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Abstract

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Keywords

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Development of a submerged membrane fungi reactor for textile wastewater treatment

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Abstract

A submerged microfiltration membrane bioreactor implementing the white-rot fungus *Coriolus versicolor* was developed for the treatment of textile dye wastewater following explorations with different fouling-prevention techniques. The optimum combination ensuring permeate quality and precluding membrane fouling comprises of placing a bundle of hollow fibers within a non-woven coarse-pore (50-200 μm) mesh-cage, so as to avoid direct deposition of sludge onto it, together with arrangements for its periodic high pressure back-washing (3 sec/10 min.) and chemical back-flushing (100 ml/m², every 3rd day). Under controlled temperature (29 \pm 1 $^{\circ}$ C) and pH (4.5 \pm 0.2), and applied HRT and an average flux of 15 hrs and 0.021 m/d, respectively, the reactor accomplished around 97% TOC and 99% color removal from the synthetic wastewater (TOC= 2g/L; Dye= 100 mg/L) for a prolonged period of observation. Realization of excellent stable pollutant removal along with alleviation of the membrane-fouling problem by employing reasonable chemical-cleaning dose presents the proposed novel system as an attractive one.

Keywords: *Coriolus versicolor*, Decolorization, Submerged microfiltration membrane bioreactor, Textile wastewater, White-rot fungi.

1. Introduction

Textile wastewater is a complex and highly variable mixture of many polluting substances ranging from inorganic compounds and elements to polymers and organic products, and induces persistent color coupled with organic load leading to disruption of the total ecological/symbiotic balance of the receiving water stream [1]. Concomitant with the in-house multi-media pollution prevention efforts, a flurry of end-of-pipe decolorization technologies are being proposed and tested at different stages of commercialization; however, to date none has emerged as a panacea for the same. Cost-competitive conventional biological options are rather ineffective, while physico-chemical processes are restricted

in scale of operation and pollution profile of the effluent. Wood rotting 'white-rot' fungi can aerobically degrade a wide variety of recalcitrant organic pollutants, including various types of dyes, through extracellular secretion of non-specific oxidative enzymes as a secondary metabolic activity in C or N-limited medium [2]. The application of white-rot fungi in large-scale waste treatment, however, has been impeded owing to the lack of an appropriate reactor system capable of coping with rather slow fungal degradation [3], loss of the extracellular enzymes and mediators with discharged water [4], and excessive growth of fungi [5]. In this context, a feasible system may be envisaged by coupling the excellent degradation capability of the white-rot fungi with the inherent advantages of a

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membrane bioreactor (MBR) i.e., suspended solids and macro-colloidal material free permeate, retention of high biomass concentration requiring a small footprint and allowing the process to be operated at a low F/M ratio, hence, yielding reduced excess sludge production. Our previous study [6] reported stable TOC and color removal (97% and 99%, respectively) from a synthetic textile wastewater by a submerged microfiltration membrane reactor implementing white rot fungi *Coriolus versicolor*. The shortcoming of that system, however, was fatal fouling of both of the two types (hollow fiber and flat sheet) of membranes explored. The membrane with horizontally mounted hollow fibers was susceptible to cake-layer fouling, while critical flux dependent pore blocking of the flat sheet membrane was detected. A cylindrical hollow fiber bundle was chosen next as a possible way out of the problem with the expectation that the average cake -layer thickness on each fiber would be reduced owing to the densely packed fiber-bundle; in addition, inter-fibril deposition may be avoided.

This paper seeks to delineate the explorations encompassing the development of a novel submerged microfiltration membrane fungi reactor for treatment of textile wastewater, which ensures permeate quality while avoiding membrane fouling.

2. Materials and methods

This study involved experiments employing a synthetic textile wastewater in a submerged microfiltration MBR, initially inoculated with pure culture of fungi, but operated, other than controlling pH (4.5 ± 0.2) and temperature ($29 \pm 1^\circ\text{C}$), under non-sterile conditions. The investigations carried out within the scope of this study may be categorized under two broad categories- explorations for abatement of the membrane fouling

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problem, and simultaneous investigations on the characteristics of the permeate.

2.1 Microorganism and Synthetic wastewater

The white-rot fungi *C. versicolor*, NBRC 9791 obtained from the NITE Biological Resource Center (NBRC), Japan was used for this study. Batch tests confirmed the decoloration and TOC removal capacity (Fig.1) of the collected fungi strain [7]. A synthetic wastewater (TOC= 2g/L) containing dye (100 mg/L; Poly S119) and starch (two common components in real textile wastewater), along with other nutrients, was utilized. Details about the media have been documented elsewhere [6]. With an attempt to minimize dispersed growth, the fungi were immobilized on Polyurethane foam cubes [8], and the cubes were added to the reactor as stable seed.

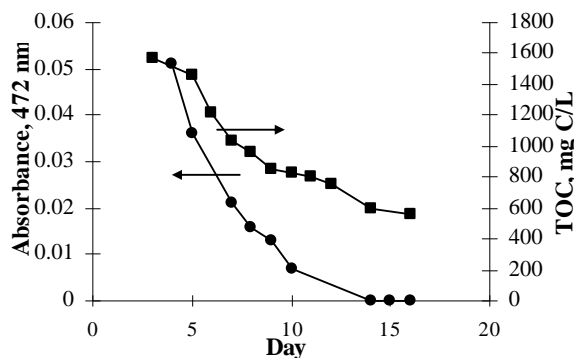


Fig.1 Color and TOC removal by fungi during batch test (Poly S119 dye, 50 mg/L) [7]

2.2 Equipment and operating conditions of the bioreactor

A laboratory scale bioreactor, made of PVC, with a working volume of 12.5 l was used in conjunction with 5 cm bundles of polyethylene hollow fibers (*Clean sui*, Mitsubishi Rayon) having a surface area and pore size of 0.97 m^2 and $0.4 \mu\text{m}$, respectively. In this study, trials with different fouling prevention set-ups were carried out. Details of the intermediate

trials have been outlined in the relevant sections. Fig. 2 depicts the reactor set-up during the final trial. Inside the reactor, the membrane was placed in a cylindrical cage (10 cm ϕ x 28 cm H) of non-woven coarse-pore (50-200 μm) mesh, which served as a pre-filter, hence, avoiding direct deposition of sticky starch and fungi mass on the membrane. In addition, as the afore-mentioned mesh-cage was observed to be inadequate in terms of retaining the critical portion in the size-distribution of the MLSS, a non-woven nylon mesh (#300) was wound around it. A diffuser supplied continuous air from the bottom of the reactor with an intensity of 4L/min for complete mixing and supply of dissolved oxygen to the microbes along with facilitating mesh-cage cleaning. Effluent was filtered out through the membrane by a suction pump with a 5 min. on/off cycle (average flux of 0.021 m/d) resulting in a HRT of 15 hrs. The central pipe of the fiber-bundle was originally used to add glue during forming the platform (located adjacent to the outlet) holding junction of all fibers. The upper half of this pipe was manually perforated; and the core of the fiber-bundle was back-flushed with NaOCl solution containing 3% Cl (100 ml/m², every 3rd day) through a pipe connected to the bottom of this central pipe (Fig. 2b). The membrane was also subject to periodic high-pressure permeate-backwashing (3sec/10min@ 100ml/min) through a V-connection at its outlet. The total back-flush/wash volume was thus restricted within 5% of the daily permeate produced. After using for a certain trial, the used membrane was cleaned chemically with NaOCl solution and reused for further experiments following confirmation of retrieval of the initial flux, or replaced by a new membrane. During the intermediate trials lasting for a short period, the MLSS concentration varied from 7-10 g/L, while in the course of the final trial it increased from 10 g/L to 55

g/L in 50 days in the absence of any sludge withdrawal.

2.3 Analytical methods

TOC was measured with a Total Organic Carbon analyzer (TOC-V, Shimadzu). Color measurements were carried out using a spectrophotometer (U-2010, Hitachi) to measure the absorbance of the sample at the peak wavelength (472 nm) of the dye used. The concentration of dyestuff was calculated from a calibration curve of 'absorbance versus concentration' and concentration values were used for calculations of decolorization efficiency. MLSS concentration was measured according to the standard methods. Transmembrane pressure (TMP), as an indicator of membrane fouling, was continuously monitored using a vacuum pressure gauge (GC 61, JUST). Also direct assessment of membrane fouling was performed through visual observation of the fouled membrane by lifting it up above the water level periodically, and occasionally performing membrane-autopsy.

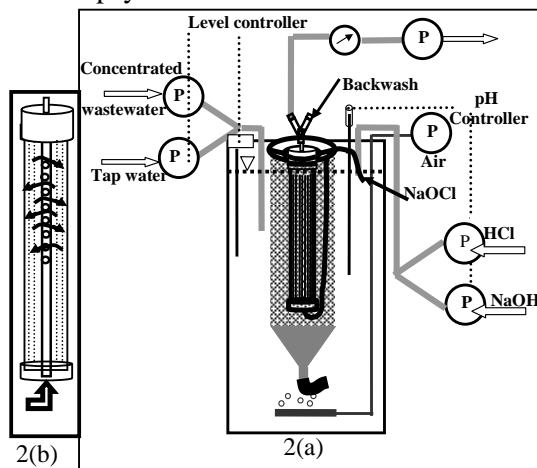


Fig.2. Schematic of the proposed bioreactor system. (a) Membrane within a mesh-cage inside the reactor, (b) Close-up of backflushing of the core of fiber-bundle.

3. Results and Discussion

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3.1 Abatement of membrane fouling

Contrary to the expectation that a hollow fiber bundle may be less vulnerable to external fouling by fungi, a fatal TMP rise was observed (90 kpa within 36 hrs.). This indicated the necessity for some sort of porous covering to prevent direct deposition of fungi on the membrane. Accordingly, in order to solve the membrane-fouling problem, different fouling remediation assemblies, as illustrated in Fig.3, were explored. The intermediate trials, conducted under similar conditions, were continued up to a TMP value of 90 kpa. Fig.4 portrays the trends of TMP increase during different trials.

The bare membrane was vulnerable to massive inter-fibril deposition of the gelatinous fungi-starch mixture, consequently exhibiting a TMP of 90 kpa within 36 hrs. The performance of the membrane with a flexible nylon cloth covering (trial 2), however, appeared to be worse than that of the bare membrane. Repeated investigations revealed that, although the nylon cloth cover could prevent both direct deposition on the membrane and the inter-fibril intrusion of fungi/ starch to a considerable extent, the gradually accumulating thick, non-porous deposition on the cover itself (unaffected by air-scouring), however, led to a very steep rise in TMP (90 kpa, 30 hrs.). The membrane module, following its withdrawal from the reactor at the end of the trial, appeared to have been squeezed and felt very rigid. This observation indicated that while a cover is a must, the use of a cover that is not so tightly pushing around the membrane is required.

In contrast to the performance of the flexible nylon cloth cover, that of the mesh-cage was rather encouraging. Trial 3 (Fig.3), involving arrangements for continuous air scouring within the cage, and also those for periodic- sonication /

removal of settled particles/ permeate-backflushing, sustained for 90 hrs. It is worth-mentioning here that periodic back-flushing (1 min./ 30 min. @ 21 ml/min.; restricting total daily back-flush volume

within 5% of total permeate) of the inner part of the fiber-bundle with permeate was introduced in order to avoid gradual build-up of rigidity of the membrane as observed during the previous trial. The continuous aeration within the cage (1L/min.) and periodic application of ultrasonic vibration (Branson sonifier, 2 min./day) were intended, in addition to cleaning the

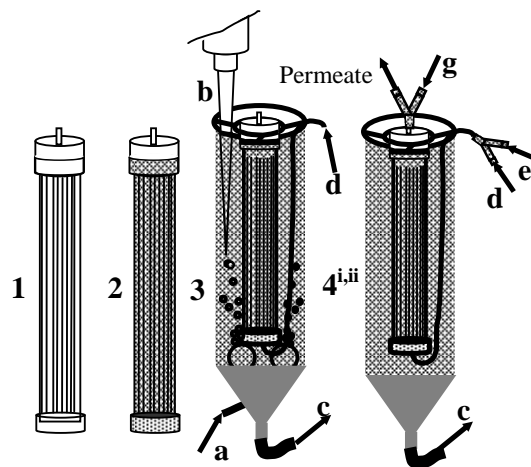


Fig.3. Schematic of different trials
1. Bare membrane; 2. Flexible nylon cloth cover pushing tight on membrane; 3. Mesh cage with arrangements for air scouring (a), sonication (b), removal of settled particles (c) and permeate-backflushing (d); 4. Mesh cage with arrangements for permeateⁱ or NaOClⁱⁱ-backflushing (d or e), permeate-backwashing (g).

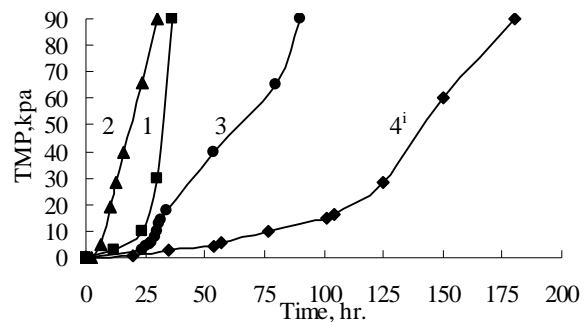


Fig.4. Variation of TMP during different trials (Numbers corresponding to those of figure3.)

membrane, to facilitate prevention of gradual build up of thick deposition on the mesh-cage. However, the moderate performance of this assembly, despite the conceptual expectations, led to further repeated investigations, which revealed that, although sonification was an effective means for cleaning the mesh-cage, the turbulence generated during its application led to intensive dispersion of the very fine fractions of MLSS trapped inside the cage. Eventually those very fine particulate matter, in addition to fouling the surface, found their way into the inner portion of the fiber-bundle, initiating irrecoverable rigidity. The continuous aeration imposed similar adverse impact on the membrane performance.

Trial 4ⁱ (Fig.3), involving back-flushing (1 min./ hr. @ 12 ml/min.) and back-washing (3sec/10min @100ml/min.) of the membrane by the permeate, but without creating any sort of turbulence (aeration/ sonication) within the cage, exhibited further improved performance. The TMP increased to only 28 kpa in 120 hrs. While the initial rise in TMP was slow, at the later stage, typically after a TMP of 15 kpa, the increase was fast, eventually reaching 90 kpa within 180 hrs. During this trial, in absence of any turbulence within the cage, the outer fibers of the fiber-bundle appeared to be clean. However, the membrane-core still seemed to be considerably susceptible to fouling by the very fine particulate fraction of MLSS (Fig.5), although to a lesser extent than that under constant aeration.

The trial employing NaOCl-back-flushing (4 min./ day @ 35 ml/min.), along with back-washing by the permeate (3sec/10min@100ml/min.) surpassed all the previous trials in terms of fouling prevention (Fig.6). With such an assembly, there was a meager rise of 12 kpa in TMP during the first two weeks. The TMP further dropped to 3 kpa and remained unchanged for a week when the duration of back-flushing with NaOCl was

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increased (from 4 min. to 10 min.), while keeping the chemical dose the same or even reducing it a bit. Around the 20th day of continuous operation with the latest combination, the fouling of the mesh-cage caused water level decrease within it. This led to increase in TMP to some extent, although not so critical. At this point, the scouring action of the applied aeration became inadequate as the MLSS concentration in the reactor rose up to around 30 g/l (Fig.6). Accordingly, this problem was solved by providing additional intermittent aeration (5min./hr.@10L/min) with a tube diffuser placed spirally around the cage. Following this, even with reduction of the chemical back-flushing frequency, applying it on every 3rd day rather than daily, the TMP remained stable around 4 kpa for the remaining period of operation. It is worth mentioning here that with chemical washing every 3rd day involving a small dose (100ml/m²), the total NaOCl consumption in three months would be 3L per square meter of membrane area, which is comparable to the dose recommended by the membrane supplier for its commercialized modules during one

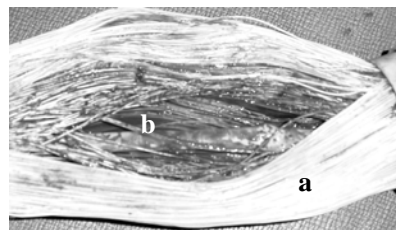


Fig.5. Appearance of fiber-bundle within mesh-cage in absence of chemical back flushing. (a) Cleaner outer fibers, (b) Fouled core.

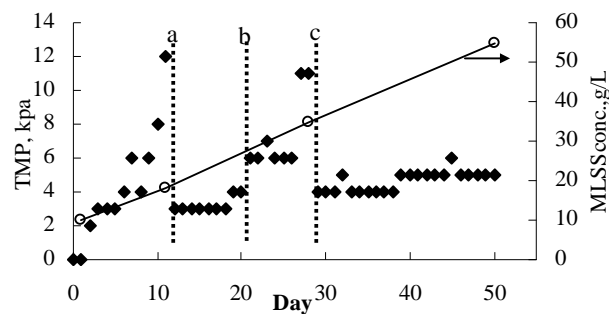


Fig.6. Variation of TMP during final trial (4ⁱⁱ). (a) Chemical-wash duration increased (volume same), (b) Onset of Outer cage fouling, (c) Amendment of outer cage fouling, and reduction of chemical-wash frequency.

prescribed cleaning every three months.

Since a tremendous increase in MLSS concentration (7-55 g/L) in the absence of sludge withdrawal was encountered, elucidation of its probable effect on the observed membrane fouling was deemed imperative. Fig.7 plots TMP values as a function of MLSS concentration during different trials. It appears that the extent of fouling was independent of MLSS concentration itself, rather more influenced by the efficiency of the fouling prevention strategies adopted. In view of the intensive dispersed growth of fungi, it is, however, obvious that immobilization of fungi on bio-carriers could not realize its intended purpose.

3.2 Pollutant removal

Table 1 furnishes the average color and TOC removal performances of the MBR during different trials. Average biological color and TOC removal, as indicated by the supernatant quality, were 68.3% and 95%, respectively. The remarkable low concentration of TOC in the supernatant may be ascribed to the good settling property of starch-the main TOC contributor in this study. All the trials revealed comparable removal efficiencies, the one associated with the nylon cloth- cover (99.9% and 98.2%, respectively), however, marginally surpassed the others. That cover, by encouraging massive attachment of starch and fungi over it, resulted in better removal through the sieving action offered by the cake-layer. Notwithstanding this seeming advantage, such cover is not acceptable from the point of view of TMP increase.

Conversely, the trials (#3,4) involving the mesh-cage exhibited slightly deteriorated performance. This may be attributed to negligible cake-layer on the membrane (unlike the previous trial), when placed amidst media pre-filtered through the mesh cage. In this case the

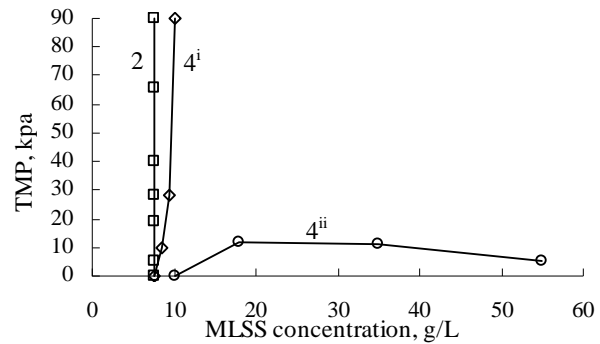


Fig.7. TMP-variation independent of MLSS concentration during different trials. (Numbers corresponding to those of figure 3).

Table 1. Average color and TOC removal performances

Trial No. ^a	Decoloration (%)	TOC removal (%)
1	99.68	98
2	99.90	98.2
3	98.42	97
4 ⁱⁱ	96.83 ^b -99.05 ^c	97-98

^a refer to fig.3; ^{b,c}Initial and Later stage(fig.8)

Note: Permeate appears slightly colored with a decoloration of 98%. Average biological color and TOC removal, as indicated by supernatant quality, were 68.3% and 95%, respectively.

soluble portion could easily go through the utilized membrane of 0.4 μ m. This hypothesis was confirmed by a simple investigation. Two samples, one collected from within the cage and another from the surrounding media, were filtered through laboratory syringe-end filters of 0.45 μ m pore size. The absorbances (472 nm) of the permeates obtained were similar to those of the permeates obtained during trial 4 (“mesh-cage”) and 2 (“nylon cover”), respectively, substantiating the role of cake-layer on the membrane surface in decoloration. Accordingly, during the initial stage of the final trial (Fig.8), a fluctuating removal performance, especially that of color, was observed. The decoloration performance of the reactor, however, improved to a considerable extent (99.05%) and remained stable throughout the later stage of operation

with the tendency of development and maintenance of an optimum layer (enough for appreciable dye retention, yet not causing a TMP rise) of fungi on the membrane surface. Stable decoloration performance was accompanied by steady TOC removal. Furthermore, considerable extent of mineralization was evidenced by the remarkable diminution of absorbance of permeate in the UV range (Fig.9).

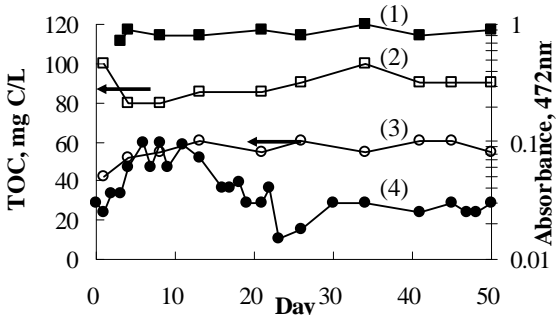


Fig.8. Variation of absorbance (1,4) and TOC (2,3) in supernatant and permeate, respectively, during final trial (4th).

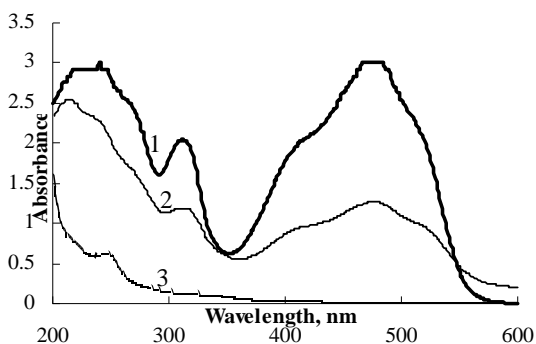


Fig.9. Typical observed change in UV-VIS spectra, indicating dye-mineralization. (1) Influent, (2) Supernatant, (3) Permeate.

4. Conclusions

Accomplishment of excellent stable pollutant removal (99% color and 97% TOC removal), along with the alleviation of the membrane-fouling problem by employing a reasonable chemical-cleaning dose (100 ml/m², every 3rd day) presents the proposed novel system as an attractive one. In view of the fact that in practical submerged MBRs the membranes are placed within a casing, utilization of the

mesh-cage in the present study does not impose much additional technical requirement. Besides, since there was a distinct indication that the system would offer reasonable performance even under further reduced chemical back-flushing frequency, realization of a cost-effective system is very likely.

Nevertheless, further scopes of optimization, for instance, dealing with excessive sludge growth due to high loading, enhancement of biological activity, assessment of performance with more representative wastewater etc., may be indicated. Especially, the extreme vulnerability of the bare membrane to fouling necessitates development of an appropriate membrane module so that the proposed system may enjoy more flexibility in terms of precluding fouling.

Notwithstanding the probable scopes of optimization, the developed laboratory scale prototype utilizing an available membrane module shows great potential in terms of devising an excellent membrane based biological treatment system for textile wastewater.

References

- [1] P.C.Vanndevivera, R.Bianchi, W. Verstraete, Treatment and Reuse of Wastewater from the Textile Wet-Processing Industry: Review of Emerging Technologies. *J. Chem. Technol. Biotechnol.*, 72 (1998) 289-302.
- [2] Y. Fu, T. Viraraghavan, Fungal decolorization of dye wastewaters: a review. *Bioresour. Technol.*, 79 (2001) 251-262.
- [3] M. T. Moreira, C.Palma, G.Feijoo, J. M. Lema, Strategies for the continuous production of ligninolytic enzymes in fixed and fluidised bed bioreactors. *Journal of biotechnology*, 66 (1998) 27-39.
- [4] F. Zhang, J. Yu, Decolourisation of Acid Violet 7 with complex pellets of white rot fungus and activated carbon. *Bioprocess Engineering*, 23 (2000) 205-301.
- [5] F.M. Zhang, J.S.Knapp, K.Tapley, Development of bioreactor system for decolorization of Orange II using white rot fungus. *Enzyme Microb. Technol.* 24(1999) 48-53.
- [6] F.I. Hai, K. Yamamoto and K. Fukushi, Different fouling modes of submerged hollow-fiber

and flat-sheet membranes induced by high strength wastewater with concurrent biofouling. Desalination (In press, DES 3212).

[7] F.I. Hai, K. Fukushi and K. Yamamoto, Treatment of textile wastewater: Membrane Bioreactor with special dye-degrading Microorganism. Proc. Asian Waterqual, Bangkok, Thailand, 2003, 2Q3F16.

[8] M. Fujita, A. Era, M. Ike, S. Soda, N. Miyata, T. Hirao, Decolorization of Heat-Treatment Liquor of waste sludge by a bioreactor using polyurethane foam-immobilized white rot fungus equipped with an ultramembrane filtration unit. J. Bioscience and bioengineering, 90(4) (2000) 387-394.