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A novel membrane-based integrated process for fractionation and reclamation of dairy wastewater

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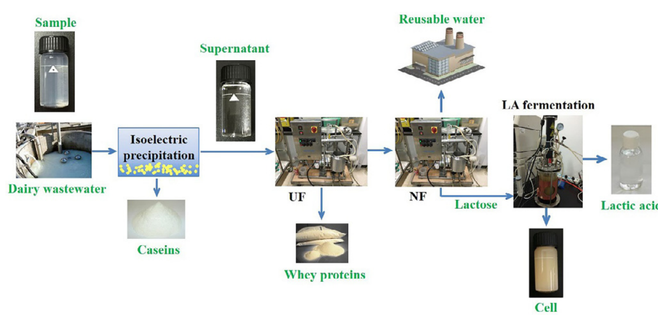
HIGHLIGHTS

- An integrated process recycles water, caseins, whey proteins and lactose.
- This process can mitigate the sludge/retentate disposal and membrane fouling.
- IP-UF pretreatment greatly reduces fouling and concentration polarization of NF.
- NF retentate with concentrated lactose is obtained with IP-UF pretreatment.
- Fermentation fed by NF retentate with IP-UF pretreatment produces more LA.

GRAPHICAL ABSTRACT



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ABSTRACT

An integrated isoelectric precipitation (IP) – ultrafiltration (UF) – nanofiltration (NF) – lactic acid (LA) fermentation process was established for recovering water, proteins, cells and LA from model dairy wastewater (MDW). This process could solve the problems of sludge/retentate disposal and membrane fouling during membrane-based wastewater treatment. The IP process greatly retarded concentration polarization and fouling of UF. However, PES membrane was still severely fouled by whey proteins. XPS results showed that carboxyl groups (C=O) belonging to the proteins was on PES10 surface, but it was not found on Ultracel PLGC membrane. Proteins in MDW with IP were completely retained by Ultracel PLGC UF membrane and lactose almost passed through. After IP-UF pretreatment, the fouling of NF membrane was greatly retarded (irreversible fouling decreased from 44.4% to 11.1% in a pilot-plant test). Thus, the volume reduction ratio was highly increased to 37 and the retentate with high strength lactose of 44.2 g L^{-1} was obtained. With a model NF retentate (45.0 g L^{-1} of lactose) without proteins, 5.42 g L^{-1} of cell mass and 37.6 g L^{-1} of LA concentration were produced by the LA producer *B. coagulans* IPE22.

1. Introduction

A steady rise in the demand for milk and milk products has led to enormous growth of dairy industries in the world. The total milk production in 2014 was estimated at about 802 million tons, a considerable increase of 3.3% compared with 2013 [1]. It is calculated that around 2% of the total milk processed is wasted, resulting in an increasing amount of dairy wastewater [2]. Receiving water can be severely polluted by dairy wastewater without proper treatment. Traditional physical-chemical and biological approaches for dairy wastewater treatment like coagulation/flocculation, anaerobic and aerobic processes have many disadvantages, such as nutrients loss, generations of greenhouse gases (i.e., carbon dioxide and

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Nomenclature

AC	activated carbon
ASR	activated sludge reactor
AnF	anaerobic fermentation
CWS	constructed wetland systems
FO	forward osmosis
J	permeate flux of pure water
IP	isoelectric precipitation
IF	irreversible fouling
LA	lactic acid
LAF	lactic acid fermentation
$L_{p,b}$, $L_{p,a}$	pure water permeabilities before and after filtrations
L_p	permeability
MBR	membrane bioreactor
MFC	microbial fuel cell
mMRS	modified De Man-Rogosa-Sharpe
MD	membrane distillation
MDW	model dairy wastewater
MWCO	molecular weight cut-off
NF	nanofiltration
PES	polyethersulphone
RC	regenerated cellulose
RO	reverse osmosis
TMP	transmembrane pressure
TN	total nitrogen
TOC	total organic carbon
UASB	up-flow anaerobic sludge blanket
UF	ultrafiltration
V_f, V_r	feed and retentate volumes
VRR	volume reduction ratio

methane) and sludge [2–4]. For instance, sludge disposal represents up to 50% of the operating cost of a municipal wastewater treatment plant [5]. Recently, there has been a paradigm shift, from disposing of dairy wastewater to recovering it [6–8]. For example, constructed wetland is one of the green technologies to recover nutrients into plant biomass [6]. Algal cultivation is also applied to convert nutrients to biomass and biodiesel [7].

Membrane-based process is a promising technology for water reuse and resource recovery from wastewater owing to its high separation efficiency, small footprint and ease of operation [9–14]. Nataraj et al. reported that hybrid nanofiltration (NF) and reverse osmosis (RO) process successfully removed contaminants and recovered water from high strength distillery wastewater [9]. Also, Aydiner et al. applied integrated membrane systems (forward osmosis (FO)-membrane distillation (MD) and MD-RO) to recycled water and whey powder from raw whey [15]. However, the major challenges facing widespread application of membrane technologies in wastewater treatment are membrane fouling, concentration polarization, and retentate disposal [12,16,17]. Membrane fouling results in lower productivity, higher operation costs and shorter membrane lifespan. For dairy wastewater, caseins and whey proteins are the principal foulants for membrane [12]. Some properties of proteins in milk are shown in Table S1. According to their properties, hydrophilic membranes are more suitable for dairy wastewater treatment [8]. As mentioned above, concentration polarization is another barrier. Thus, many innovative membrane modules, such as vibratory shear-enhanced filtration system and rotating disk membrane module, are designed to reduce concentration polarization [16]. In addition, pretreatment is an alternative to reduce concentration polarization and fouling, especially at industrial scale [18,19]. Van der Bruggen et al. demonstrated that microfiltration was efficient to avoid the fouling of large particle in NF [18]. Nataraj et al. also used microfiltration as pretreatment step for electrodialysis in treatment of paper industry wastewater [19]. In our previous work, isoelectric precipitation (IP)-NF-anaerobic fermentation process was utilized for recovering dairy wastewater [12]. IP process was firstly proposed as a low-cost approach to reclaim caseins in dairy wastewater. Also, caseins removal greatly mitigated NF fouling and reduced the negative impact of proteins on fermentation. Regarding NF process, it produced reusable water, and the resulting retentate could be utilized for fermentation to produce hydrogen and volatile fatty acids which were capable of precipitating caseins.

However, there are still several problems to be solved in the IP-NF-anaerobic fermentation process. On the one hand, there are whey proteins in wastewater after IP, which probably foul the membrane during the NF concentration process. On the other hand, separation of volatile fatty acids and biogas produced by fermentation are also challenging [20,21]. Thus, to better recover the resource from wastewater, an integrated IP-UF-NF-lactic acid (LA) fermentation process is proposed, as illustrated in Fig. 1. Firstly, IP significantly reduces the fouling and concentration polarization in UF process. Secondly, UF pretreatment can recover high value-added whey proteins and further mitigate fouling during NF concentration. Thirdly, due to IP-UF pretreatment, NF can produce retentate with high strength lactose which is suitable for LA fermentation. Compared to conventional biological processes, such as anaerobic and aerobic processes, LA fermentation produces high value-added lactate, and its extraction and purification is more facile than those of volatile fatty acids. It also yields the cells which can be used as animal feed, rather than deleterious sewage sludge from the aerobic process. This strategy can also be applied for similar biodegradable wastewater if proven feasible.

This work aims at evaluating the feasibility of the integrated IP-UF-NF-LA fermentation process for recovering water, caseins, whey proteins, cells and high strength LA from dairy wastewater, focusing on its advantages of membrane fouling mitigation and retentate/sludge utilization. UF membranes with various pore sizes and surface properties were employed for separating proteins from dairy wastewater, and transmembrane pressure (TMP), antifouling performance, solutes retention and foulants composition on the membrane surface were examined. The effect of pretreatment on NF was also assessed in terms of TMP and irreversible fouling (IF). A pilot-scale test of NF process was further carried out to obtain retentate with high strength lactose. Furthermore, simulated NF retentates with and without proteins were used as the substrate for LA fermentation by the thermophilic *Bacillus coagulans* IPE22 [22]. Fermentative parameters were examined in terms of lactose consumption, LA production, and cell mass accumulation.

2. Materials and methods

2.1. Wastewater

2.381 g of commercial milk powder (Yili Hi-Calcium Skim Milk Powder, Inner Mongolia Yili Industrial Group Co., China) was dissolved in one liter of deionized water for preparing MDW. The MDW was freshly prepared and centrifuged at 8500 rpm in a