



# Biomass assisted microfiltration of chromium(VI) using Baker's yeast by ceramic membrane prepared from low cost raw materials

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## ABSTRACT

This work deals with the preparation of ceramic microfiltration membrane from inexpensive raw materials such as kaolin, quartz, calcium carbonate by uniaxial dry compaction method. The prepared green membrane was initially dried at 100 °C for 24 h, 200 °C for 24 h and finally sintered at 900 °C for 6 h. The properties of the membrane such as porosity, flexural strength, chemical stability and hydraulic permeability were investigated. The fabricated membrane possessed an average pore diameter of 1.32 μm, porosity of 30% and flexural strength of 34 MPa. Furthermore, the chemical stability of the membrane was found to be excellent. Eventually, the separation performance of the membrane in terms of flux and removal of chromium(VI) ion using baker's yeast biomass as a function of applied pressure, pH, metal ion concentration and biomass dosage was also studied. The removal of Cr(VI) was found to be strongly dependent on the initial pH of the solution. At lower pH, the metal solution shows higher removal due to higher binding of the metal ion with biomass. It was also observed that the removal of Cr(VI) ion increases with increasing the biomass concentration and decreases with increasing the metal ion concentration. The removal of Cr(VI) was found to be independent of the applied pressure. The maximum removal of Cr(VI) was found to be 94% with the permeate flux of  $2.07 \times 10^{-5}$  (m<sup>3</sup>/m<sup>2</sup> s) for a metal solution concentration of 100 mg/L.

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## 1. Introduction

Ceramic membranes received significant interest due to their potential application in several industrial processes. For process application, ceramic membranes offer many advantages such as excellent chemical, thermal, mechanical stability and higher separation efficiency [1–5]. The development of newer ceramic materials adopting newer fabrication techniques is currently the subject of numerous studies. Commercial membranes available in the market are generally manufactured from alumina, which are relatively expensive [6]. In order to reduce the cost of the membrane, nowadays, low cost clays are utilized for the fabrication of membrane. These include raw clay, Moroccan clay, Tunisian clay, sepiolite clay, Algerian clay, dolomite and kaolin [7–12]. Kaolin is one of the cheapest membrane raw materials easily available in India. Many researchers have reported the use of kaolin as a starting material with other additives for membrane applications [13, 14]. Recently, Neelakandan *et al.* have used the low cost clays (kaolin, feldspar, ballclay, quartz, pyrophyllite and calcium carbonate) for the preparation of ultrafiltration (UF) membranes [15].

The presence of heavy metals such as chromium, copper, lead, nickel etc., in wastewater is hazardous to the environment and human health. Chromium metals are known to be very toxic to living organisms, causing allergies, eczema, irritations and respiratory track disorders [16, 17]. Generally, chromium element exists in nature mainly in two oxidation states, such as Cr(III) and Cr(VI). Between the two forms, Cr(VI) is the most toxic. Large quantities of chromium containing wastewater generated from various processes industries particularly, leather tanning industry, pigment producing plants and metallurgy need to be treated before discharge into the natural water body. The World Health Organization recommended that the maximum allowable concentration in drinking water for total chromium should be 0.05 mg/L. The discharge of Cr(VI) to surface water is regulated to below 0.05 mg/L by USEPA and European Union, while total Cr, Cr(III), Cr(VI) and other forms are usually regulated to below 2 mg/L. Conventional treatment technologies for the removal of heavy metals and chromium ions from waste streams including coagulation, adsorption, electrochemical treatment and ion exchange are often uneconomical and operational feasibility poses problems. Moreover, the final metal recovery requires additional treatments, which complicate the process. Membrane filtration has been applied in several studies for the removal of chromium ions, usually in integrated processes. Pagana *et al.* [18] employed an innovative process, combining adsorption-permeation membrane process for the removal of chromium ions and achieved 99% removal efficiency. In another

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work, they have proposed a novel scheme of combined adsorption-ultrafiltration (AUF) process for the removal of arsenic and chromium from water using nano-sized  $\text{Fe}_2\text{O}_3$  adsorbent and  $\gamma\text{-Al}_2\text{O}_3$  ultrafiltration membranes (average pore diameter of  $\sim 3\text{--}4$  nm) [19]. Irena et al. [20] used a polyelectrolyte-assisted UF system to study the removal of chromium ions from aqueous solution and observed very high Cr(VI) removal efficiency at a pH of 6. Aroua et al. [21] employed a polymer-enhanced UF system to remove Cr(III) and Cr(VI) from aqueous solution using different types of water soluble polymer and achieved complete removal of metal ions. Also, it is reported that the use of biosorbent or biomass for the removal of heavy metals is cost effective and it can be obtained from various fermentation industries [22]. The use of non-living biomass has many advantages in comparison to the use of living biomass. Some of these are greater binding capacities with toxic metals and ease of availability from numerous fermentation industries. It is well known that the yeast cells can uptake heavy metals from aqueous solution in wide external conditions and can be successfully applied for the removal of heavy metals from wastewater [23, 24]. Moreover, the influences of physical and chemical properties of metals and environmental condition (pH, temperature) on metal uptake by yeast cells are well described [25]. Brady et al. [26] investigated the removal of heavy metals bound with yeast cells using hollow fiber cross-flow microfiltration [26]. Recently, Bayhan et al. [27] reported the removal of divalent heavy metal mixtures ( $\text{Ni}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ ) using yeast (*saccharomyces cerevisiae*) by membrane filtration (anisotropic cellulose acetate membrane with pore size of  $0.2\ \mu\text{m}$ ). They achieved higher rejection and flux for Pb as compared to other metals (Cu and Ni) [27].

The above extensive literature review clearly indicates that UF membranes (whose pore size is about  $0.2\ \mu\text{m}$ ) are mostly used for the removal of metal ions. On the other hand, the yeast cells have relatively narrow size distribution ( $2\text{--}5\ \mu\text{m}$ ). Therefore, the usage of membranes with larger pore sizes would be further useful for the simultaneous enhancement of flux and minimization of irreversible fouling. In summary, the objective of this work is to examine the separation potential of a low cost microfiltration membrane for the removal of Cr(VI) assisted with baker's yeast biomass.

This work addresses the fabrication of a low cost ceramic microfiltration (MF) membrane using inexpensive raw materials. Eventually, adsorption-MF principle is investigated as a novel separation scheme for the removal of Cr(VI) from its aqueous solution. The structural characteristics, mechanical and chemical stability of the membrane were investigated. Water flux and hydraulic permeability were also evaluated in order to determine the membrane performance. Separation performance of Cr(VI) bound with biomass was carried out and the effect of process parameters such as pH, metal ion concentration and biomass dosage was also investigated.

## 2. Experimental

### 2.1. Membrane synthesis

The raw materials used for the synthesis of membrane are kaolin (50 g), quartz (25 g) and calcium carbonate (25 g). These raw materials were mixed in a ball mill at 40 rpm for 20 min and the resulting powder was then sieved using 30 mesh standard screen. A requisite amount was pressed at a pressure of 50 MPa with the help of stainless steel mold to yield a disc shaped membrane. The prepared green membrane (62 mm diameter and 5 mm thickness) was first dried at  $100\ ^\circ\text{C}$  for 24 h and  $200\ ^\circ\text{C}$  for 24 h for the complete removal of moisture. Consequently, the membrane was sintered at  $900\ ^\circ\text{C}$  for 6 h with a heating rate of  $2\ ^\circ\text{C}/\text{min}$  in a muffle furnace. The sintered membrane was polished at both sides using silicon carbide abrasive paper (No.C-220) to obtain a flat surface. The polished membrane was cleaned with Millipore water in an ultrasonic bath [make: Elma

(India); model: T460] for 15 min to remove the loose particles created during polishing.

### 2.2. Membrane characterization

Porosity of the prepared membrane was evaluated by Archimedes' principle, in which the membrane was first dried in hot air oven at  $110\ ^\circ\text{C}$  for 6 h to remove all moisture present in the membrane and its dry weight ( $W_1$ ) was measured. It was then immersed in water for 24 h at room temperature. After that, the membrane was taken out and water on the outer surface was removed using tissue paper and the wet weight of the membrane ( $W_2$ ) was measured. The porosity of the membrane was determined using the following relation [28]

$$\varepsilon = \frac{W_2 - W_1}{W_1} \quad (1)$$

The chemical stability of the ceramic membrane was estimated by soaking the membrane into acid and alkali solution individually using HCl (pH 1) and NaOH (pH 14) for one week at atmospheric condition. The stability was estimated in terms of mass loss before and after corrosion. The flexural strength of the membrane was tested in a three point bending method on  $55\ \text{mm} \times 5\ \text{mm} \times 5\ \text{mm}$  rectangular bars using Universal Testing Machine (DUTT-101, M/s Deepak Polyplast, Mumbai, India).

A dead-end filtration setup was used for the determination of water flux and hydraulic permeability [29]. Prior to the experiment, the pure water was passed through the membrane at a high pressure than the operating pressure to remove any loose particles present in the pores. Eventually, the water flux was measured at various applied pressures ( $70\text{--}345$  kPa). At each applied pressure, the collection of first 50 mL of the permeate was discarded and time taken for collection of second 50 mL of the permeate was measured for flux determination. The reported values are average of three readings and the data is regressed by a linear curve to obtain the average hydraulic permeability.

### 2.3. Preparation of yeast biomass and feed solution

The baker's yeast (*saccharomyces cerevisiae*) was procured from SAF yeast Co. Ltd., Chembur, Mumbai, India. The yeast cells were washed three times with Millipore water, centrifuged and dried at  $80\ ^\circ\text{C}$  for 24 h in a hot air oven to deactivate the cells [30]. The stock solution of chromium(VI) was prepared by taking potassium dichromate (Analytical grade, Merck India Ltd) and dissolving it in appropriate amount of Millipore water. A calculated amount of the dried yeast biomass was added to 100 mL of Cr(VI) solution in 250 mL Erlenmeyer flasks. Before adding biomass, the pH of the solution was adjusted by the addition of 1 N  $\text{HNO}_3$  or 1 N NaOH. The suspension was then shaken at a constant temperature ( $25\ ^\circ\text{C}$ ) in an incubator shaker at 200 rpm for 24 h, which was sufficiently long for adsorption-equilibrium. After shaking, the solution was subjected to the microfiltration experiments.

### 2.4. Microfiltration of chromium(VI)

Filtration experiment was performed in a dead-end filtration setup in batch mode operation [29]. All experiments were conducted at room temperature ( $25\ ^\circ\text{C}$ ). In all experiments, the feed volume of 100 mL was filled into the filtration setup. The first 10 mL of the permeate was discarded and time taken for collection of second 10 mL of the permeate was measured for the permeate flux calculation. The percent removal (R) was evaluated according to the following expression

$$R(\%) = \frac{C_f - C_p}{C_f} \times 100 \quad (2)$$

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