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Fouling control of a membrane coupled photocatalytic process treating greywater

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ABSTRACT

Fouling in membrane coupled photocatalytic reactors was investigated in the case of greywater treatment by establishing the link between product type, dose, irradiation time and fouling rates in a cross flow membrane cell fitted with a 0.4 μm pore sized polyethylene membrane. Rapid fouling occurred only with shower gels and conditioners and was linked to changes in the organo-TiO₂ aggregate size postulated to be caused by polymers within the products. Fouling was reduced to a negligible level when sufficient irradiation was applied demonstrating that the membrane component of the process is not the issue and that scale up and implementation of the process relates to effective design of the UV reactor.

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1. Introduction

A wide range of new engineered nanoparticles are becoming available for use in water and wastewater treatment (Jefferson, 2008). Recent examples include nano silver coatings on socks to inhibit microbial growth and hence odour (Ross, 2004) and zero valent iron nanoparticles for groundwater remediation (Huang et al., 2008; Ahmadimoghaddam et al., 2008). Whilst research is continuing into developing new nanoparticles actual uptake of the existing ones is rather limited in water treatment (Jefferson, 2008). The problem is a classical chemical engineering one: how to implement (scale up) a nano scale process at the meso or macro scale (Wintermantel, 1999). For context, water treatment facilities vary greatly but typically treat flows in the order of 10s–100s of ML d^{−1}. Converted to nanoparticles, which are typically in the

size range 1–100 nm this equates to 7.2×10^{26} particles and thus there is a challenge. The nanoparticles provide very large specific surface areas with which to provide high mass transfer and reaction kinetic coefficients yet fixing such a large number of very small particles so that they remain in the treatment process and do not exit with the product water is extremely challenging. Typical energy and operating costs for water treatment are in the order of 0.5 kW h m^{−3} and €0.2–0.5 m^{−3} which means solutions cannot be overly complicated or complex. Reported solutions to the problem involve either immobilisation to solid substrate (Rachel et al., 2002) or retention by filtration with membranes (Rivero et al., 2006; Chin et al., 2007).

One embodiment of this concept is the membrane chemical reactor (MCR) (Parsons et al., 2000; Jefferson et al., 2001) which utilises nano sized titanium dioxide (TiO₂)

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particles in combination with a UV light source to generate highly reactive hydroxyl radicals which have a redox potential of 2.33 V, only surpassed by F_2 (Huang *et al.*, 1993). The TiO_2 particles are retained in the system by means of a membrane filtration unit that is configured externally to the membrane but operated in an air lift, low pressure manner equivalent to that of a submerged membrane system (Le Clech *et al.*, 2003). Long term trials for the treatment of greywater have shown it to be an effective system comparable to that of a membrane bioreactor (Pidou *et al.*, 2008a,b). For instance, average effluent residuals of below 10 mgL^{-1} for bio-chemical oxygen demand (BOD), below 1 NTU for turbidity, below 2 mgL^{-1} for suspended solids (SS) and no pathogens were observed throughout the trial at a hydraulic residence time of 2 h (Pidou *et al.*, 2008a,b). The observed residual levels mean that the technology is viable for treating greywater to the most stringent water quality standards available for urban reuse (Pidou *et al.*, 2007). Consequently, it provides an alternative to biological systems such as membrane bioreactors (MBRs) where the small scale of operation, proximity to the end users and the potential for toxic shocks provides a relatively high process failure risk (Jefferson *et al.*, 1999; Knops *et al.*, 2007). Similar high performance of photocatalytic systems have been reported for the treatment of dyes (Molinari *et al.*, 2002; Mozia *et al.*, 2007), humic acid (Lee *et al.*, 2001; Fu *et al.*, 2006; Erdei *et al.*, 2008), bisphenol A (Thiruvengatachari *et al.*, 2005; Chin *et al.*, 2007) or pesticides (Oller *et al.*, 2006; Lhomme *et al.*, 2008) hence the appropriateness of the technology as a treatment solution is well established.

However, during the greywater investigation significant membrane fouling was observed. Consequently, the system could only be run for about 10 days at a flux of $5\text{ L m}^{-2}\text{ h}^{-1}$ (LMH) before a chemical cleaning of the membrane was necessary (Pidou *et al.*, 2008a,b). This was found to be contradictory to results of a previous study in which the MCR pilot plant was operated in batch mode (Rivero *et al.*, 2006). Very little or no fouling was observed during the batch experiments for fluxes up to $120\text{ L m}^{-2}\text{ h}^{-1}$. Such differences in operation are surprising but the results obtained during the batch operation tests can be explained by the fact that the greywater was rapidly treated and consequently for the higher fluxes the TiO_2 was dispersed in fairly clean water and very little or no fouling was observed. This suggests that the fouling propensity of TiO_2 changes significantly in the presence of a waste, in this case greywater.

A paucity of literature on operation of such photocatalytic hybrid membrane systems, especially for medium to high strength organic wastes, potentially limits the uptake of the technology to full scale operation. Specifically two key questions remain unanswered: (1) how to develop systems that can treat sensible flows whilst ensuring all the TiO_2 in the system is active and hence degrades the organics and (2) how to ensure the membranes does not foul in systems that answer question 1.

The current paper addresses question 2 by examining the impact of different greywater products on the fouling behaviour of the system elucidating the major changes in the system when fouling occurs.

2. Materials and methods

2.1. Filtration system

A bench-scale filtration system was used to replicate the fouling experienced when operating the membrane chemical reactor (MC-R™) (Water Innovate Limited, UK). Trials were conducted to study the influence of different parameters on titanium dioxide (TiO_2) and its properties to foul membranes. This system was composed of a 9-l PVC tank in which the TiO_2 and greywater slurry was placed. The slurry was pumped across the membrane module (Perspex, $28\text{ cm} \times 20\text{ cm} \times 8\text{ cm}$) and back to the reactor at a cross flow velocity of 0.16 m s^{-1} . The treated water was permeated through the membrane by a peristaltic pump (505Du, Watson-Marlow, UK) and a pressure transducer (RS components, UK) was fitted in the permeate line to record the trans-membrane pressure (TMP). Because it was a batch system, the permeate was sent back into the tank to avoid any volume loss. The membranes used in the module were polyethylene sheets with a pore size of $0.4\text{ }\mu\text{m}$ and a surface area of 0.019 m^2 .

2.2. Methods

A range of products (all bought in a supermarket) including shower gel, shampoo, bathroom cleaner, conditioner, hand soap and bubble bath were diluted in tap water at a concentration of 2 or 3 g L^{-1} and placed in the reactor with 5 g L^{-1} of TiO_2 . These concentrations were chosen because they represent the upper range for organic concentrations reported in the literature in terms of COD in feed greywaters from sampled sites around the world (Pidou *et al.*, 2007). The COD concentration of the products were measured prior to use at 360, 280, 280, 250, 190 mg L^{-1} for the shower gel, conditioner, bubble bath, shampoo and hand soap respectively. Indeed, concentrations of products of 3 g L^{-1} corresponded to solutions with COD concentrations between 570 and 890 mg L^{-1} . The TiO_2 dose was selected from previous trials which demonstrated it be the optimum concentration for greywater treatment (Rivero *et al.*, 2006). Ingredients of each of the products as listed in provided in Table 1. Tests using the flux step method generally used to characterise membrane fouling in MBRs (Le Clech *et al.*, 2003) were then carried out and the fouling rates determined for the different slurries. Fouling experiments were conducted under dark light conditions to best replicate practical system where the membrane is configured such that it is not in direct contact with the UV light to prevent damage to the membrane structure (Rivero *et al.*, 2006). All tests were duplicated. For the photo-catalysis tests, the slurry was placed in a stirred tank under a 100 W UVA lamp (Black-Ray, CA, USA) for a range of time ranging between 0.5 and 2 h.

2.3. Analytical procedures

Particle sizes were measured with a Malvern Mastersizer 2000 particle analyser (Malvern Instruments Ltd, Worcestershire, UK). It was not possible to measure the particle size with the TiO_2 concentration of 5 g L^{-1} as the detection cell saturated

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