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Application of Ultrafiltration Membrane Technology in Pulp and Paper Mills in Indian Context: A Review

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Abstract: In general, Indian pulp and paper mills generate 25–225 m³ wastewater/tonne of paper. Pulp and paper mill effluent carries high pollution load in terms of COD, BOD, absorbable organic halides (AOX) and is dark brown in colour mainly due to the lignin and lignin derivatives. Stringent application of new environmental norms in the form of CREP (Corporate Responsibility for Environmental Protection) has forced many mills to close down their operation. Hence, large proportion of water now being discharged need to be recycled back into the system at the appropriate intake points with/without treatment. There are several conventional treatment processes employed in Indian paper industry which effectively remove BOD, TSS, and also COD but do not remove colour, AOX, heavy metal and process chemicals as per the desired level to recirculate the process water in good condition.

Membrane technology is being adopted increasingly in the paper mills for closing their mill production process. Application of membrane technology is one such option which can improve the recycled water quality by removal of heavy metal contaminates, COD, TSS, colour, AOX, and lignin. This shall reduce the load on conventional recovery evaporation system with no significant loss of inorganic compounds. Membranes also provide an effective method of closing the chelating stages and allow the filtrates to be used as bleach plant wash liquors. Membrane filtration also allows efficient recycling of white water. The present paper reviews current status of ultrafiltration in terms of R&D efforts and its application to the paper industry with due focus of system closure in Indian pulp and paper mills.

Key words: Ultrafiltration membrane, pulp and paper mill, system closure, paper mill effluent.

Introduction

The pulp and paper industry is one of the 17 most polluting industries listed by the CPCB (Central Pollution Control Board). Huge amount of water is being used in the manufacture of pulp and paper. Water utilized finally leaves as wastewater unless it is appropriately recycled. Pulp and paper mill effluent carries high pollution load in terms of COD, BOD, absorbable organic halides (AOX) and is dark brown in colour mainly due to the lignin and lignin derivatives (Kulkarni, 2003).

In view of the stringent environmental norms in the form of CREP (Corporate Responsibility for Environmental Protection) laid down by CPCB, large proportion of water now being discharged will have to be recycled back into the system at the appropriate intake points with/without treatment. Conventional biological treatment processes effectively remove BOD, TSS, and also COD but do not remove colour, AOX, heavy metal and process chemicals as per the desired level to recirculate the process water in good condition. For this purpose, recent R&D efforts are on for the use of membrane filtration as a wastewater treatment technology in pulp and paper industry.

The major effluent generating sections of paper mill are: pulping section, bleaching section (E-stage, C and/or D-stage), coating, paper machine, de-inking (in the case of waste paper based mill) and finally common effluent. As membrane filtration is based on pore size and different pore size membrane are used for each type

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of effluent, it is well suited for the system closure with segregated approach. Though all membrane filtration processes mentioned above are being used in paper industry but, ultrafiltration has been found wide in the applications. The present paper reviews status of ultrafiltration in terms of R&D efforts and its application to the paper industry from different sections as well as common effluent with focus on the system closure. The summary is given in Table 1.

Application of Ultrafiltration on Pulp Mill Effluent

The effluents from the pulp mill is black liquor containing complex mixtures of aromatic phenolic compounds ranging from low molecular weight monomeric to high molecular weight polymeric substances known as lignin and its derivatives. UF treatment can be a suitable system for concentrating black liquor or separation of high calorific lignin compound and inorganic chemicals. The separated streams are further used as by-products (Bhattacharjee, 2006). Chakravorty et al. (1987) found that U.F. treatment of wash plant effluent could produce very clear water by removing colour and turbidity to the tune of 90% besides chemical oxygen demand (COD) reduction to the order of 70%. Neytzell-de Wilde (1987) found 80% removal of lignin and 57% removal of TOC in 5000 molecular mass limit (MML) and 61% and 37% in MML 100,000 respectively from pulp mill effluent. Kulkarni et al. (1998) discussed the techno-economic viability of ultrafiltration process in non-woody spent liquors and calculated that ultrafiltration of non-woody spent liquors improve the nature of permeate. Its anaerobic degradation efficiency is also improved to 70.9% as against 40.5% of original liquor. Wallberg Ola et al. (2003) studied lignin removal from kraft black liquor by ultrafiltration and during the investigation, 80%, 67% and 45% lignin retention was observed. After diafiltration and concentration, the retention of Fe, Mg and Mn in the end of the experiment was 98%, 100% and 99%, respectively. Guangli Liu et al. (2004) analyzed treatability of kraft spent liquor by ultrafiltration. The result showed 90% retention in lignin, the removal of silica was 80% and total efficiency was also 80% of UF membrane. Bhattacharya et al. (2005) studied biological treatment followed by these membrane processes thus resulting in a clear liquid with negligible chemical oxygen demand (COD) and less than 0.9 mg/L of total reducing sugar (TRS). Bhattacharjee et al. (2006) found that influence of membrane disc rotation increases the flux substantially, more than so obtained by stirrer rotation.

Application of Ultrafiltration on Bleach Plant Effluent

Application of Ultrafiltration on Combined Bleach Plant Effluent

Rune (1980) observed that the total solids in the bleach effluent are normally around 0.7% and the liquor can be concentrated upto 25-50 times by UF. Even if only 50% of the total solids are kept in the concentrate, 90% of the colour is retained. Zaidi et al. (1992), Afonso and Pinho (1991), and Falth (2000) compared the efficiency of (1) ultrafiltration and (2) ultrafiltration plus dissolved air flotation for bleach plant effluent. The results showed 54%, 88%, and 100% removal of TOC, colour, and SS, respectively by ultrafiltration alone. Ultrafiltration plus dissolved air flotation resulted in 65%, 90% and 100% removal of TOC, colour, and SS, respectively. Merry (2001) achieved 50% of the target reduction in the COD, with a 98% reduction in volume, on the soft wood with a polyethersulphone membrane.

Meuller et al. (2003) concluded that about 50% of the bleach plant COD, BOD and colour will be returned to the recovery system, but less than 20% of Ba, Ca, Fe, Mg and Mn and about 35% of the oxalate will be recirculated after using ultrafiltration. Also only about 5% of the total chloride will be returned. With the eucalyptus effluents AOX and COD reductions of about 80% and 70% respectively were obtained. Ultrafiltration and partial closure of alkaline filtrates in ECF bleaching can reduce final bleach plant effluent COD by as much as 60% and AOX by as much as 40%. Anna-Karin Nordin et al. (2006) observed that the change in original design parameter can improve the performance of membrane and increase the flux. It was observed that increasing the inlet pressure from 0.7 MPa to 1.1 MPa in the last stage would increase the average flux around 2.5 times.

Removal of different contaminants in different stages by ultrafiltration separation is discussed below.

Application of Ultrafiltration on C-Stage Effluent

In the last 20 years most of the research work on this subject is based on the work of Pfister and Sjostrom (1978), that determined the distribution of molecular weight (MWD) of the bleached Kraft effluent using ultrafiltration technique in range (i) 30% MW < 1000, (ii) 50% 1000 < 10000, (iii) 10% 10000<25000 and (iv) 10% MW> 25000. Jokela and Salkinoja-Salonen (1992), using aqueous and non-aqueous size exclusion chromatography (SEC), showed that over 85% of the chlorinated material present in the C-stage effluent is of low molecular weight (<1000 Da). Most of the substances in

Table: 1 Current Status: Ultrafiltration membrane technology to close water circuit in pulp and paper mill

S. N.	*Process*	*Membrane Specification*	*R&D Result*	*Current status in world*	*Current status in India*	*Reference*
1.	Pulp Mill Effluent	Cut-off MWCO, 4000, 5000, 8000, Cellulose triacetate membrane	COD reduction 70%, Colour and turbidity 90%, lignin removal 80%, TOC removal 57, silica 80%, and total efficiency 80% of UF membrane	Pilot scale	Pilot scale	Chakravorty, B. et al. (1987), F.G. Neytzell-dewilde (1987), Guangli Liu et al. (2004), Chiranjib Bhattacharjee et al. (2006). Wallberg Ola et al. (2003)
2.	Common Bleach Plant Effluent	Polysulphone membrane	Removal efficiency; Colour 88% to 90 %, BOD 50 %, TOC 54%, SS 100%. AOX 80%, COD 70%	Applicable on commercial scale	Pilot scale	Rune glemenius (1980), Zaidi et al. (1992), Afonso and Pinho (1991), Falth (2000), Meuller et al. (2003)
3.	C-Stage Effluent	Polysulphone membrane, Cut-off 100-1000	Removal efficiency; Chloride content 60%, Sodium sulfate and Carbonate 90%	Lab scale	Lab scale	Pfister and Sjostrom (1978), Zadorecki Paul (1987), Jonsson and Petersson (1988, 1989), Salkinoja-Salonen (1992), Moudgil, B.M. et al. (1994)
4.	E-Stage Effluent	Polysulphone membrane, flow velocity 3-6 m/s (10-20 ft/s), pressure 1200 kPa (174 psi).	Chemical recovery nearly 100%, Colour removal 90%, Removal COD 50%, Removal BOD_7 10%, Water retain 96%, Conductivity 40%, TOC 65%, TSS 100%	Applicable on commercial scale	Lab scale	Lundahl H. et al. (1980) Jansoon, A.S. (1987, 1989), M. Nystrijm et al. (1988), Rosa et al. (1995), Surendra S. et al. (1999), Richard Greaves (1999), De Pinho M. (Fredrik Filth et al. 2001)
5.	Coating Plant Effluent	Polyethersulfone, Polyaramide, Polyvinylpyrrolidone, cut- off 50000 (MWCO) Working pressure of 20-28 psig	Nearly 100% water is recovered	Applicable on commercial scale	Lab scale	Stridsberg et al. (1992), Stina Nygard et al. (1998), Surendra S. et al. (1999)
6.	Paper Machine Effluent	Hydrophilic C30G membrane	Removal efficiency: 99% Suspended solid COD 10-20%, SS and Turbidity 100%	Pilot scale	Lab scale	Elefsiotis P. et al. (1995), Jutta Nuortila-Jokinen et al. (1996), Ramamurthy P. et al. (1996), Milan Teppler et al. (1999)
7.	De-Inking Effluent	Polysulfone hollow-fiber membranes, MWCO 500,000	Removal efficiency of chemicals close to 100%	Pilot scale	Lab scale	Desaulniers et al. (1970) Bradley H. et al. (1997), Bruno C. et al. (1998)
8.	Common Effluent	Polyethylenemine (PEI), Polyvinyl alcohol (PVA)	Removal efficiency Turbidity 90%, 54% of Fe and 70% of COD	Pilot scale	Lab scale	B. Chakravorty et al. (1987), Nuortila-Jokinen, J. et al. (1994) Koyuncu, I. et al. (1999) M. Vieira et al. (2001), S.C. Low et al. (2005)

the C-stage effluent are too small to be retained, even by a very dense ultrafiltration membrane. This makes C-stage effluent difficult to be treated with ultrafiltration. The large amount of effluent is another factor that makes the membrane treatment of C-stage even more difficult (Jonsson and Peterson, 1989). They conducted experiments by mixing C-stage and E-stage effluents in their actual proportion, however results were not very encouraging. Moudgil et al. (1994) however investigated a method for purging chlorides from the kraft liquor cycle using aqueous-aqueous separation technology. In laboratory tests it was shown that using PEG (poly ethylene glycol) polymer and salt cake solution, a 60% reduction in chloride content can be achieved, with 90% recovery of sodium sulfate and carbonate, and 98% recovery of the PEG.

Application of Ultrafiltration on E-stage Effluent

E-stage effluent is very well suited for ultrafiltration process because of its comparatively small volume and high molecular weight substances in it. Lundahl et al. (1980) found total removal of colour (90%), 40% reduction in COD, and 10% reduction in BOD_7. Nystrijm et al. (1988) observed that sufficient chlorolignin removal can be achieved if ultrafiltration is carried out at pH 10, which is approximately the pH of the first caustic stage in the bleach process.

Jansson (1987, 1989) observed that ultrafiltration of plant E- stage effluent contribute can reduce TOCl (Total organic chlorides) by 60-80%, COD by 50-80%, AOX (absorbable organic halides) by 90%, colour by 90%, BOD_7 by 25-50%. Rosa et al. (1995) studied colour removal experiment of E-stage effluent through ultrafiltration, and found 85% colour removal efficiency by the ultrafiltration with a thin film composite (TFC) membrane of 10.8 kDa, while total colour removal was achieved using a TFC nanofiltration membrane of poly (trans-2,5-dimethyl) piperazinthiofurazanamide. Singh et al. (1999) achieved nearly 100% recovery of the chemicals. Richard (1999) achieved 50% reduction in COD in oxygen bleaching by ultrafiltration in the pilot scale study. De Pinho et al. (2000) carried out flotation/ ultrafiltration experiments to treat E1 stage effluent and achieved the removal efficiencies in terms of conductivity, TOC, colour, and TSS, 40%, 65%, 90%, and 100% respectively. Filth et al. (2001) found that the retention of the substances causing the COD is approximately 40%.

Application of Ultrafiltration on Coating Plant Effluent

Paper coatings are composed of pigments, binders and flow modifiers. Ultrafiltration of coating plant effluent has three advantages. Firstly no coating effluent is released; secondly, the concentrated effluent can be recycled; and thirdly, permeate can be used as wash water. Stridsberg et al. (1992) successfully achieved concentrated coating plant effluent from 0.5% to 46% in a cross flow rotational filter system. Stina et al. (1998) concentrated regenerated coating colour using UF and mixed with the fresh base coating colour at three different concentrations (5, 10, and 15 %) and three different solid contents (30, 40, and 50%) and paper quality at different coat weight was tested, for opacity, brightness, gloss, IGT and Bendtsen roughness. No significant differences were detected in any of the paper quality parameters. Singh et al. (1999) achieved separation of clay particles close to 100%. VSEP (Vibratory Shear Enhanced Processing) has shown 60% total solids recovery. They can be recycled to the coating preparation system and 99% TSS reduction was observed while the coating effluent was concentrated from 1.5-2% to 60% by weight (New Logic).

Application of Ultrafiltration on Paper Machine Effluent

Jonsoon et al. (1985) achieved a separation of 99% for suspended solid from paper machine white water with a laboratory scale ultrafiltration process. Nuortila-Jokinen et al. (1995) and Milan et al. (1999) observed 10-20% reduction in COD, and 100% reduction in SS and turbidity. Elefsiotis et al. (1995) indicated that TDS, soluble COD, and TOC removal efficiencies were affected by membrane molecular weight cut-off, but were independent of the operating temperature, in the 20 to 60 ^{o}C range. Except for fatty acids, where average removals exceeded the 90% level, the separation efficiency of the process for all other parameters (TDS, soluble COD, TOC, and resign acids) was rather moderate, ranging from 10 to 41%. Nuortila-Jokinen et al. (1996) stated that the removal of chemical oxygen demand (COD) is usually lower than 50% depending on the source of the effluent and the type of membrane used. Ramamurthy et al. (1996) reported that ultrafiltration is a very promising method to close white water circuit. Tardif et al. (1997) compared an aerobic membrane biological reactor (MBR) and ultrafiltration (UFT) technique for white water recirculation. Both processes achieved greater than 98% removal of fatty acids and modest success at removing total dissolved solids and colour.

Application of Ultrafiltration on De-Inking Plant Effluent from Wastepaper based Paper Mill

The main contaminants in wastepaper-based mill come from de-inking plant. There are some works reported to treat de-inking plant effluent by ultrafiltration membrane technique. Desaulniers et al. (1970) attained a maximum solids content of 31% when ultrafiltering an aqueous dispersion of activated carbon particles in a batch cell using an ultrafiltration membrane. Bradley et al. (1997) found that ultrafiltration achieved complete retention of the flexographic pigment, producing clear permeates. Stable flux was observed during the experiments, while the ink concentration steadily increased from 0.05% to 0.15%, indicating that flux was independent of solids concentration. Bruno et al. (1998) concluded that ultrafiltration efficiency remained close to 100% resulting in clear permeates for all flexo inclusion rates studies.

Application of Ultrafiltration on Common Effluent

Nuortila-Jokinen et al. (1994) observed that COD and residual lignin were reduced by only 50% using UF. Nuortila-Jokinen et al. (1998) studied ultrafiltration for common effluent treatment and concluded that, depending on the membrane material and cut-off, UF removes about 30-60% of the COD. Koyuncu et al. (1999) did the pilot plant study and concluded that, overall removal efficiencies of COD, Colour, Conductivity, NH_3-N were found as 90–95%, 95–97%, 85–90% and 80–90% respectively with 85–90% recovery after UF and RO membranes for all these wastewater streams. Vieira et al. (2001) observed PVA (polyvinyl alcohol) and PEI (polyethyleneimine) removal by the PVDF (polyvinylidene fluoride) membrane was over 95% for all conditions used in the experiments. When PVA was used, 54% of Fe and 50% of COD removal was achieved in 24 h of contact, and 83 and 55% in 120 h of contact, respectively. Low et al. (2005) observed combined VSEP (Vibratory Shear Enhanced Processing) and membrane bioreactor system for treatment of pulp and paper mill effluent. They concluded that VSEP was placed in extreme operating conditions with the TMP (trans-membrane pressure) 15 times higher than the SMBR (submerged membrane bio reactor) that offered a 7.8 times higher in the initial flux. The intensive membrane oscillation, 26 times higher than the SMBR with lengthwise oscillation, managed to maintain the stabilized flux at 70% of the initial value. This value was still 6.8 times higher than that of the SMBR with lengthwise oscillation.

Conclusion

It is clear from the above discussion that the application of ultrafiltration membrane in pulp and paper industry has already come out from pilot plant phase to plant phase in some of the mills. The best approach would be to segregate the waste water from each section like pulping effluent, paper machine and bleaching effluent and apply ultrafiltration treatment operations. It is advisable as ultrafiltration treatment is highly efficient for specific effluent stream and also it is a costlier treatment method. By segregating streams and treating it by ultrafiltration, fresh water consumption can be reduced. A very good example is to treat coating plant effluent to recover chemicals approximately 100% and reuse the water. Similarly E-stage effluent can be treated separately and permeate can be used in paper section while energy recovery is possible from the concentrate. Hence reduction in freshwater consumption and pollution load can be achieved simultaneously. Recent advance like VSEP membranes can further address the problem of fouling. It needs due consideration in pore size, pore structure of the membrane and oscillation of membrane setup.

In India application of ultrafiltration has not been carried out in mills but lab and pilot scale work is in the progress. The ultrafiltration process is expected to reduce the load on the biological treatment process while improving the efficiency of the treatment. At the same time, the concentrated stream, intended for recycling to the recovery system, contains only a fraction of the troublesome inorganic substances of the alkaline bleach effluent. Effective plan for recirculation of various treated streams would enable mills not only in complying the environmental standards but can also reduce the operational cost.

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Calendar of Events

Climate Change and Urban Poverty–Infrastructures of Development
28 January 2009
Dhaka, Bangladesh
Website: http://www.bwpi.manchester.ac.uk/events/conferences/index.html
Contact name: Shahana Siddiqui
Organized by: BRAC Development Institute; Brooks World Poverty Institute

6th Everything About Water Expo 2009
28 to 30 January 2009
New Delhi, India
Website: http://www.eawater.com/expo2009
Contact name: Shivani Gupta
Organized by: Everything About Water

International Conference on CLIMATE CHANGE IMPACTS AND ADAPTATION STRATEGIES FOR BANGLADESH
18 to 20 February 2009
Dhaka, Bangladesh
Website: http://www.buet.ac.bd/climate2009/
Contact name: M. Habibur Rahman
Organized by: International Training Network (ITN) - BUET, Department of Civil Engineering, Bangladesh University of Engineering and Technology, Dhaka, Bangladesh

The 4th Annual Water Symposium
20 February 2009
Sydney, New South Wales, Australia
Website: http://www.legalwiseseminars.com.au
Contact name: Gemma Goodingham
Organized by: Legalwise Seminars

International Conference on Implementing Environmental Water Allocations
23 to 26 February 2009
Port Elizabeth, South Africa
Website: http://www.wrc.org.za
Contact name: Cilla Taylor, Conference Secretariat
Organized by: Water Research Commission -SA/DWAF-SA/IUCN/IAHS

44th Central Symposium on Water Quality Research
23 to 24 February 2009
Burlington, Ontario, Canada
Website: http://www.cawq.ca/en/44c/
Contact name: Naomi Warrier
Organized by: Canadian Association on Water Quality (CAWQ), in concert with the National Water Research Institute (NWRI) of Environment Canada

Mega Water Expo 2009
26 to 28 February 2009
Chennai, Tamil Nadu, India
Website: http://watertoday.org
Contact name: S.Shanmugam
Organized by: Water Today

Managing Waste in a Changing Climate
4 to 6 March 2009
Launceston, Tasmania, Australia
Website: http://www.taswasteandclimatechange.com.au
Contact name: Kylie Hood
Organized by: Tasmanian Department of Economic Development & Tourism

5th World Water Forum
16 to 22 March 2009
Istanbul, Turkey
Website: http://www.worldwaterforum5.org/index.php?id=1870&L=0
Contact name: 5th Forum Secretariat
Organized by: World Water Council