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Water and Papermaking
3. Measures to Clean Up Process Water

As was noted in Part 2 of this series, the build-up of dissolved and finely divided materials in the process water, or white water, of a paper mill can hurt product quality and also the efficiency of papermaking operations. Such problems are expected to become increasingly important, given current trends in:

- *reduced usage of fresh water*
- *larger proportions of recycled fibres*
- *continuing pressure to increase both product quality and rates of production.*

This article reviews various measures that have been developed to remove contaminants from white water. The best-known strategies include: membrane filtration, the use of coagulating chemicals; biological treatments under anaerobic or aerobic conditions; enzymatic treatments; use of oxidizing agents, and multiple-effect evaporation.

Whichever strategy or combination of strategies is adopted at a given mill, it is recommended also to take measures to reduce the amount of fresh water employed, thus reducing the volumetric flows that need to be purified.

Effective washing of the incoming pulp, as well as a good retention aid system, can reduce the burden on a kidney system used for white water purification.

The word "kidney" has been used when describing a wide variety of approaches by which paper technologists can remove dissolved and finely divided materials from the process water of paper machines.[1-30] Although implementation of such measures to clean up white water can add complexity and new expense to the operation of a paper mill, the arguments in favour of using kidney technology tend to grow stronger over time.

As discussed by others,[5,11,19,23,31-32] the costs associated with internal purification of white water often can be offset by decreased costs of purchasing and heating fresh water, as well as in treating and paying discharge fees associated with liquid effluent.

Early in the 1900s it was common for paper machine systems to use 500 to 1000 m^3 of fresh water per ton of product,[18] and then dump the excess water directly into a river. Subsequent technological progress has included:

- the development of save-alls to minimize loss of fine materials from a paper machine system,[33]
- development of efficient retention aid systems,[34] and
- implementation of wastewater treatment systems at pulp and paper mills.[35]

Increased costs and permitting issues related to the use of fresh water and discharge of liquid effluent have prompted technological developments aimed at increased internal recirculation of water within paper mills.[8,11-12,23,36]

The forming of a sheet of paper can be viewed as a separation operation. A fine screen excludes the fibres, as well as various fine materials and chemicals associated with those fibres, allowing most of the water to pass on through. However, as discussed in *Part 2*,[37] the retention efficiency can vary a great deal for different components of the furnish. Monomeric salt ions and sugar-like materials derived from the pulping and bleaching of wood tend to build up on a white water system, especially in cases where efforts have been undertaken to minimize liquid effluent from the system.[18,23,38-41]

Before considering different means to clean up white water, two common alternative approaches should be mentioned. The first of these involves adjusting the chemical additives to the system.[42] The idea is to compensate for effects of enriched salt ions, colloidal matter, and dissolved organic material in the white water so that acceptable levels of paper quality and paper machine runnability are maintained.

Additives used for such purposes include biocides, high-charge cationic additives, retention aids, cationic starch, sizing agents, etc., all of which may be at an increased dosage compared to what would have been needed if the white water had been cleaner.

Another likely alternative involves a decision not to reduce fresh water use beyond a certain point.[19,38] The idea is to find a level of contamination that can be tolerated, in terms of product quality and runnability.

A third possibility is to integrate mill operations with a municipal wastewater facility, using the treated municipal water, after further purification, as a source of process water.[38]

At each paper mill site, it is important to weigh the advantages and costs of such approaches, relative to exclusive reliance on any of the kidney-based technologies, as will be discussed below.

Ways to clean up white water

Table 1 (overleaf) summarizes some key strengths and suggested deficiencies of different reported strategies for white water quality improvement. Many of these strategies rely on implementation of one or more "kidneys," i.e. measures to remove dissolved solids, organic matter, or suspended minerals from the water. *Figure 1 (overleaf)* suggests how a kidney system can be incorporated into the processes of a paper mill that uses dry bailed fibre as a main component, as in the case of a containerboard recycling operation.

When comparing the different strategies listed in *Table 1*, one may consider "evaporation" as a kind of benchmark. Though evaporation can be considered as expensive,[4-5] it tends to be highly reliable, as well as capable of producing rather pure water. The only materials that tend to "carry over" to the condensed water are certain volatile low-mass alcohols and acids originating from the processing of wood.[4-5] Thus, adjustments of pH are sometimes needed in order to obtain the best results when evaporation is used to concentrate and remove salt ions from a paper machine system.[4]

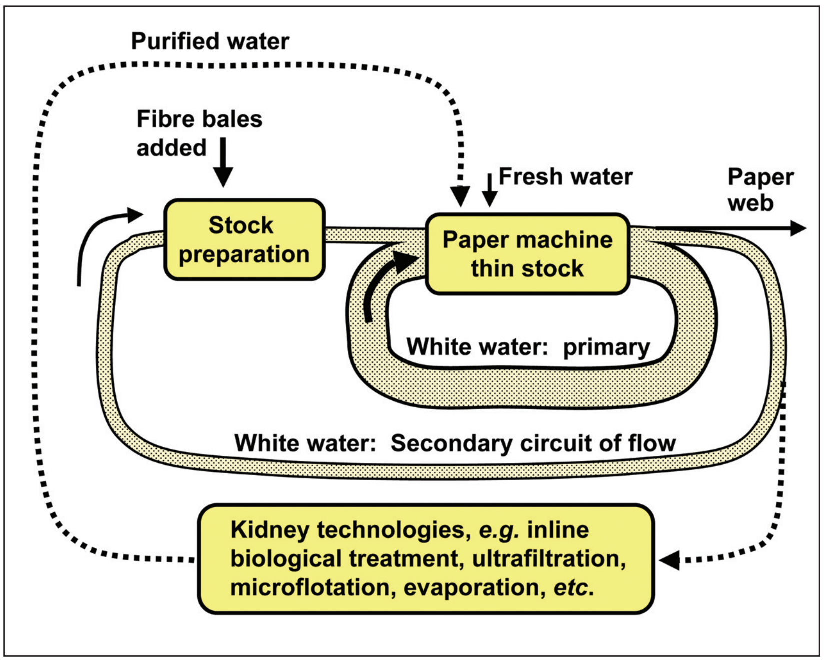

Figure 1 Generalized diagram showing how an inline biological treatment or other "kidney" water purification system can be incorporated into a paper mill process, replacing some of the fresh water intake

Membrane Filtration of White Water

Membrane systems have become widely recommended for white water purification, especially in light of advances in their durability and throughput.[1,23,26,30,43-44] In particular, ceramic membrane filters can withstand temperatures associated with the paper machine wet end, and even those of thermomechanical pulping,[26] conditions under which some synthetic polymer membranes fail.

Membrane filters separate various materials from liquid based on their size.[45] In theory, one could employ a one-step filtration with a so-called "nanofiltration" system capable of excluding even some large monomeric ions[13,26,46] or a reverse osmosis membrane that can exclude essentially everything except water.

However, there is often a tradeoff between exclusion capability vs. throughput and resistance to fouling. Ultrafiltration membranes are rated to exclude very large molecules, such as polyelectrolytes, whereas microfiltration systems have pore sizes selected to exclude bacterial cells and fine particulate matter suspended in the water. Throughput is often enhanced by a cross-flow design, as in the case of tubular filters, providing a means for the flow to minimize the caking of excluded materials on the membrane face.[26]

So, what pore-size category tends to be most advantageous for practical purification of white water? A case can be made that ultrafiltration, excluding materials larger than about 10 nm, is an ideal choice. Research has shown that it is the higher-mass polymeric material and colloidal byproducts of wood processing that tend to interfere the most with cationic retention aids and other wet-end additives,[43,47-49] and such materials can be substantially excluded by ultrafiltration.[12,23,43]

White Water Clean-Up Measure	Advantages Relative to Some Alternatives	Disadvantages Relative to Some Alternatives
Membrane filtration	Simple; moderate in cost; effective against bacteria	Prone to fouling and low volumetric throughput
Chemical coagulation	Useful as a pretreatment for membrane filtration	By itself not effective for most "non-substantive" [40] substances
Biological treatment	Very effective to reduce biological oxygen demand of the process water	When used alone, there is danger of poor dewatering, brightness loss, salt buildup
Enzyme treatment	A way to more precisely target the desired effects on white water	At most, solves only some of the problems related to white water
	components	
Oxidative treatment	Accelerates chemical breakdown of organic matter	By itself, does not remove anything; sometimes makes removal more difficult
Evaporation	Most reliable way to achieve high-quality condensate and brine	Tends to set a cost threshold, which competing approaches must beat

Table 1. Summary of strong and weak points of different general approaches to the cleaning up of white water in a paper machine system

Though it might be expected that microfiltration membranes would permit higher fluxes of filtration, compared to the finer pore sizes used in ultrafiltration, it appears that white water tends to equalize the performance of membrane filters across this range of pore sizes. Foulants such as bacteria and pitch can occlude the pores of a micro-filter system, due to a similarity in size.[26] In another study, microfiltration membranes were found to be suitable for exclusion of stickies from recycled paper pulp.[45]

Though membrane systems generally can handle wide ranges of levels of contamination in water, their volumetric throughput tends to be a limiting factor. Attempts to exceed a critical level of volumetric throughput sometimes accelerates plugging of membrane pores by suspended material near to the critical size.[26]

Due to these considerations, papermakers who contemplate the use of membrane technology for white water purification usually also take measures to minimize the net volumetric flows of fresh water and liquid effluent to and from the system.[26]

Technologies to recirculate white water multiple times, for such purposes as pulp dilution and showers, have been reviewed by others.[11,18,22-23,36]

As will be seen from some of the discussion that follows, there often can be an advantage of using membrane technology in combination with other water treatment measures. The expected drop-off in membrane throughput often can be delayed by pretreating aqueous input streams.[46] Conventional pretreatment strategies, before membrane filtration, include chemical coagulation, adsorption, sedimentation, and flotation.

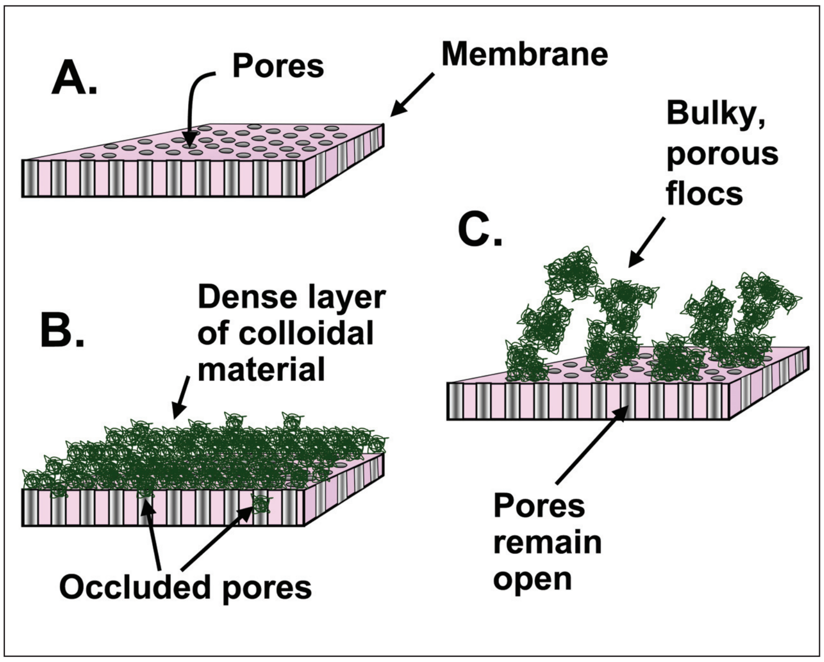

Figure 2 Schematic illustration of effect of coagulative pretreatment of white water before ultrafiltration. A: Representation of a filtration membrane; B: Same membrane fouled by very fine material from white water; C: White water treated with coagulant before ultrafiltration, yielding a bulkier, more porous sediment

Chemical Coagulation of White Water

Paper technologists are often familiar with coagulation phenomena, due to the fact that aluminum sulfate (“alum”), polyaluminum chloride (PAC), and low-mass, high-charge cationic polymers are commonly used to balance the colloidal charge of a paper machine, the purpose being to stabilize operations and enhance the performance of high-mass acrylamide-type retention aids and other additives. Such additives also can be used, at least as a first step, in the clarification of white water.[27]

By causing suspended materials to be agglomerated into larger-size units, coagulation can serve as a kind of pretreatment for other kidney operations, such as membrane filtration. *Figure 2* suggests a mechanism to explain why coagulation has the potential to increase throughput and reduce the rate of blockage of pores in an ultrafiltration membrane. Coagulation gathers the suspended material into larger agglomerates. Coagulated matter generally forms a more porous layer of sediment, and there is usually less chance that fine pores of the membrane become blocked.

Though acrylamide copolymers (PAMs) usually are considered only as a second additive for a two-component treatment of white water, a recent study compared different cationic PAM products as sole additives for the clarification of water resulting from thermomechanical pulping.[50]

Best performance was obtained at a medium-high level of cationic charge. Results were not highly dependent on the molecular mass of the additive. These results suggest an electrostatic mechanism of coagulation (charge neutralization or charged patches), rather than bridging between surfaces by the acrylamide copolymer molecules.

An innovative way to bring about the coagulation of suspended matter in an aqueous solution involves treatment with direct or alternating current. The effect appears to result at least partly due to the dissolution of metal ions from a sacrificial anode, such as aluminum.[1] Electrochemical activation has been incorporated into the design of a clarifier that has been used to treat effluent from a paper mill.[15]

The simplest approach, from the standpoint of paper machine operations, is just to add the coagulant to the stock system, and then follow up with addition of a high-mass acrylamide copolymer, i.e. a “retention aid,” just before the stock reaches the forming section of the paper machine. In this way, the fine matter becomes incorporated in the product and purged from the aqueous system.

This type of strategy often works well, provided that the colloidal material in the white water doesn't reach excessive concentrations or contain highly colored or smelly matter. In the latter case, a flotation treatment *(see next section)* might be used to remove the fine matter from the white water. Chemical coagulation often makes flotation systems more effective.[24]

Flotation of White Water

Whitewater treatment strategies based on flotation are already well known to many papermakers in the form of flotation saveall s.[51] Such devices use bubbles of air to lift a mat of solid materials, including fibre fines, to the surface of a shallow pool of water, where the solids can be skimmed off and returned to the paper machine system. Most flotation units in paper mills employ pressurized water, to which air has been added. Release of the pressure, as the water enters the bottom of a flotation cell, causes air to come out of solution as very fine bubbles.

For purposes of water clarification, it can be expected that dissolved-air flotation (DAF) units will be especially effective in the case of particulate matter having at least some hydrophobic character.[52] For this reason, it is not surprising that DAF units are commonly used to separate ink droplets from water. It has been found that very small bubbles ("microflotation") can be effective for removal of very finely divided matter from aqueous solution.[3,28,46]

A novel idea to enhance the performance of flotation systems, for purposes of water purification, involves the application of direct current to a pair of electrodes near to the entrance to a flotation cell.[1] Tiny bubbles of hydrogen and oxygen, generated by electrolysis at the two electrodes, provide the needed lift to colloidal and particulate matter in the water.

Biological Treatment of White Water

Most papermakers are familiar with biological treatment as an operation that occurs after papermaking, so that water discharged to the environment meets local requirements. However, as part of a system for within-mill treatment of white water, biological treatment can reduce the level of biological oxygen demand (BOD).

By using controlled growth of bacterial sludge, which is removed from the system, much less "food" remains, and slime problems can be reduced. Inline biological treatment has been implemented in some "closed" paper mills where the discharge of liquid effluent has been sharply reduced or eliminated.[10,17]

Biological treatment systems can be designed to run either in the absence of air (anaerobic systems) or with sufficient agitation or air injection to keep the system oxygenated during bacterial or fungal growth (aerobic systems). A further choice may involve the temperature of operation. Paper machines with fully closed white water systems often run at relatively high temperatures, too high for the majority of bacteria to thrive. However, thermophilic aerobic bacteria have been found to perform well in white water treatment systems at temperatures of 55-60ºC.[17,20,30]

Anaerobic biological treatment systems have been installed in several paperboard recycling mills in Germany.[18,22] According to the reports, the systems have worked well, making it possible to produce high quality packaging board with a closed white water system. Biocide use was discontinued in at least one case.[22] One of the expected advantages of anaerobic treatment systems is that the amount of sludge produced tends to be minimized. In some cases, the collection of methane and other gases, as byproducts, can be counted as an advantage.

Despite the demonstrated effectiveness of anaerobic biological treatment of white water, there are reasons that aerobic treatment may be preferred.[18] For one thing, it is desirable to end up with well-aerated water after treatment, thereby helping to avoid anaerobic conditions when the water is returned to the paper machine system. Anaerobic conditions in white water chests, for instance, have been associated with the production of foul smells and corrosion.[3]

Even in the absence of biological treatment, it has been recommended to aerate the white water used in mills where the amount of fresh water has been sharply minimized.[53] The combination of aerobic biological treatment, followed by microflotation has been highly recommended.[23] In another successful installation, aerobic biological treatment was followed by microfiltration.[12]

Recent publications have claimed superior performance in the case of moving bed biofilm reactors.[20,54] As illustrated in *Figure 3 (overleaf),* such systems employ plastic "carriers," onto which some of the bacterial material becomes attached. Because the carriers remain suspended in the reactor vessel, it is not necessary to recirculate "activated sludge" from a clarifier back to the reactor.

Biofilm systems have been demonstrated with water from a thermomechanical pulping operation under thermophilic bacterial conditions (55-60ºC).[20,30] High rates of reduction of biodegradable materials were observed in the course of a trial lasting several months.

Recent trials were carried out with a combined anaerobic / aerobic treatment sequence.[7,29] It was found that the key to a successful treatment was to accurately supply the amounts of phosphorous and nitrogen needed for bacterial growth, without leaving any significant excess of the nutrients in the water phase after the biodegradable content of the system has been consumed in the production of bacterial sludge. Such excess would tend to support slime growth in the paper machine system when the water is returned to the white water circuit.

In one of the cited studies[29] the majority of reduction in chemical oxygen demand occurred in the anaerobic stage of treatment. In addition to overcoming problems with the buildup of volatile fatty acids in the system, the dual-biological treatment also results in a substantial reduction in the level of sulphate in the system.

In a related study it was found that an anaerobic/aerobic biotreatment sequence was able to digest wood resins, reducing problems related to pitch.[13] The authors suggested that biotreatment be followed by nanofiltration to remove residual bacterial material, which would tend to decrease drainage rates and lower the brightness of paper products. Membrane bioreactors can combine both functions, providing support for activated sludge and exclusion of cellular material from the accepted filtrate.[15]

Because biological treatment systems are well-suited to remove low-mass organic molecules, but not designed to remove low-mass inorganic electrolytes, it has been suggested that inline biological treatment strategies be accompanied by a second type of kidney technology.

The output from a biological treatment operation is often turbid, and it may contain viable bacteria cells. It has been suggested that evaporation (see later) could be used to remove salts.[8]

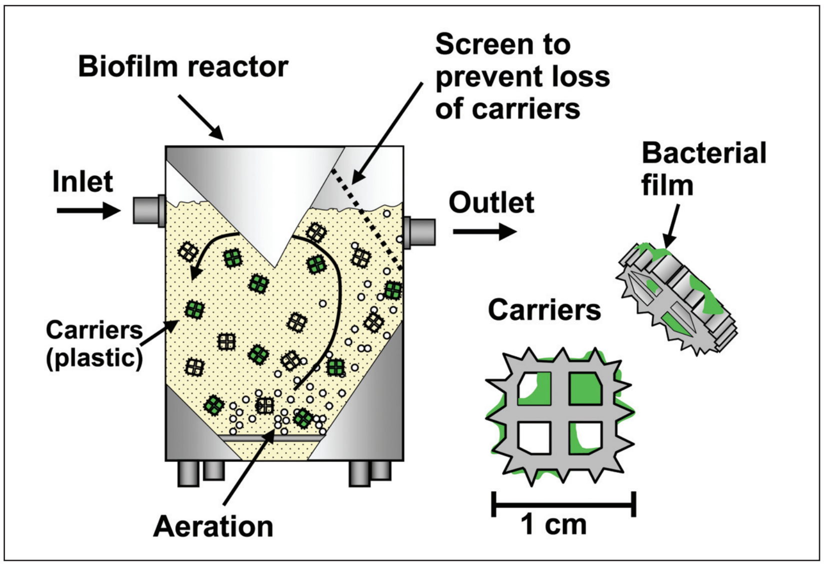

Figure 3 Main features of a moving bed biofilm reactor using aerobic conditions. Aeration also agitates the mixture, keeping the plastic carriers in suspension. The carriers hold biofilm, eliminating the need for activated sludge recirculation.

However, it may be more cost-effective to follow biological treatment with reverse osmosis, using membranes that are rated to exclude even monomeric ions. As a bonus, biotreatment is expected to increase throughput when using such a membrane.[26,55]

In some cases, it appears that substantial quantities of inorganic ions can be purged from the system as a component of biological sludge. For instance, the conditioning of biologically treated water by addition of CO_2 gas has been found to precipitate $CaCO_3$, which is retained in the sludge, reducing the level of hardness in the water returned to circulation.[25]

Enzymatic Treatment of White Water

Enzymatic agents are produced by living cells, including bacteria. In fact, much of what happens during biological treatment of water can be attributed to the action of various enzymes. By using enzymes, rather than living bacteria or fungi, there can be opportunities for greater control and selectivity.

For the treatment of white water the enzyme laccase appeared quite effective. Specific contaminants were removed from thermomechanical pulping water.[28] However, in the cited study the net biological oxygen demand of the system actually increased, due to the biomass of the enzyme itself. Fungal extracts have been found effective to break down wood extractives and carbohydrates.[9,14,56] Related enzyme treatments have been found to reduce pitch problems.[57]

Earlier, it was noted that hemicellulose-derived macromolecules of relatively high molecular mass tend to form strong complexes with various cationic additives, rendering them less effective. One solution, as suggested earlier, involves use of an ultrafiltration membrane capable of excluding that high-mass material.

A contrasting approach is possible with the use of enzymes. The idea is to cut up the hemicellulosic macromolecules into smaller bits that no longer have sufficient chain length to form complexes with cationic additives. It appears that this type of mechanism accounts for the effectiveness of pectinase in reducing the cationic demand of water from peroxide-bleached thermomechanical pulp.[48-49]

Another likely reason to add enzymes to white water is to increase rates of drainage. Substantial increases in dewatering rates are often observed when cellulases are added to the whole furnish.[58-60] In theory the effect is due to dissolution of very fine fibrillar cellulosic material. Due to their extremely high surface area per unit mass, cellulosic colloidal fines are expected to be the first to become substantially dissolved when cellulase is added to whole furnish.[61]

But, the addition of cellulase to whole furnish can raise concerns about partial degradation of the long fibres[60] In principle, such degradation of fibres would be avoided by separate treatment of white water. However, it might be uneconomical to hold large volumes of white water for a sufficiently long time to allow effective enzymatic treatment in the absence of fibres.

Oxidative Treatments of White Water

Oxidation is another way to break down various biodegradable materials in white water. A potential advantage over some other approaches, including some kinds of biological treatment, is that oxidation treatments can be optimized to break down recalcitrant organic compounds, including dyes, pesticides, and surfactants.[62] A combination of ozone treatment with photocatalysis (TiO_2 particles and ultraviolet radiation) has been reported to be highly effective.[62-63]

A potential drawback of oxidative treatment is disruption of bacterial cells, producing fragments that are in a size range likely to plug pores of ultrafiltration membranes.[26] Another potential barrier to implementation of white water treatment schemes based on oxidation is the need for such treatments to be followed by washing operations or cooling, depending on the treatment conditions.

Distillation of White Water

Evaporation systems can produce water that is free of salts and most organic matter,[44] with the possible exception of some low-boiling organic materials, such as low-mass alcohols[5] and acetic acid.[4]

Advances in technology have lowered the costs associated with evaporation,[5] and the application is most recommended for situations in which relatively small volumes of highly contaminated water need to be treated. Economies can be achieved by carrying out the evaporation in a series of stages (or "effects") and using reduced pressures, similar to what happens in a kraft chemical recovery operation.

Because evaporation is extremely effective at excluding salts from the distillate, the method has been recommended to be combined with biological treatment,[8] In one series of trials, three different designs of evaporation systems all yielded water that exceeded the quality of locally available fresh water.

Washing as a Possible Alternative

In addition to the "kidney" options discussed in the previous sections, papermakers also need to consider opportunities for improved washing of pulps before the materials reach the paper machine system. By eliminating contaminants from the furnish to begin with, some of the costs associated with white water purification may be avoided in some cases.[23]

The clearest advantages, based on such an approach, would be expected in paper mills employed virgin unbleached kraft pulp, a process in which the salts and lignin byproducts can become converted to pulping chemicals and/or energy during the recovery cycle.

There is a need for more study to determine the optimum degree of washing that can be justified after mechanical pulping or the recycling of waste fibres. Improved washing has the potential to increase the quality of the paper product and reduce the costs associated with wet-end chemicals.

Retention of White Water Components on Fibres

Enhanced use of retention aids is yet another approach that ought to be considered, at least in some cases, before investing in kidney technologies. Often the most effective retention aid treatments, from the standpoint of purifying white water, begin with addition of a high-charge cationic agent. For example, polyethyleneimine (PEI) has been shown to decrease the turbidity of white water when added to the furnish.[64]

Especially effective retention of colloidal fines is expected when a first additive is able to at least partly neutralize the surface charges, and a second additive, a very-high-mass flocculant, is able to bind fibre fines and other materials onto cellulosic fibres. As noted in *Part 2* of this series,[37] however, such strategies have potential to lower the strength and brightness of the paper, depending on what kinds of materials are present in the white water.

Recently it has been shown that zeolites, having a nanoporous structure, are able to adsorb large amounts of contaminants, including calcium[65-67] and manganese ions.[68] By reducing the level of Ca^{2+} there is a reduced likelihood of scale formation and pitch deposits. Adverse effects of manganese were mentioned in *Part 1* of this series.[69] Reportedly, zeolites can be used as a substitute for precipitated calcium carbonate (PCC) filler.[70] Improvements in retention and drainage were attributed to cleaner white water.

Summary

Based on the literature cited in this article, the following general statements can be made:

1. Several different viable technologies already have been demonstrated for the inline purification of paper machine process water. Some of the most attractive approaches include ultrafiltration (tubular membrane units, etc.), inline anaerobic, aerobic, or combined biological treatments, chemical coagulation of white water, and distillation with multiple-effect evaporators.

2. Synergistic effects often can be expected by combining two or more kidney-type strategies that have different strong points. For instance it is often advantageous to pretreat white water with high charge cationic fixatives prior to membrane filtration in order to slow down losses in throughput. Likewise, it can be advantageous to follow biological treatment by either ultrafiltration or microflotation, so that any cellular material is excluded from the purified water. At the same time, the biological treatment yields a very low biological oxygen demand (BOD) and a low potential for slime growth in the system.

3. Before considering any of the kidney technologies described in the literature, it is recommended first to decide whether similar improvements in the cleanliness of white water might be achieved just by washing pulp better before it enters the paper machine system. Alternatively, the effective use of a retention aid programme can effectively purge fines and colloidal material (but not monomeric ions) from the white water system; in some, but not all such cases there may be no measurable adverse effect on paper properties.

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