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„WASTEWATER TREATMENT IN PULP & PAPER INDUSTRY, NEW DEVELOPMENTS"

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SUMMARY

Concepts for wastewater treatment of pulp and paper mill effluents have steadily been developed in recent years according to the *best available technique* (BAT). Depending on the production and wastewater characteristic biological stages consist of an anaerobic or aerobic high loaded first stage followed by a low loaded activated sludge system. Aerobic wastewater biofilters are used as tertiary treatment and in single or two-stage biofiltration plants for the full biological treatment after primary treatment. By applying an *advanced oxidation process* (AOP) with a combined ozone and biofiltration process, the BAT quality standards are exceeded by far. New developments and large-scale applications are presented in this paper.

KEY WORDS

Moving bed biofilm reactor (MBBR), expanded granular sludge blanket reactor (EGSB), membrane bioreactor (MBR), one - two stage biofiltration, tertiary biofilters, advanced treatment, ozonation, advanced oxidation process (AOP), treated effluent reuse

CONTENT

1. INTRODUCTION

Wastewater load and wastewater characteristic for pulp and paper mills strongly depend on the type of process for chemical pulp mills or type of production and paper grades in paper mills.

New wastewater treatment concepts have continuously been developed during the last years considering process combinations according to the type of production and resulting effluent, long time experience, site specific conditions as well as regional legal requirements. Basic design standards must follow these preconditions representing state of the art rules. The European Union has documented the conditions and rules for the respective key industries in the *Best Available Technique* (BAT) *Reference Documents* (BREF), for the pulp and paper industry in the reference Document for the production of Pulp, Paper and Board [BREF (2015)].

Aerobic high loaded stages as the first biological stage are generally using the suspended carrier biofilm process (also called moving bed biofilm reactor MBBR). Anaerobic high loaded stages are using the expanded granular sludge blanket process (EGSB). In case of the anaerobic process as first biological stage the energetic use of the biogas is an essential part of the economic considerations. The low energy demand and the low production of biological excess sludge are further economic (and ecologic) advantages of the anaerobic process. A downstream low loaded activated sludge system is used for the final biological treatment as biological second stage.

The advantages of the MBR process – which as a single stage process has to be compared to aerobic two stage processes – are smaller bioreactor volume (due to higher biomass concentrations) and no secondary clarifier due to the membranes retaining the activated biomass. The MBR therefore needs much less area compared to a standard aerobic system. The treated effluent is virtually free of suspended solids. The MBR is predestined for internal water circuit treatment in papermills (kidney systems) and treated effluent reuse. However, only few MBR systems are in operation for pulp and paper effluents so far.

Aerobic wastewater biofilters are used for secondary treatment in single or two-stage biofiltration plants for the full biological treatment at low to medium BOD concentrations following primary treatment and as tertiary treatment (tertiary biofilters) after the secondary biological treatment (down flow the secondary clarifier in an activated sludge system).

Finally, an advanced treatment step has to be added when water quality of the receiving river requires far lower organic concentrations than can be achieved by a wastewater treatment plant designed according to BAT.

By applying an *advanced oxidation process* (AOP) with a combined ozonation and biofiltration process an effluent quality for discharge surpassing BAT is reached (Emerging technique).

Existing plants shall be checked according to the rules for new processes. A chemical mechanical treatment will be sufficient only very rarely for a limited time. Whenever they do not meet the state-of-the-art requirements, a renovation must be prepared. The instant of the execution then depends on different factors (requests, financing).

In order to adopt the state of the art for existing plants, the best possible variant of process combinations has to be chosen, regarding existing parts, predictable development of the effluent to be treated and of the authority’s requirements and above all the local situation.

New developments enabling improved operation with further developed systems and innovative process combinations are presented here.

2. DETERMINATION OF BASIC DESIGN PARAMETERS

For determination of basic design parameters and selection of the most effective and reliable treatment process, a link to the production is indispensable. Wastewater load and wastewater characteristic for pulp and paper mills strongly depend on the type of process for chemical pulp mills (see e.g. kraft pulp, sulphite pulp, integrated chemical pulp and papermills) or type of production and paper grades produced in paper mills (see e.g. integrated mechanical pulp and recycling fibre paper mills and non-integrated paper mills).

If we focus to the paper production within this short overview, it must be pointed out that the type of production essentially influences the organic load of the resulting sewage water, but also the specific wastewater consumption and thus the concentration of all dissolved substances. An overview of results found for a set of paper products according to a survey made in German papermills effluent after mechanical treatment in the early 1990’s (basically being still representative) is published [Möbius, Cordes-Tolle, (1993)], [Möbius (2016)].

Brief information about the recommended wastewater treatment processes according to our current level knowledge related the type of pulp and paper grades produced and resulting typical BOD concentration levels (and resulting BOD-load as basis for design of the bioreactors) is shown in **Table 1**.

Table 1: Recommended effluent treatment concepts related to main pulp and paper grades

Pulp and Paper production		Recommended biological process
Kaft pulp		Pure oxygen activated sludge cascade system MBBR + activated sludge
Sulphite pulp		Anaerobic for condensates Pure oxygen activated sludge cascade system MBBR + activated sludge system
Woodfree paper	BOD < 70 mg/l BOD > 70 mg/l BOD > 200 mg/l	Biofilters Two-stage biofilters Activated sludge cascade system
Coated paper	BOD 150 - 250 mg/l BOD > 200 mg/l	Activated sludge cascade system MBBR + activated sludge system
Printing paper (mechan. and secondary fibres)	BOD < 1000 mg/l	MBBR + activated sludge system
Packaging paper (secondary fibres)	BOD < 1000 mg/l COD > 1500 mg/l	MBBR + activated sludge system Anaerobic + activated sludge system

Designing a wastewater treatment plant for a pulp and papermill, as well for a greenfield mill as for an extension to an existing plant, primarily a decision on the treatment system is required. Preparing this, it is necessary to take into consideration a forecast of quantity and quality of the production wastewater to be treated (as consequence of future production capacity and type of production), and the required effluent quality for discharge.

The task often will need a variant study in which at least two systems with a complete dimensioning and cost estimate (capital and operation) are examined. If the wastewater to be treated cannot be examined in sufficient extent (because the production plant has to be set up first or the treatment has to be chosen for a planned change of production), published information and benchmark together with specific experience of experts will help to determine the required data.

The use of statistically evaluated data for dimensioning of waste- water treatment plants is necessary. The load used for dimensioning of bioreactors – BOD for aerobic, COD for anaerobic reactors – always refers to the 24 hr per day filtered samples for load calculation. In order to take into account, the statistical fluctuations of loads, and make different plants comparable, the 80th or 85th percentile of the data set is used as dimensioning value (in practice taken as mean value plus standard deviation mv+s of the data set used). In principle, the hydraulic dimensioning of the hydraulic related reactors such as primary and secondary clarifiers has to use the hydraulic peak flow. This is described as a short time value and met sufficiently well by use of the maximum hourly wastewater quantity. Unless daily hydrographs are not available, it is acceptable to calculate $Q_{h,max}$ by multiplying $Q_{d/24}$ with a proper factor, generally 1.6 to 2 before a hydraulic buffer and 1.3 - 1.6 following a hydraulic buffer. The factor itself is dependent of the statistical quality of Q_d, which well may be a mean value of yearly data. However, the design of pumps and pipes need to have the security factor according to the experiences of the operators of Pulp and Papermills.

An overview of effluent treatment process processes proven in the pulp and paper industry is given in **Figure 1**, whereby the key position of the biological processes is to be emphasized. This result from the fact that the most economical way to achieve low BOD treated effluent concentrations is by biological methods.

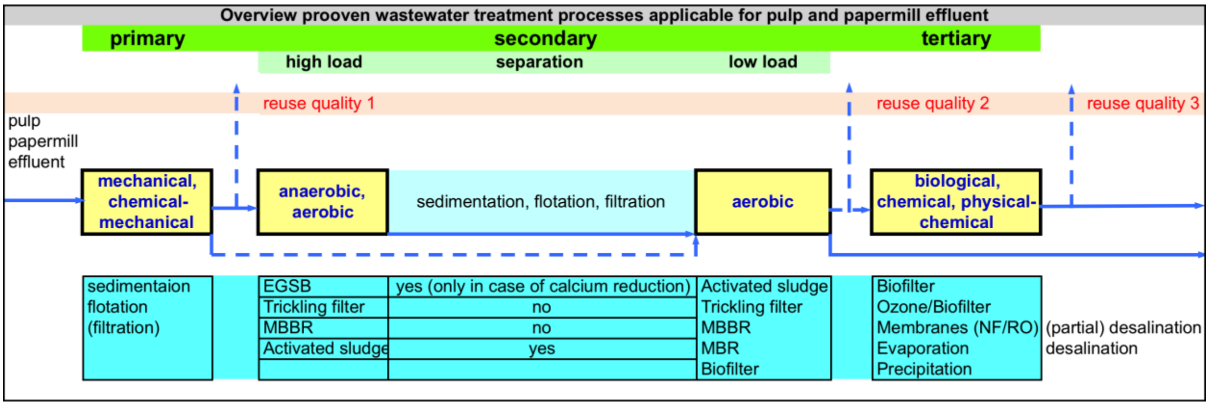

Figure 1: Proven wastewater treatment process combination

3. WASTEWATER TREATMENT SYSTEMS

3.1 MOVING BED BIOFILM REACTORS (MBBR)

Generally, a moving bed biofilm reactor (MBBR) is used as an aerobic high loaded first biological stage, followed by a low loaded activated sludge reactor. A BOD removal efficiency of 50 to 60 % is achieved with design load in the range of 4 to 7 kg of BOD per m³ of reactor volume and day. Two-stage designed MBBR’s with the same volume load achieve a higher BOD removal efficiency up to 70 to 75 % of BOD [Helble (2002)], [Helble, Jansen (2002)].

The type of carrier media and the biomass control of the carrier media could be optimised the last years. **Figure 2** shows the aerated surface with the airlift pump on the left used for control of the biofilm thickness. **Figure 3** and **4** shows different carrier media types applied for pulp and papermill effluent treatment today. Both have a big influence on operation, performance and efficiency of the process.

Some types of carriers developed in the 1990’s did not succeed since they lead to big problems with incrustations and thick biofilms. Newly developed carriers claim to be more stable and increase the efficiency of the process so much, that multiple BOD volume loads compared with the standard systems are possible without loss of elimination efficiency.

Figure 2: MBBR

Figure 3: Carrier (© Anox)

Figure 4: Carrier (© Mutag)

3.2 EXPANDED GRANULAR SLUDGE BLANKET REACTORS (EGSB)

After a period, when different anaerobic processes were tested (for example contact sludge, film, hybrid) and used in pulp and paper industry, the up flow anaerobic sludge blanket (UASB) process got standard (**Figure 5**). The hybrid reactor as a most promising combination of pellet reactor in the lower part and film reactor in the upper part, which has been in widespread use in North America, could not succeed in Europe.

The UASB reactor was mainly in use in packaging paper production (paper for corrugated board) starting in the 1990’s. Since more than 10 years now the dominating process is a further developed granular anaerobic process called expanded granular sludge blanket (EGSB) process. **Figure 6 - 10** shows examples of dominating EGSB reactor types.

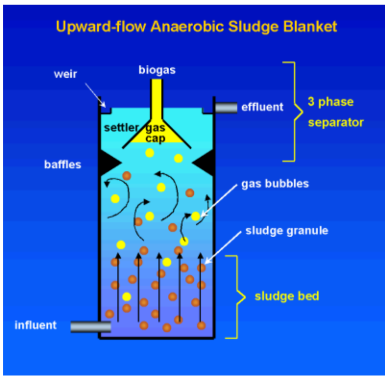

Figure 5: Principle of the UASB

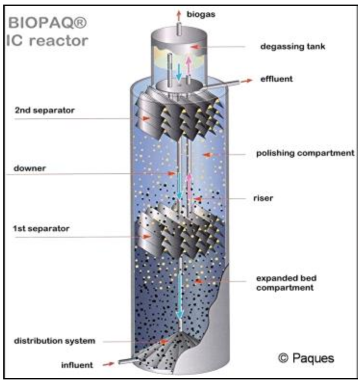

Figure 6: © BIOPAQ IC

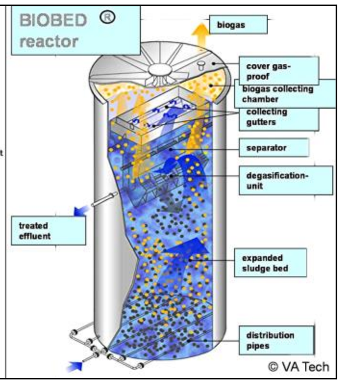

Figure 7: © BIOBED

Figure 8: © R2S EGSB

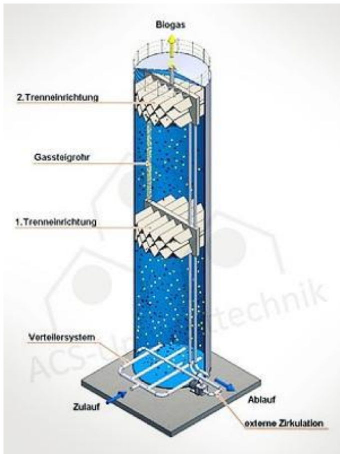

Figure 9: © ACS EGSB

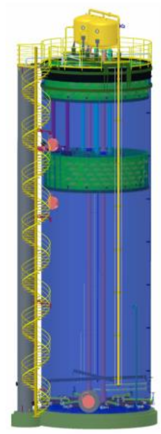

Figure 10: © Econvert IR

Today anaerobic high loaded stages are using the expanded granular sludge blanket process (EGSB). In the last two years several new developments in this compartment claim to reach similar or better elimination efficiency in BOD and COD elimination with much higher COD volume load than the above mentioned standard EGSB processes. Typical COD volume load for design range between 20 - 25 kg of COD per m^3 reactor and day.

In case of the anaerobic process as first biological stage the energetic use of the biogas is an essential part of the economic considerations, especially when a refund may be expected in case of production of so-called green energy, like in Germany from the EEG ("Erneuerbare Energien Gesetz" or renewable energy act). Another possibility can be cleaning the biogas to biomethan ("green gas"), supply into the natural gas net and decentralized usage in combined heat and power units. Additional costs for the biogas treatment and the green gas supply have to be evaluated in each case. A microturbine instead of a conventional combined heat and power unit (CHP) might be more cost efficient. The low energy required for operation and the low production of biological excess sludge are further economic (and ecologic) advantages of the anaerobic process.

However, there are several process conditions for a reliable application of anaerobic treatment. If the ratio of COD to SO_4 is too low, the process will not work properly due to the sulphate reduction and formation of sulphides. The key figure is the ratio of COD eliminated to chemically reduced sulphur, which optimal should be higher than 100 and not below 15

[Möbius, Demel, (2016)]. If the calcium concentration exceeds a certain level depending on the calcium carbonate content in the recycling paper itself and the level of a more closed-circuit additional measures to avoid massive calcium carbonate precipitation must be implemented. One possibility applied in papermills is the stripping of the formed CO_2 and additional precipitation by pH increase with caustic soda done in an additional reactor after the EGSB before entering the activated sludge system. A recirculation of a certain volume back to inflow of the EGSB reactor might even stabilise the process and avoids precipitation in the EGSB-reactor itself. The process only works economical when the organic substrate concentration is high enough, given as COD for instance more than 1.000 to 2.000 mg/l depending on the conditions. For lower COD concentrations (about < 2 g/l COD) the volume load has to be decreased in order to get a good elimination rate. So the invest costs per COD eliminated are increased, which makes the system too expensive compared to anaerobic high loaded reactors (MBBR) below a certain COD concentration, differing from case to case. When for instance the anaerobic high loaded reactor has been feasible for 1,5 g/l COD, with the new systems it might be feasible even below 1 g/l. Since the anaerobic treatment causes much less operation cost than the aerobic, this would decrease the total cost for many paper mills in the near future [Helble, Möbius (2008)].

3.3 MEMBRANE BIOREACTORS (MBR)

The membrane bioreactor process (MBR) is a well-developed and proven technology. The membrane materials and module systems have been developed continuously within the last years [Helble, Möbius (2007)], [Möbius, Helble (2009)]. Several installations have been built for municipal and industrial applications, but only few full scale MBR plants are in operation in pulp and paper industry [Wozniak, Renvoise (2007)], [Bauer, Helble (2014)].

The available technical MBR systems using either low pressure submerged membranes (**Figure 11, 12**) or classical horizontal cross flow tubular modules or as a new development aerated vertical cross flow tubular modules.

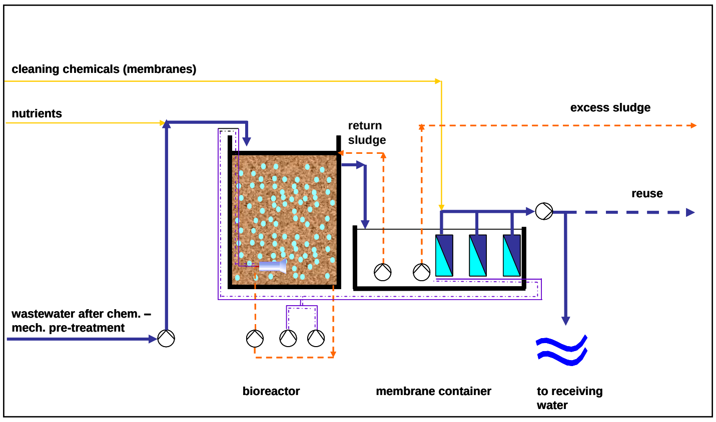

Figure 11: MBR schematic

Figure 12: Membrane container with flat sheet modules

The advantages of the MBR process – which as a single stage process has to be compared to aerobic two stage processes – are smaller bioreactor volume (due to higher biomass concentrations) and no secondary clarifier due to the membranes retaining the activated biomass. The MBR therefore needs much less area compared to a standard aerobic system. Depending on the pore size and membrane material high molecular dissolved organic compounds (> 0,05 µm) are hold back and might be further biodegraded, if possible, due to the higher sludge age. Our recommendation for MLSS design according to the available information is a moderate mixed liquor concentration of 10 – 12 g/l. The specific sludge loading in the activated sludge bioreactor related to biomass, also called F/M-ratio is recommended in the range of 0.08 – 0.12 kg/kgd. An optimal BOD removal efficiency (BOD effluent < 5 mg/l) is required to minimise the fouling potential on the membranes. The MBR is predestined for internal water circuit treatment in papermills (kidney systems) and treated wastewater reuse.

The costs of the system (invest and operation) are higher than for the standard aerobic two-stage system suspended carrier reactor plus low load activated sludge treatment. So it will be used only in cases, where the above mentioned advantages justify the higher costs. A new aspect might be the use of ceramic membranes, which is not new at all, but could be more compatible regarding the costs due to new development. This, however, still needs more research and experience.

3.4 BIOFILTRATION, ADVANCED TREATMENT

The Biological Aerated Filtration (BAF) represents a biological treatment with fixed film biomass. Biological treatment and filtration of suspended solids takes place in one and the same tank (**Figure 12**).

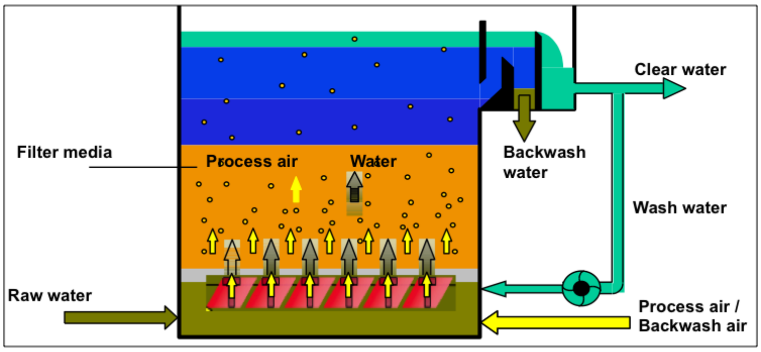

Figure 12: wastewater biofilter

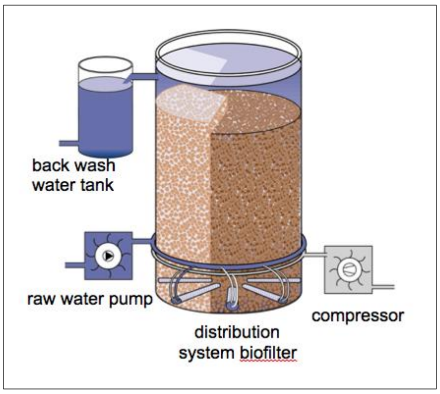

Figure 13: wastewater biofilter with modified wastewater distribution and aeration system

The practical experiences show that the biological up flow filtration - depending on the objective – is particularly suitable to eliminate carbon, ammonium, nitrogen and phosphorus. At the same time, an advanced removal of suspended solids is achieved. A process air diffuser below the strainer plate distributes the oxygen needed for the oxidation of carbon and ammonium. New developments provide a modified economic wastewater and air distribution system with modular biofilters (**Figure 13**) [Rüdiger (2005)]. The modular concept of biofilters simplifies possible later upgrading. An equal oxygen supply on the total filter cross-section must be assured. Denitrification (mainly for municipal wastewater treatment) can be achieved by recirculation or by injection of external carbon source such as raw sewage or methanol.

Biofilters are adapted for biological treatment of low and medium concentrated paper mill wastewaters, i. e. up to about 80-200 mg/l BOD as single stage plants after chemical-mechanical pre-treatment [Möbius (1999)]. For BOD concentrations below about 80-150 mg/l a one stage biofilter can be used. A two-stage biofilter (see fig. 3) is used for wastewater up to about 250 mg/l BOD.

Tertiary biofilters for advanced treatment are installed downstream of an activated sludge system. As advanced treatment process in pulp and paper industry generally carbon eliminating processes are used, mainly the tertiary wastewater biofilter (for COD polishing of 10 to 20 % and elimination of suspended solids).

Depending on the objectives the height of the filter material can be varied between 2 - 4 m. By choosing the appropriate filter material a high concentration of attached biomass and high suspended solids retention can be guaranteed simultaneously. Typical filtration velocity for design is 4 - 8 m/h as specific surface load. Depending on the BOD concentration volume loads of 6 - 8 kg per m^3 of media and day are achieved. In two stage biofilters a higher BOD volume load can be accepted in the first stage for design.

3.5 TERTIARY TREATMENT WITH ADVANCED OXYDATION PROCESS (AOP): OZONATION IN COMBINATION WITH BIOFILTRATION

The combined process uses the chemical / biochemical oxidation (chemical oxidation with ozone; biochemical oxidation with biofiltration) in an optimized ozone application representing an efficiency standard far above the European BAT level. The process is attributed to the group of advanced oxidation pro-cesses (AOP). The combination makes use of the effect of partial oxidation in which with reduced consumption of ozone (far reduced operation costs) of non-biodegradable (persistent) COD becomes biodegradable. For advanced treatment of the resulting biodegradable organic compounds biofiltration is applied (see chapter 3.4). High elimination rates of persistent COD and other compounds are achieved simultaneously, such as, AOX, colour, chelating agents (i.e. DTPA, EDTA), optical brighteners, surface active substances, micro pollutants, microorganism (disinfection).

The principle of this AOP is given in **Figure 14**. COD elimination generally is about 50 %, with a two stage system 85 % COD elimination may be reached. AOP with pressurized ozone reactors with shorter hydraulic retention time (HRT approx. 5 min at around 3 bar) and depressurized ozone reactors (HRT approx. 25 min, atmospheric pressure) are in large-scale applications in papermills [Kaindl (2005)].

The depressurised ozone reactor concept with ozone injection and diffusion system is schematically shown in **Figure 15**. The objective of this application is a further reduction of the specific ozone consumption required for the removal of persistent COD and overall improved energy efficiency [Haase, Helble (2015)].

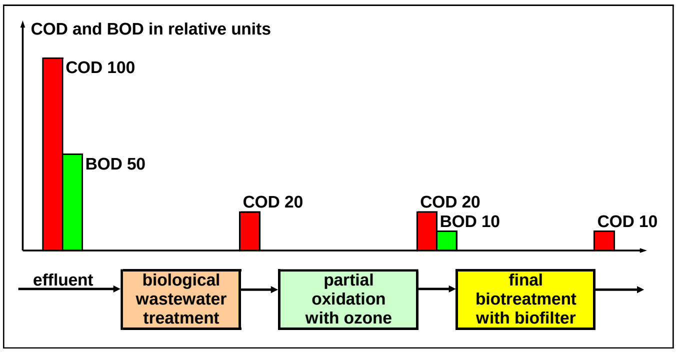

Figure 14: AOP for tertiary treatment

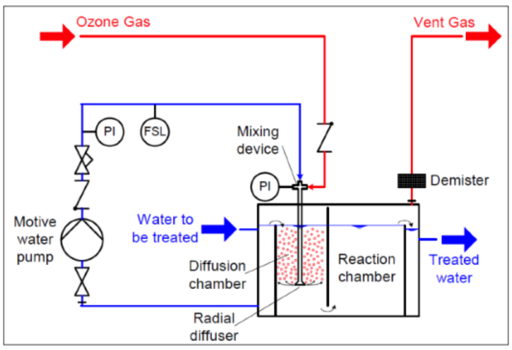

Figure 15: depressurized ozone reactor

The tertiary treated effluent by AOP is predestined for effluent reuse and to decrease the fresh water consumption. Tertiary treatment, however, is not a standard procedure in pulp and paper industry. It is used only when standard mechanical biological treatment is not sufficient to meet the quality requirement of the receiving water, or a high reuse rate is required

4. REUSE OF TREATED EFFLUENT

For reuse in paper production a treated effluent has to meet certain requirements, which are strongly depending on the type of production [Helble, Möbius (2001)]. Generally, the reused effluent should have a low concentration of suspended solids, BOD and – more or less, depending of the production – COD. Colour might be a quality parameter for white papers, but certainly only when a high percentage of the fresh water shall be substituted by treated effluent.

In any case the treated effluent must not contain any remarkable concentration of iron.

Microorganism contamination shall be low, but the effluent does not have to be sterile.

Regarding these requirements, quality of the different treatment systems can be evaluated as follows:

The standard system high load first stage (either anaerobic or aerobic) plus low loaded activated sludge treatment generally delivers a treated effluent containing a concentration of suspended solids SS below about 30 mg/l (in case of bad settling in the secondary clarifier much more), a rather low BOD concentration and a COD concentration depending on the biodegradability of the wastewater (between about 100 and 500 mg/l). The colour is often brown, except for paper production using only BCP as fibre stock, generally with a higher intensity than the untreated effluent. Microbiological contamination is not a problem for recycling rates up to about 50 %. The effluent can be improved for reuse by a sand filter (or even better tertiary biofilter) following the secondary clarifier. However, when settling is very bad (bulking sludge), the filter will be not operable, and the reuse has to be stopped.

The secondary biofilter is a good option for reuse of the water. In cases when this system is operable the organic concentrations are low anyway. The suspended solids concentration after the filter is low, but some abrasive broke of the carrier material (generally expanded clay granules) will be in the treated water. The colour will be more intense than before treatment, but effluents, which can be treated with biofilter, have a bright colour anyway. The microbiological contamination will be in the same range than with activated sludge plant.

In terms of suspended solids and microbiological contamination the MBR treated effluent has the best quality for reuse in the production. Suspended solids are normally below 3 mg/l.

Regarding colour, COD and some other parameters like complexion agents the runoff of an advanced oxidation process (ozonation plus biofilter) has the best quality for reuse. However, suspended solids and microorganisms are determined by the final step, which is the biofilter.

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