



# Modeling of arsenic, chromium and cadmium removal by nanofiltration process using genetic programming

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## ABSTRACT

In this paper, genetic programming (GP) as a novel approach for the explicit formulation of nanofiltration (NF) process performance is presented. The objective of this study is to develop robust models based on experimental data for prediction the membrane rejection of arsenic, chromium and cadmium ions in a NF pilot-scale system using GP. Feed concentration and transmembrane pressure were considered as input parameters of the models. The ions rejection is considered as output parameter of the models. Some statistical parameters were considered and calculated in order to investigate the reliability of each model. The results showed quite satisfactory accuracies of the proposed models based on GP. The results also nominated GP as a potential tool for identifying the behavior of a NF system.

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## 1. Introduction

One of the most serious environmental contaminants which recognized as threaten to human lives are heavy metals. Their higher toxicity, accumulation and retention in human bodies are the main problems that they make [1]. Arsenic, Chromium and Cadmium have been made wide attractive attention from environmentalist as one of the main toxic pollutants in drinking water.

Obviously, removal of these pollutants are kind of interest. Nanofiltration (NF) is a pressure-driven process which its characteristics fall among ultrafiltration (UF) and reverse-osmosis (RO) ones [2]. In fact, in comparison with UF and RO, NF allows obtaining higher fluxes than reverse osmosis and better rejections than ultrafiltration [3–6]. Because of its benefits, such as low energy consumption, high permeation flux, and unique separation capability for different valence ions, nanofiltration has been widely used in different applications, for instance, heavy metals removal, water softening, color removal and reduction of chemical oxygen demand (COD) and biological oxygen demand (BOD) [1,2,6–10]. However, for a successful implementation of the process it is necessary to obtain some information about the performance of a given membrane. On this basis, modeling plays an important role in description of a NF process and provides useful information about it.

Several efforts have been made to develop models that have a reasonably good description of a NF process. Many NF models based on extended Nernst–Planck equation have been presented

so far. One of these models, which widely have been used, is Donnan-steric partitioning pore model (DSPM) [11–13]. This model describes the transport of ions in terms of porosity ratio, an effective membrane thickness and charge density. DSPM has undergone several modifications by taking into account the hindrance effect of the ions through the pores in the membranes [14], concentration polarization [15], and dielectric constant [16]. Some other models based on extended Nernst–Planck equation have been developed for description of a NF process in separation of heavy metals. Gomes et al. [6] analyzed the nanofiltration process for separating Cr (III) from acid solutions and the results were interpreted with a suitable mathematical model that includes the polarization effect and extended Nernst–Planck equation. Garba et al. [1] proposed a model in combination of the extended Nernst–Planck equation with the film theory. The proposed model was applied to predict cadmium salts rejection through a nanofiltration membrane and reasonably good results were obtained.

Kedem and Katchalsky [17] proposed a model which describes the transport of the solute through ultrafiltration, nanofiltration, and reverse osmosis membranes by irreversible thermodynamics. This model, which have used by many investigators [17–23], treats NF membranes as a gray box and characterizes them in terms of salt permeability and reflection coefficient.

The above mentioned mathematical models were derived from physical descriptions and understanding of NF process. These models usually are mathematically complex, computationally expensive and require detailed knowledge of the filtration process [22,24]. Therefore, alternative methods for description of a NF process by using available process data and extending it to a mathematical model, which can be applied simply on unavailable data,

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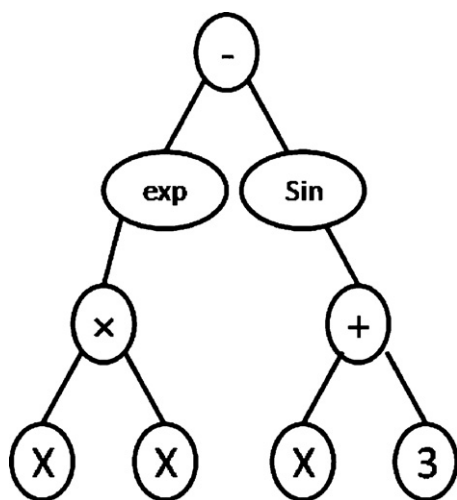


Fig. 1. Tree representation of  $\exp(X^2) - \sin(X+3)$ .

are so worthwhile. Genetic algorithm (GA) and especially genetic programming (GP), which is one of the GA's branches, is such tool that can be used in wide range of problems. Some few remarkable works in the field of using GA in membrane technology have been done. Majority of these studies concentrated on using GA as an optimizing tool to develop better performing membranes [25–28]. Lee et al. [29] used GP for modeling and prediction of the microfiltration (MF) membrane fouling rate in a pilot-scale drinking water production system. Their results were satisfactory. Thus, to the best of our knowledge, there is no work in the field of modeling of a NF process by means of GP.

In this study, modeling of As (V), Cr (VI) and Cd (II) removal from wastewater in terms of rejection as function of transmembrane pressure (TMP) and initial concentration of pollutants were done using GP. The goal is to exploit GP flexible tree structure for building a prediction model for removing each of above mentioned ions by using NF process. In the literature, none reported work was found to use this approach in order to assess the separation of heavy metals by NF process.

## 2. Genetic programming

GP is a systematic method for getting computers to solve a given problem by automatically generating algorithms and expressions on the basis of natural evolution. These expressions are coded as a tree structure with its nodes (functions) and terminals (leaves) [30]. The function set chosen is  $\{+, -, \times, /, \sin, \cos, \exp, \log, \text{abs}\}$ . Moreover, the function set may also include other mathematical functions, conditional operators or any user-defined operators [31]. The terminal set chosen contains the independent variable  $X$  or an integer. For example, Fig. 1 shows the representation of the function  $\exp(X^2) - \sin(X+3)$ . To “read” trees in this fashion, one resolves the sub-trees in a bottom-up fashion. The GP model is explicit and free from conceptual designs, thus it has few requirements about the domain knowledge of the problem and is therefore less problem-dependent [32,33]. Fig. 2 shows a brief flowchart of GP. One loaded the problem data, initial population of programs created randomly and fitness value, which quantifying how well the program solves the problem, for each of them evaluates. New generations of programs are iteratively created by selecting parents based on their fitness and breeding them via variety of genetic operators including crossover, mutation and reproduction. In crossover operation, two parents are selected and two offspring generated from them; the offspring's are produced by swapping random sub-trees of the

parents. The mutation operator generates one offspring from one parent by substituting a randomly selected sub-tree with a newly generated sub-tree. If this replacement violates the depth limit, mutation just reproduces the original tree into the new generation. The reproduction operator simply selects an individual and copies it to next generation.

Because better individuals are selected more often and given the chance to pass their best characteristics to their offspring, the population tends to improve in quality along successive generations. This evolutionary process continues until a given stop condition, e.g. acceptable fitness or maximum number of generation, is verified. Finally, the best-so-far individual is designated as the result and is produced as the output of the GP operation.

## 3. Experimental

### 3.1. Membrane and chemicals

The nanofiltration membrane used in this study, was a flat sheet one that had an aromatic polyamide layer with  $63.6 \text{ cm}^2$  effective filtration area, denoted as UTC-70UB and manufactured by Toray Japanese Company.

All salt solutions were prepared fresh using reagent grade chemicals dissolved in pure water produced by a New Human Power I unit (Humancorp). Arsenate As(V), Chromium Cr(VI) and Cadmium Cd(II) solutions were prepared from  $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ,  $\text{K}_2\text{Cr}_2\text{O}_7$  and  $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ , respectively. All salts were procured from Merck.

### 3.2. Experimental setup

The nanofiltration experiments were carried out on a crossflow filtration setup as shown schematically in Fig. 3. This apparatus consists on a NF disk module, a feed tank, a washing solution tank, a high pressure pump, pressure gauges installed on the feed and retentate pipes connected to the module, a thermometer, a heat exchanger, a rotameter installed on permeate outlet as flow indicator and some valves. Additionally, a bypass line was considered for pump exit stream. The friction between fluid and tubes and also dissipation of energy in pump result in increasing temperature during the tests, thus for controlling the temperature of process, a heat exchanger was used.

### 3.3. Procedure

The experiments were done after a through washing and rinsing of the membrane. The experiments were carried out in batch mode which both permeate and retentate were returned to feed reservoir to keep a constant concentration. The samples were taken after flow conditions became steady-state for each of runs. The stabilization time for each run was about 30 min. Maximum volume of permeate, which was obtained in all the tests, was about 50 ml. The affect of this volume on variation of the retentate concentration was neglected. After doing filtration process in each feed concentration level, all parts of apparatus were washed and rinsed with pure water. Washing of the membrane was performed at 5 bar at least for 10 min to insure that the initial membrane pure-water permeability was restored. By this procedure, no fouling was obtained in any of experiments.

The experiments were conducted at different TMPs in range of 5–14 bar and feed concentrations in range of 100–400  $\mu\text{g/l}$  for arsenic and chromium and 20–80  $\mu\text{g/l}$  for cadmium. The feed reservoir temperature was maintained approximately at  $30^\circ\text{C}$ . The more details have been introduced in the previous paper [10].

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