Separation and Purification Technology 125 (2014) 126–135

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Feasibility study of microfiltration for algae separation in an innovative nuclear effluents decontamination process

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ARTICLE INFO

Article history:
Received 15 October 2013
Received in revised form 21 January 2014
Accepted 23 January 2014
Available online 4 February 2014

Keywords:
Micro-algae
Microfiltration
Membrane fouling
Hydrodynamic actions

ABSTRACT

Bio-remediation technologies often offer efficiency, cost and environmental impact benefits against physico-chemical technologies. Concerning the remediation of radionuclide-containing water, a few bio-based technologies have been proposed but none is currently operational in highly radioactive environments. A new radio-tolerant micro-alga, isolated from a nuclear facility, possesses properties that offer new decontamination prospects for the nuclear industry or for the clean-up of environmental water. A pilot-scale treatment unit based on this alga is currently under development for the decontamination of radioactive water. It includes separation and/or concentration steps relying on membrane filtration. This work aims at verifying the feasibility of microfiltration as separation step for the targeted algae separation. Recommendations about the choice of operating conditions limiting and/or controlling the membrane fouling are provided with the objective to enhance the separation efficiency. Lab-scale dead-end filtration tests were implemented and the key factors involved in the separation performances were investigated. Membrane characteristics, biomass composition, and hydrodynamic conditions were considered. Organic membranes provided adequate filtration performance. Membrane fouling was essentially induced by a rapid reversible algae deposit and to a lesser extent by irreversible pore blockage caused by smaller particles and dissolved organic matter. To cancel the reversible fouling, hydrodynamic actions such as stirring and back-flush efficiently prevented algae deposit, allowing higher filtration productivity. This study demonstrates the feasibility of membrane separation for micro-algae harvesting at laboratory-scale and specifies the suitable working conditions.

1. Introduction

The nuclear industry generates radionuclides, including radioactive metals, carbon-14 and tritium, which are found in liquid and gaseous effluents. These effluents must consequently be decontaminated before release in order to reduce their activity below controlled thresholds [1]. Technologies frequently used to separate radionuclides from liquid effluents include evaporation, solid/liquid separation by filtration, centrifugation or decantation possibly associated to a chemical precipitation/flocculation process, membrane filtration, sorption, and ion exchange [2,3]. These methods are efficient and robust; however, they are expensive and do not completely remove some radionuclides such as carbon-14 and tritium.

Alternative technologies are needed to reduce radioactive releases in aqueous effluents. Remediation technologies based on micro-organisms are already used in a variety of industrial applications [4–6] and often offer efficiency, low cost and environmental impact benefits against physico-chemical technologies [7]. They may constitute interesting alternatives in the nuclear field as well, but only a few bio-based technologies have been proposed. One study concerns lab scale experiments for the decontamination of a highly radioactive water issuing from Fukushima accident [8]. Real-scale implementation has scarcely been performed and, to our knowledge, only two processes were reported to have been tested at this scale for uranium treatment, whose radioactivity is low [9,10]. The availability of microorganisms adapted to nuclear

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http://dx.doi.org/10.1016/j.seppur.2014.01.039
1383-5866/

conditions, *i.e.* tolerant to high ionizing radiations, nutrient stress and metallic stress, would facilitate the development of such bio-applications for the nuclear industry. Actually, living photosynthetic biomasses, especially microalgae, are a potential remediation matrix because they can take up inorganic carbon-14 via photosynthesis, organic carbon-14 via metabolic pathways as well as a large panel of radioactive metals such as silver-110 m and cobalt-60. A new autotrophic green unicellular micro-alga was recently isolated from a used fuel storage pool of a nuclear facility. This alga is extremely radio-tolerant since it survives to radiation doses of up to 20 kGy and has a high capacity to concentrate the main radionuclides present in nuclear effluents, including carbon-14 and the gamma-emitters ^{238}U, ^{137}Cs, ^{110m}Ag and ^{60}Co [11]. It is therefore of great interest for the development of new remediation solutions for nuclear effluents and for the clean-up of environmental water. The implementation of a pilot-scale treatment unit, based on this micro-alga and including different tasks, is currently under development for the decontamination of radioactive effluents. To ensure the objectives of the process, algae have first to be produced in a growth medium and harvested before ensuring the treatment of the contaminated effluent. Moreover, after the treatment of the effluent, algae must be separated from the decontaminated effluent. The development of such an unusual process is subject to several constraints linked to the use of a biological matrix in a nuclear environment. Firstly, during the harvesting step, algae have to be strictly separated from the growth medium in order to comply with regulatory constraints in nuclear industry. Secondly, after the decontamination step, the separation technology should allow the whole retention of the algae to ensure a perfect treated water quality in terms of suspended solids retention.

In microbiological processes, biomass harvesting and/or dewatering are crucial steps; depending on the final objective (*e.g.* biomass separation from treated water or biomass concentration), the chosen technology can lead to cell concentration or to complete dewatering [12]. The frequently used technologies are based on usual liquid–solid separation techniques, such as centrifugation, filtration, flotation and sedimentation. In most cases, separation must not modify cells integrity and viability. The choice of the separation technique integrates biological constraints, separation efficiency and energy cost. It should be noticed that in algae field, the separation step is currently a technological lock due to high energy cost [13]. Micro-algae harvesting usually relies on centrifugation, sedimentation, flotation or flocculation [12–14]. Recently, membrane filtration has emerged as a promising advanced method for micro-algae harvesting with several advantages not requiring chemical coagulants or flocculants, which reduces the operational cost while ensuring the quality of the biomass [15]. Furthermore, according to the above-mentioned specific constraint of our pilot-scale treatment unit, membrane filtration seems to be the most appropriate separation technology in our specific case.

This work aims at demonstrating the feasibility of microfiltration as separation step for micro-algae separation and at optimizing the filtration efficiency with regard to selectivity and permeability constraints and to biomass fate. Laboratory-scale experiments were performed on a frontal filtration unit to identify the key factors involved in the separation performances, notably in membrane fouling. Three factors known to impact membrane permeability were considered and tested: (i) the membrane characteristics, namely pores diameter and material, (ii) the biomass composition and (iii) the hydrodynamic conditions, namely stirring and back-flush conditions. With the objective to provide recommendations about the operating conditions to be employed at pilot-scale, a particular attention was paid to the influence of the biomass characteristics on the filtration efficiency in the context of the decontamination process; filtration conditions permitting a rapid and easy micro-algae harvesting without cell damage were specifically explored.

2. Experimental

2.1. Algal suspension

2.1.1. Strain, culture and storage conditions

The micro-alga *Coccomyxa actinabiotis* used is described in [11]. Algal biomass was grown continuously in a 10 L photobioreactor, sparged with air at 50 ± 10 L h^{-1}, under continuous illumination of 100 ± 20 μeinstein m^{-2} s^{-1}, at 24 ± 2 °C, using a modified Bold Basal Medium (Sigma–Aldrich, Saint Louis, MO) diluted twice with deionized water. In most cases, freshly produced biomass was immediately used for filtration assays. To assess the impact of biomass ageing on filtration, it was also stored for 1–15 days. Three storage modes were tested, corresponding to the fate of biomass in the context of the decontamination process. In storage conditions I (SCI), the freshly produced biomass was stored under light (100 μeinstein m^{-2} s^{-1}) with air stirring and nutrient addition; in storage conditions II (SCII), it was stored under light (100 μeinstein m^{-2} s^{-1}), with stirring but without any nutrient addition; in storage conditions III (SCIII), it was stored away from light (15 μeinstein m^{-2} s^{-1}), without stirring and with no nutrient additions.

2.1.2. Characterization of algal suspensions

Algal biomass consists of an aqueous phase and suspended particles, including algal cells. These two phases were characterized by conventional analytic tools set out below.

2.1.2.1. Particles characterization. Algal density was measured using a Mallassez counting cell and correlated to dry weight (DW). The size of algal cells was determined by microscopic observation. Biomass also contains some cell debris and potentially some bacteria depending on storage conditions. Debris was not quantified but bacterial population was estimated by counting.

2.1.2.2. Aqueous phase characterization. The aqueous phase contained nutrients, dissolved organic matter, and cell content released by cell damage. Dissolved organic matter comes from extracellular polymeric substances (EPS) produced by algae, especially when cells grow under stressful conditions [16]. They are mainly carbohydrates and proteins [17]. Free EPS were separated from algal cells by centrifugation at 2000*g* for 5 min as described in [15]. Protein and polysaccharides content was characterized by colorimetric methods. Extracellular proteins were quantified using Bradford method (Kit UP36858A, Interchim, Monluçon, France); the detection limit for this method is 1 μg mL^{-1}. The phenol–sulfuric acid method from [18] was employed for polysaccharides measurement. The average molecular weight of polysaccharides was determined using size-exclusion chromatography; the size distribution was not identified.

2.2. Filtration unit

2.2.1. Experimental device

The dead-end filtration apparatus employed for filtration experiments in both membrane selection and assessment of the impact of the suspension nature and hydrodynamic actions consisted of a laboratory pressurized and stirred rig (Spectrum Laboratories Inc, Rancho Dominguez, CA). The rig was composed of a 0.4 L filtration cell providing a surface of 38.4 cm^2. Trials were conducted at constant temperature (24 ± 2 °C), in a batch mode under constant pressure (20 kPa). Compressed air was supplied by a valve placed