still widely used for anaerobic digestion of wastes with a high concentration of particulate matter, such as sewage sludge and the organic fraction of municipal solid waste.

The hydraulic retention time (HRT) in CSTR-type reactor is determined by the specific growth rate of the slowest-growing microorganism in the system. This generally means that very high HRT values are required to achieve an acceptable level of degradation (roughly 25 days). The high HRT values make the CSTR concept less feasible for treatment of the wastestreams considered here, containing primarily dissolved organic compounds at moderate concentrations (4–30 kg of COD per cubic meter of wastewater).

Uncoupling the biomass or solid retention time (SRT) from the HRT can increase the volumetric treatment capacity of an anaerobic bioreactor treating dissolved substrates. Initially, biomass separation from the biomass-wastewater mixture in the CSTR-type reactor was achieved by gravitational sedimentation in an external settler. During operation, settled biomass is fed back to the CSTR-type reactor, increasing the volumetric degradation rates in the system. This approach is called the ***Contact process***, and is fully comparable to the aerobic activated-sludge process.

However, the maximum biomass

MICROBIAL ASPECTS OF ANAEROBIC DEGRADATION

In environments where inorganic electron acceptors are absent, microorganisms depend on fermentation reactions to biodegrade organic substrates. In these methanogenic environments, the final products of the fermentation reactions are methane, carbon dioxide and ammonia.

From an energy perspective, the fermentation of organic substrates to methane and carbon dioxide yields much less energy for the microorganisms than the oxidation with inorganic electron acceptors (Table 4). This is the main reason why methanogenic degradation of organic substrates only occurs in the absence of external electron acceptors.

Furthermore, the free energy yields are directly coupled to the biomass yields of the microorganisms. Consequently, the amount of biomass formed per unit of organic substrate converted is much higher in aerobic or denitrifying environments compared to methanogenic environments. Since the rate of electron transfer per unit of biomass is basically independent of the electron acceptor [6], the different energy yields form the basis of the main differences between biological wastewater-treatment concepts for the removal of organic compounds.

Degradation of complex substrates in methanogenic environments involves a complex network of different types of microorganisms. Full conversion to methane and carbon dioxide results from a biological chain reaction, where one type of microorganism generates the substrate consumed by the subsequent organism in the chain. A schematic overview of the individual steps in the degradation of polymeric substrates in methanogenic environments is shown in Figure 2.

Hydrolysis of polymeric substrates (such as carbohydrates, proteins and fat) to the corresponding monomers is generally assumed to be catalyzed by extracellular enzymes that are excreted by acidogenic bacteria. These acidogenic bacteria ferment the monomers to a limited number of alcohols and volatile fatty acids. In the subsequent step of the chain reaction, acetogenic bacteria anaerobically oxidize alcohols and fatty acids to the two main precursors of methane production; namely, acetate and molecular hydrogen. Methanogenesis through acetate fermentation, and carbon dioxide reduction with molecular hydrogen, are both catalyzed by *Archaea*, which represent a distinct group of microorganisms. An example of a mixed methanogenic culture degrading a phthalate is shown in Figure 3. ❐

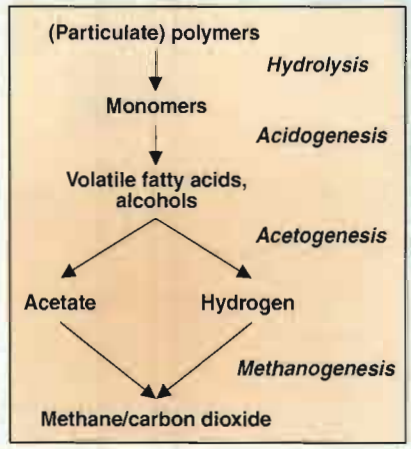


FIGURE 2 (left). Shown here is the stepwise degradation of organic materials in an anaerobic or methanogenic treatment environment

FIGURE 3. This microbial culture — a mixed, anaerobic culture enriched on an *ortho*-phthalate — consists of at least three different types of microorganisms: anaerobic, phthalate-oxidizing bacteria; and acetate-fermenting and CO_2-reducing methanogenic *Archaea*

TABLE 4. ENVIRONMENTALLY IMPORTANT ELECTRON ACCEPTORS AND THEIR STANDARD REDOX POTENTIALS (E^{01}) AND STANDARD FREE ENERGY CHANGES (ΔG^{01}) FOR REDUCTION*

Electron couple	Reaction	E^{01} (V)	ΔG^{01} (kJ/e)
Mn III/Mn II	manganese reduction	1.48	-142.6
O_2/H_2O	aerobic respiration	0.82	-78.7
Fe III/Fe II	iron reduction	0.77	-74.3
NO_3^-/N_2	denitrification	0.75	-72.1
H^+/H_2	proton reduction	-0.41	39.9
SO_4^{2-}/HS^-	sulfate reduction	-0.26	25.1
CO_2/CH_4	methanogenesis	-0.24	23.6

* Corrected for a biologically favorable pH of 7.0.

concentration that can be achieved in these systems is usually limited to 4–6 kg per cubic meter of reactor. This relatively low value for the biomass concentration is the result of the poor settling characteristics of methanogenic sludge, partly due to biogas formation in the settler. Consequently, this technology has only been applied on a very limited scale for treatment of industrial wastewater containing dissolved organic compounds. Nevertheless, the contact process remains a good option for wastewaters containing high concentrations of suspended solids and/or fats.

Upflow anaerobic sludge bed (UASB) reactors [9] combine a reaction compartment with an internal settler and a biogas separator. Wastewater is evenly distributed in the bottom (reaction) section of the reactor and flows through the sludge bed. In the sludge bed, the organic pollutants are converted into biogas. The biogas provides adequate mixing of the sludge-water mixture to avoid preferential channeling in the sludge bed, and excludes the need of mechanical mixing.

Biogas is collected in the three-phase separator that is operated at a low overpressure to increase the gas-to-liquid-exchange surface area. The sludge-water mixture flows to the settler section in the top of the reactor, where biomass is allowed to settle and return to the reaction compartment.

It was found in the 1970s that in this type of reactor, very high biomass concentrations (on the order of 20–30 kg of biomass per cubic meter of reactor) could be achieved with moderate-strength wastewaters (about 5 kg of COD per cubic meter of wastewater) at low HRT-values (around 10 h), but very high SRT-values (around 50 d). The effective uncoupling of the SRT and HRT in such systems resulted in higher treatment capacities.

The high biomass concentrations in UASB-type reactors can largely be attributed to the spontaneous formation of large, dense biomass conglomerates in the reactor compartment. This so called methanogenic granular sludge (shown in Figure 5) plays a key role in the high SRT values that can be achieved in this type of reactors.

Methanogenic granular sludge has a diameter of 0.5 to 3.0 mm and a biomass concentration of approximately 100 kg dry matter per cubic meter. Due to the absence of inert carrier material, the density of such granules is slightly higher than water (about 1.05 kg/m^3), enabling both good mixing in the reactor compartment and good settling in the internal settler. These characteristics make granular sludge a perfect biomass carrier. The combination of a simple construction and a high volumetric treatment capacity has made the UASB reactor concept the dominant anaerobic bioreactor type, with more than 800 currently in use worldwide.

Expanded granular sludge bed

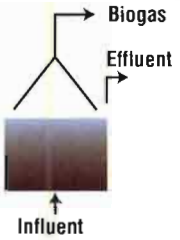
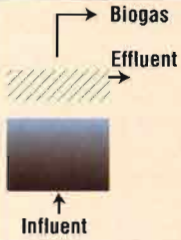
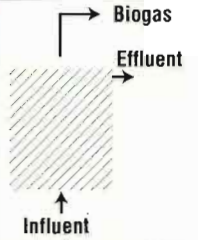
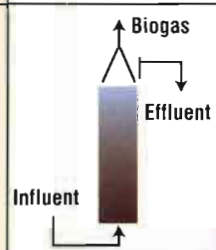
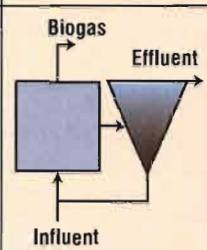
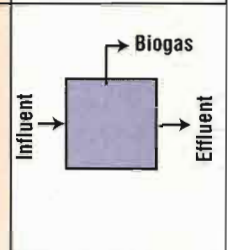
			
Reactor type	Upflow anaerobic sludge bed (UASB)	Hybrid	Anaerobic filter (AF)
Max. biomass conc., kg/m^3	25	25	15
Max upflow velocity, m/h	1	1	1–2
Reactor height, m	6	6	6
			
Reactor type	Expanded granular sludge bed (EGSB)/ fluidized-bed (FB)	Contact process	Continuously stirred tank reactor (CSTR)
Max. biomass conc., kg/m^3	35/20	5	3–12% of influent COD
Max. upflow velocity, m/h	15/30	—	—
Reactor height, m	15	—	—

FIGURE 4. Shown here is a comparison of the reactors most often used for commercial-scale anaerobic treatment of industrial wastewater, and the optimal boundary conditions for each

(EGSB) reactor. A novel variation on the UASB reactor concept is the EGSB reactor. This is comparable to UASB reactors, except that much higher liquid upflow velocities are applied: typically between 5 and 20 m/h, compared to less than 1 m/h in UASB reactors. To achieve this, EGSB reactors are constructed as tall reactors with a limited diameter and relatively small footprint.

The high liquid upflow velocities can avoid preferential channeling in the sludge bed, making wastewater distribution in the bottom of the reactor less critical. Consequently, treatment capacities and biomass concentrations are generally higher than those associated with UASB reactors.

Other bioreactor types that have been investigated intensively use biomass immobilized on either a stagnant support material (i.e., ***fixed-film reactors***) or a mobile support material (i.e., ***fluidized-bed reactors***). Both of these reactor types do not have the hydraulic limitation encountered in UASB-reactors. However, the desired biomass quantity and quality control is hard to achieve using these reactor types. Furthermore, the volume occupied by the carrier supports inside the reactor reduces the effective volume that is available for biomass formation; hence, volumetric treatment capacities are generally lower.

Hybrid reactor. A final variant comprises a combination of the UASB and the fixed-film reactor. In such a hybrid design, biomass separation is not achieved using an internal settler (as with UASB reactors), but with a packing material installed at the top of the reactor. Due to their relatively recent commercialization, these reactor types have — until now — found only limited application in anaerobic wastewater treatment.

Bioreactor design

If the substrate composition and strength of a wastestream are known, a basic design of a high-rate anaerobic bioreactor can be established, with relatively limited knowledge. The only parameters that must be known are:

- The overall biomass yield ($Y_{x/s}$) value for growth of the biomass (X) on the substrates (S) in the wastewater
- The maximum specific growth rate (μ^{max}) of the slowest-growing organism in the system, and
- The maximum active biomass concentration that can be achieved in a biomass-retention reactor systems (all reactor types shown in Figure 4, except the CSTR-type reactor).

The biomass yield ($Y_{x/s}$) values for full conversion of a wide array of organic substrates to methane and carbon dioxide are in the range of 0.03 to 0.12 kg/kg-COD removed from the wastewater. The actual values depend on several variables, such as the type of substrate, the temperature and the solid-retention time of the system. Numerous biomass yield values for anaerobic growth can be found in the literature [*11*].

If no biomass yield measurements are available, the total biomass yield of the mixed microbial population can be estimated, as long as the wastewater composition is known. Estimation of $Y_{x/s}$ is based on the concept of free energy conservation by bacteria. Free energy generated in a chemical reaction catalyzed by microorganisms is conserved and utilized for biomass production. An example calculation for prediction of the biomass yield is shown in the box titled Sample Problem, on p. 63.

The second step in the procedure is the definition of the slowest-growing organism in the system. For anaerobic bioreactors treating readily degradable soluble substrates, this is typically the process of acetoclastic methanogenesis, or acetate fermentation to methane and carbon dioxide. The maximum specific growth rate of the type of methanogenic organism that is normally dominant in methanogenic bioreactors, *Methanosaeta*, amounts to roughly 0.08/d at 35°C. [*14*]. To achieve effluent acetate concentrations in the grams per cubic meter range, this implies that the actual biomass growth rate in the reactor should not exceed about half of the maximum specific growth rate, or 0.04/d.

Then, combining the estimated biomass yield ($Y_{x/s}$, kg/kg-COD) and the biomass growth rate (μ, 1/d) with the typical biomass concentrations (X_R) in the different types of bioreactors (Figure 4) allows for the design of the

SAMPLE PROBLEM: ESTIMATION OF THE BIOMASS YIELD FOR ANAEROBIC BIOMASS DEGRADING ETHANOL

For estimation of the biomass yield, the stoichiometry of the energy-generating (catabolic) reaction equation needs to be derived:

$$C_2H_5OH \rightarrow 1.5CH_4 + 0.5CO_2 \quad (3)$$

$$\Delta G_{CAT} = -91.6\,kJ/reaction \quad (4)$$

From the stoichiometry of the chemical reaction, the standard free energy change under standard conditions can be calculated from the free energy of formation values of the reactants that can be found in review papers and textbooks [5, 12]. Standard free energy change values (ΔG^0) need to be corrected for the biologically favorable pH value of 7 ($\Delta G^{0'}$).

Free energy generated in the catabolic reaction is invested in the conversion of a carbon source to biomass (anabolic reaction). From an extended literature review, Heijnen [6] proposed that the amount of free energy required for biomass formation (ΔG_{AN}) depends only on the carbon chain length (C) and the COD-equivalence of the carbon source (COD, mol O_2 per mole of carbon source) for growth. The ΔG_{AN} value was found to be independent of the electron donor-acceptor couple. The following empirical relationship between these two variables and ΔG_{AN} was established:

$$\Delta G_{AN} = 200 + 18\cdot(6-C)^{1.8} + \exp\left\{\left[\left(3.8-\frac{COD\cdot 4}{C}\right)^2\right]^{0.16}\cdot(3.6+0.4\cdot C)\right\} \quad (5)$$

For ethanol, a ΔG_{AN} value of 706 kJ per mole of biomass formed can be calculated.

Now, the following equation for the biomass yield can be derived as a function of ΔG_{AN}, ΔG_{CAT} and the COD-equivalence values for the substrate (COD_S) and biomass (COD_X)

$$Y_{X/S} = \frac{-\Delta G_{CAT}}{\Delta G_{AN} - \frac{COD_x}{COD_s}\cdot\Delta G_{CAT}} \quad (6)$$

Using a general molar biomass composition of $CH_{1.8}O_{0.5}N_{0.2}$, a COD_X value of 1.05 mole O_2 per mole of biomass can be calculated. Using this equation, the estimated biomass yield for anaerobic growth of biomass on ethanol of 0.12 C-mol/mol is 0.032 g biomass per g ethanol-COD. ❐

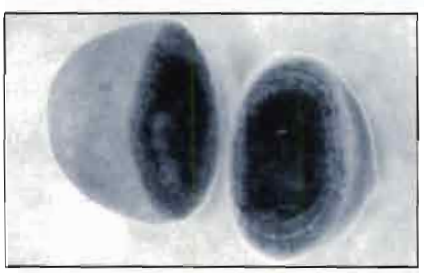


FIGURE 5. UASB-type anaerobic reactors often produce granular biomass, such as that shown here

anaerobic bioreactor. A mass balance for substrate over the reactor can be written as:

$$\frac{dS}{dt} = \frac{Q}{V}\cdot(S_{inf} - S) - \frac{\mu}{Y_{X/S}}\cdot X_R \quad (7)$$

where:

Q = Influent liquid flowrate, m^3/d

V = Reactor volume, m^3

In steady state, this equation can be solved to obtain the hydraulic retention time (HRT, in days) required to achieve an effluent concentration S:

$$HRT = \frac{V}{Q} = \frac{(S_{inf} - S)\cdot Y_{X/S}}{\mu\cdot X_R} \quad (8)$$

A boundary condition that needs to be checked for the different types of biofilm reactors is the maximum value for the hydraulic load. Based on the typical reactor height (h) of the reactor types shown in Figure 4, the liquid upflow velocity (v_{up}, m/h) can be calculated using the following equation:

$$v_{up} = h/HRT \quad (9)$$

If the calculated liquid upflow exceeds the value shown in Figure 4, biomass washout may impede sufficient solids retention in the reactor.

Figure 6 presents the HRT values required as a function of the COD concentration to be removed. The lower limits of the curves are the result of the hydraulic operational limit. The figure shows that the UASB and hybrid reactors concepts are not very attractive for the treatment of strongly dilute wastewaters, due to their limited hydraulic capacity.

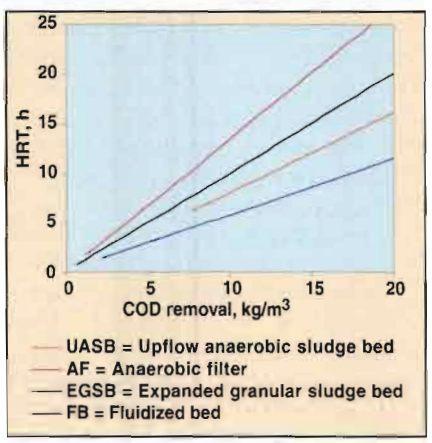


FIGURE 6. This comparison shows the hydraulic retention time required as a function of the COD concentration to be removed for different types of high-rate anaerobic bioreactors. Lower limits of the curves are set by the hydraulic operating limits. Assumptions used are μ = 0.04/d, and $Y_{x/s}$ = 0.06 kg/kg COD

Flow regime

The mixing regime in the reaction compartments of UASB, EGSB, hybrid and fluidized-bed reactor configurations approaches completely mixed at full scale. Consequently, spatial variations in both the substrate concentration and the biomass composition in the reactor compartments of these competing designs are small.

Still, elevated concentrations of organic substrates may locally occur in reactors that are operated at low hydraulic loading rates, such as UASB and hybrid reactors. In case of toxic but biodegradable compounds (such as formaldehyde) in the wastewater, small spatial-concentration variations may give rise to progressive toxification of the sludge bed [4]. This limitation can be overcome in reactors that are capable of handling high hydraulic loads (EGSB, fluidized-bed designs), by recirculation of the reactor effluent. For an example, see Ref. [15].

In other specific cases, a plug-flow regime is preferred over a completely mixed regime. A treatment system that enables a plug-flow regime can readily be achieved either by placing two or more high-rate anaerobic bioreactors in series, or by compartmentalizing the anaerobic bioreactor.

In general, a plug-flow regime is preferred in case of: (a) a wastewater that contains a mixture of slowly and rapidly degradable substrates, or (b) a wastewater that contains a substrate that gives rise to inhibition of the degradation of other substrate(s). One particular type of wastewater that exhibits both of these characteristics is the wastewater generated during purified terephthalic acid (PTA) production. PTA wastewater consists of an acetate, benzoate, terephthalate and para-toluate mixture. The first two are readily degradable in methanogenic environments, whereas the latter two are only degraded after long lag periods and at a considerably lower rates. It has been demonstrated that pre-removal of acetate and benzoate enables higher rates of degradation of terephthalate in the second stage [8]. Two specific characteristics of methanogenic terephthalate explain this observation:

• Terephthalate degradation is

EXAMPLE: REACTOR DESIGN

As an example, the bioreactor volume required to treat a wastewater with the composition and flow shown in Table 5 is calculated. It is assumed that a UASB-type reactor is designed, and that a COD-removal efficiency of 95% ($\eta_S = 0.95$) must be achieved. The organic material in the influent is assumed to be fully biodegradable in methanogenic environments. The required reactor volume can then be calculated according to:

$$V = \frac{Q \cdot \eta_S \cdot S_{inf} \cdot Y_{X/S}}{\mu \cdot X_R} = \frac{2000 \cdot 0.95 \cdot 5 \cdot 0.04}{0.04 \cdot 25} \quad (10)$$

giving a required reactor volume of 380 m^3.

Assuming a typical reactor height for UASB-type reactors of 6 m, the corresponding liquid upflow velocity amounts to 1.3 m/h. Thus, the maximum upflow velocity exceeds the proposed maximum value of 1 m/h shown in Figure 4. Relatively high upflow velocities may lead to limitations in biomass retention in UASB-type reactors; consequently, an EGSB, fluidized-bed or fixed-film reactor would be preferred for this example.

Another point that is useful to check is the solid-retention time (SRT) that needs to be achieved in the reactor, in order to enable a steady-state biomass concentration of 25 kg/m^3, as proposed for UASB-type reactors. The SRT can be calculated from the actual biomass growth rate, according to:

$$SRT = 1/\mu \quad (11)$$

giving an SRT value of 25 days. Given that the hydraulic retention time (HRT) amounts to only 4.5 hours, this means that the biomass needs to remain in the reactor roughly 130 times longer than the liquid. Provided that the biomass is present as methanogenic granular sludge (such as that shown in Figure 5, this can readily be achieved in UASB-type reactors, demonstrating the potential of the system. ❐

TABLE 5. EXAMPLE WASTEWATER COMPOSITION AND FLOW

Name	Abbr.	Unit	Value
Wastewater strength	S_{inf}	kg-COD/m^3	5
Influent flowrate	Q	m^3/d	2,000
Biomass yield	$Y_{X/S}$	g/g-COD	0.04
Biomass growth rate	μ	d^{-1}	0.04

strongly inhibited by benzoate and acetate, which both are intermediates of terephthalate degradation

- As described above, the microorganism with the lowest specific growth rate determines the actual SRT that needs to be achieved in the bioreactor. Since terephthalate-degrading biomass grows even more slowly than methanogens, they determine the actual SRT required

In summary

Anaerobic biological wastewater treatment is, in many cases, a highly attractive option for treatment of various chemical and petrochemical wastewaters. Compared to aerobic treatment, anaerobic treatment offers the following advantages:

- Lower biomass production per unit of organic substrate removed
- Lower nutrient requirements, which are specifically important for chemical and petrochemical wastewaters, which tend to be nutrient-deficient
- Production of a potential energy source (methane-bearing biogas), which can often be used onsite, as opposed to energy consumption for aeration in aerobic systems
- A temperature optimum that is typical of many CPI wastewater streams

The full potential of anaerobic treatment has only in the past two decades begun to be recognized. This has resulted in the current construction rate of roughly four new full-scale anaerobic treatment plants per year. Ongoing research and engineering efforts will surely expand the reach of anaerobic biological treatment in the years to come. ■

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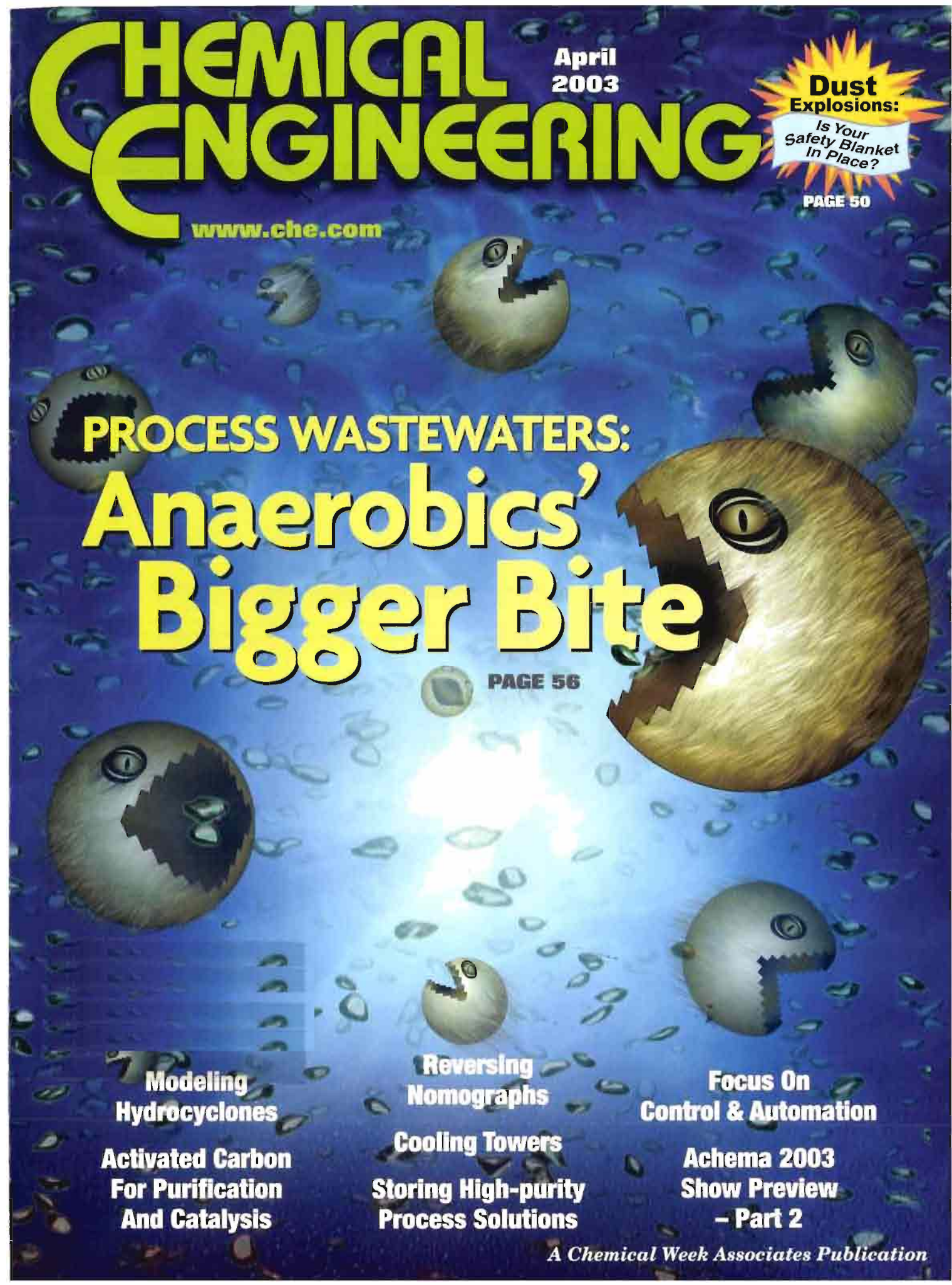

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