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# Carwash wastewater treatment by micro and ultrafiltration membranes: Effects of geometry, pore size, pressure difference and feed flow rate in transport properties

Alinne Clície Salgueiro Pinto[a], Luiza de Barros Grossi[a], Rafael Alvarenga Carvalho de Melo[a], Thalles Macedo de Assis[a], Vanessa Mendes Ribeiro[a], Miriam Cristina Santos Amaral[b], Kátia Cecília de Souza Figueiredo[a,*]

[a] Department of Chemical Engineering, School of Engineering,Federal University of Minas Gerais. Avenue Presidente Antônio Carlos, 6627, 31270-901, Belo Horizonte, Minas Gerais, Brazil

[b] Department of Environmental Engineering, School of Engineering,Federal University of Minas Gerais. Avenue Presidente Antônio Carlos, 6627, 31270-901, Belo Horizonte, Minas Gerais, Brazil

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## ABSTRACT

Micro and ultrafiltration hydrophilic membranes were investigated for carwash wastewater reclamation. The effects of geometry as well as operating conditions such as pressure difference across the membrane and feed flow rate were investigated aiming the characterization of permeate flux and quality and evaluation of water reuse. The effluent showed initial turbidity of 85 NTU, in addition to total organic and inorganic carbon of 4.1 and 58 mg/L, respectively. Tests in flat cellulose commercial membranes revealed that both micro and ultrafiltration showed good retention of solids and organic matter, as shown by turbidity and chemical oxygen demand. In order to increase the productivity, commercial hollow fiber polyetherimide membranes were investigated. The initial flux was 440 L/m$^2$h, with pressure difference of 0.4 bar and Reynolds number of 400. The final permeate with recovery rate of 80% showed a total organic and inorganic carbon of 2.7 and 35.4 mg/L, respectively. The rejection was 98.6%. The results showed that this approach has high potential to be used for water reuse depending of the number of cycles or recycling ratio is aimed.

## 1. Introduction

Water scarcity has become one of the most difficult challenges to the populations worldwide. The improvement in water management is mandatory and, as the consequence, there are many regulations in order to preserve fresh water, increase the reuse percentage and dispose effluents [1]. Carwash industries are among the activities that consume large volume of water and can benefit from recycling programs. In the washing of one vehicle, depending on the type of carwash installation and the size of the car, it is reported that the average of 150–600 L of wastewater is produced [2]. Considering the large volume of wastewater, its complex composition and the quest for the sustainability of carwash industries, the effluent treatment system is not just a process to meet the environmental discharge requirements but also is an opportunity to recover valuable water for reuse purpose [3].

The use of fresh water per vehicle has already been limited in some countries such as the Netherlands, the Scandinavian countries (both with 60–70 L/car) and also Australia (100 L/car), which leads to an increasing demand for recycling the wastewater stream [4,5]. Besides that, the use of rollover systems are always preferable compared to the automatic ones, specially due to water demand, which is 80% lower [6].

Attempts to increase the fraction of recycled water in the process have been investigated for over 20 years [7], but the main barriers to accomplish this goal are the local quality of fresh water, the broad range of degreasing agents used to perform the carwash, the fluctuation in physicochemical and biological properties of such effluent along the year, as well as the system used in the facility (rollover or automatic). As the result, many different processes have been proposed, such as filtration by means of sand filter or membranes, flocculation, air dissolved flotation, sedimentation and adsorption (ion exchange). The use of process integration to decrease energy consumption and meet the sustained requirements is a real tendency [8]. Regarding the presence of high organic matter content in some effluents, it is also desirable, in

* Corresponding author.
*E-mail address:* katia@deq.ufmg.br (K.C. de Souza Figueiredo).

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some cases, to reach the decrease in chemical and biochemical oxygen demand, COD and BOD, respectively, by using activated sludge, ozonation or reaction with HF, for instance [5,7,6,9,10,11,12,13,14].

Upon considering the complexity of the system and its variability [15], the use of decentralized systems is seen as a potential solution to treat and reuse the effluent [16,6]. This approach can diminish some of the fluctuations stated above and increase the removal of some pollutants from the effluent.

The properties of water for reuse have been established by World Health Organization, WHO, and US Environmental Protection Agency [17], but they are very stringent, which makes the treating processes unpractical. A less conservative point of view considers that the main risks that should be attained in such systems are the chemical, usually associated with corrosion, and microbial, regarding the operator health safety [6].

The use of microfiltration to treat the effluent of carwash and decrease the risks (both chemical and microbial) is an attractive alternative due the ability of such membranes to retain suspended solids and cells, with lower demand for chemicals compared to other methods, such as flocculation/sedimentation or flotation [18]. However, ultra and nanofiltration membranes are mainly used to address the removal of macromolecules and medium molecular weight species, in addition to sterilization. For instance, Boussu and coworkers studied both ultra and nanofiltration processes to carwash reclamation and verified the importance of water recycling to meet the environmental requirements [4]. The use of hybrid systems are also investigated in other papers, such as the addition of bio carriers to non-woven membranes [19] and coagulation/adsorption in addition to membranes [4].

Kiran et al. [5] investigated the use of bentonite-modified ultrafiltration cellulose acetate and poly(ether sulfone) membranes for the treatment of carwash effluent. The results showed that the hydrophilic cellulose acetate membrane had better performance in COD and turbidity removal, as well as lower flux decrease due fouling [5]. Lau et al. [9] investigated nano and ultrafiltration commercial membranes aiming effluent reuse. The results showed that the hydrophilic negatively charged NF270 membrane was able to reject ions and total dissolved solids as well as COD and suspended matter (which caused turbidity) with low flux decrease (15–10%) in 3 h of test. The authors confirmed that hydrophilic membranes had a superior performance compared to the hydrophobic ones, and showed that the process is very dependent of feed solution properties [9]. Hamada and Miyazaki [10] investigated the use of a mixture of flocculants as the pre-treatment of ultrafiltration. After flocculation, the supernatant was fed to cellulose acetate hollow fiber modules with 32 and 48 $m^2$. The resulting flux was 780 $L/m^2h$, for 100 kPa during 6 months, and the COD and BOD contents in permeate varied from 3.7 to 15.7 mg/L and from 2.5 to 14 mg/L, respectively. In summary, the research concerning the use of membrane in carwash industry is mainly limited to the use of ultrafiltration and nanofiltration and the main problems when using these processes were related to membrane fouling [20,21].

In this context, the goal of this work was to investigate the performance of hydrophilic ultra and microfiltration membranes in terms of permeate quality and membrane fouling, with no pretreatment, since the use of chemicals can increase the operating costs. Tests were conducted for a real carwash wastewater. The application of such route was evaluated for reuse purpose and the impacts of recirculation in some parameters, such as conductivity, were addressed.

## 2. Materials and methods

### 2.1. Carwash wastewater

The wastewater sample was supplied by Autoposto Trevinho, a gas station in Belo Horizonte, Minas Gerais, Brazil, equipped with a rollover carwash system. The grab sample was collected from the washing box, before any kind of treatment. Sampling was made in September (CW1), October (CW2) and November (CW3), aiming to address the fluctuation of physicochemical properties of wastewater. This period comprises very low air humidity (in September) as well as the beginning of the raining season in our region (October and November), so that it would correspond to a real fluctuation of the wastewater physicochemical properties profile. Samples were properly stored to avoid changes in such parameters.

### 2.2. Membranes

Four flat membranes were investigated in order to treat the effluent: three microfiltration membranes with average pore diameters of 0.8, 0.45 and 0.22 μm, and one ultrafiltration membrane with molecular weight cut-off of 100 kDa (equivalent to about 0.01 μm). All membranes used were cellulosic, provided by Millipore®. Membrane pore size was chosen in order to evaluate the size of the species contributing to turbidity. The use of hydrophilic membrane material was set to minimize membrane fouling, according to the literature [5,9]. The reported contact angle ranged from 55 to 60°.

Hollow fiber polyetherimide membrane module with mean pore diameter of 0.4 μm, produced by PAM Membranas Seletivas Ltda, with $2.2 \times 10^{-2}\ m^2$ and packing density of $125\ m^{-1}$ was selected to increase separation productivity. Membrane was outside-in type. The water contact angle of such membrane was 89°.

### 2.3. Experimental procedure

The experimental strategy comprised 4 steps. The characterization of physicochemical properties of samples (1), choice of membrane pore size and geometry (2), evaluation of the best process parameters for wastewater reclamation (3) and definition of recovery rate and permeate quality (4) were addressed in order to study the treated effluent for car washing reuse and the circulation effect on wastewater parameters.

The pH, turbidity (2130 B), conductivity (2510 B), total solids (2540 B), fixed and volatile suspended solids (2540 E), total and soluble chemical oxygen demand (5220 D), COD, oil and grease (5540C), total organic carbon (5310 B), TOC, and inorganic carbon (Horiba, EMIA-Pro), IC were measured according to the standard methods [22] and in triplicates. Bioassays with the bacteria Aliivibriofischeri were performed, using Microtox® Model 500 Analyzer (SDI) equipment, to determinate the acute toxicity of the samples. The tests were performed according to manufacturer's standard protocol.

Tests with membranes were conducted with CW1, the sample collected in September, due to the highest values for turbidity and COD contents.

For the choice of pore size, the flat membranes were placed in a dead end non-stirred filtration system. Sample volume was 180 mL. Test temperature was 25 °C. The experimental apparatus, with effective permeation area of 50 $cm^2$, is schematically presented in Fig. 1 (a). The membrane was pre-treated according to the manufacturer aiming the removal of top protective layer. Usually, the films were immersed in distilled water for 12 h, followed by water permeation (200 mL) in the same pressure difference of the test. Then, 180 mL of the effluent CW1 (density, ρ, of 1.004 g/mL) were permeated through the membranes. Turbidity, pH, conductivity and COD were evaluated in order to choose membrane pore size to run the separation.

Membrane flux was calculated according to Eq. (1), in which m is permeate mass, ρ is its density, A is permeation area, 50 $cm^2$, and Δt is the time interval, while relative flux ($J/J_0$) was the ratio between the flux at a particular time, related to the initial one. Permeability, P, in $L/hm^2bar$, was calculated according to Eq. (2), in which ΔP is the pressure difference across the membrane.