



Comparative assessment on lead removal using micellar-enhanced ultrafiltration (MEUF) based on a type-2 fuzzy logic and response surface methodology

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ABSTRACT

Treatment of wastewater containing heavy metal ions is a growing challenge due to their coexistence in many industrial effluents. In the present investigation, an interval type-2 fuzzy logic controller (IT2FLC) and Response surface methodology (RSM) based model was developed to predict and modelling of lead removal and permeation flux in separation processes using micellar-enhanced ultrafiltration (MEUF). The Box-Behnken experimental design method investigates the effect of parameters such as surfactant concentration, solution pH and surfactant to metal concentration ratio (S/M) on MEUF performance. The simulation results reveal that the output variables of MEUF performance could be predicted with a high degree of accuracy. Predicting the performance of a MEUF separation process, prior to its ultrafiltration experiment, was introduced as the most prominent feature of this study. This can offer an intelligent method for evaluation of MEUF presentation instead of conventional method, which is both time saving and cost effective.

1. Introduction

The discharge of wastewater containing heavy metal ions and dissolved organic pollutants from various chemical and petrochemical industries (coal refining, electroplating, batteries manufacturing, metal finishing, textiles, and paints etc.) into the environment has been a serious issue since they are non-biodegradable, highly toxic and carcinogenic in nature [32]. This has led to increasing concern due to their harmful impact on human health and aquatic life. Thus, the treatment of polluted aqueous effluents for simultaneous removal of both the hazardous contaminants is a very important environmental challenge for the researchers [50]. Lead is classified as a hazardous waste and is highly toxic to humans, plants and animals. When animals and plants are exposed to it, it causes death of these species, whereas in humans it causes anaemia, brain damage, mental deficiency, anorexia, vomiting and malaise [42,46]. Among heavy metal exposures, lead exposure is one of the most common exposures that can lead to significant neuropsychological and functional reject in humans [33]. In particular, lead (II) is considered as highly toxic heavy metal, which is commonly used in a number of industrial applications such as storage battery manufacture, television tube, printing pigments, fuels, photographic material and explosives manufacturing. Lead bearing wastewater

resulted from all these industries must be disposed of after treatment [44]. There are various methods applied for the removal of heavy metals from the industrial effluents. Some conventional methods of heavy metal-containing wastewater treatment include precipitation, electro-deposition, evaporation, ion exchange, and crystallization [42,15]. However, industries are looking for competing alternative technologies which may overcome some of the inherent disadvantages of these methods [31]. Membrane separation processes of different types of membranes show great promises for commercial application [37,3,49]. However, both RO (Reverse Osmosis) and NF (Nano-filtration) processes use relatively "dense" membranes. Permeability of these membranes is low and thus, to get the desired throughput, a high operating pressure is required [2]. MEUF process is suitable for separation of heavy metal-ions having low concentration in wastewater [15]. Here we choose Pb^{2+} as heavy metal ions for its removal because it is generally available in low concentration in wastewater and also it has comparatively higher molar mass [15]. Therefore, it is quite easy to form precipitation with surfactant at particular pH.

Micellar-enhanced ultrafiltration is one of the promising methods for the separation of such heavy metals contaminated effluents [44] which has been used for the removal of various organic and/or inorganic pollutants from aqueous phase [42] and also several kinds of

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Nomenclature

MEUF	Micellar-Enhanced Ultra-Filtration
LCA	Life Cycle Assessment
S/M	Surfactant to metal ratio
FIS	Fuzzy Inference System
IT2FL	Interval type-2 fuzzy logic
T2FLC	Type-2fuzzy logic controller
MFIS	Mamdani Fuzzy Inference System
ANFIS	Adaptive Neuro-Fuzzy Inference System
R^2	Coefficient of determination
MAE	Mean Absolute Error

MAPE	Mean Absolute Percentage Error
CMC	Critical Micelle Concentration
SDS	Sodium Dodecyl-Sulphate
MF	Membership Function
RSM	Response Surface Methodology
T2FL	Type-2 fuzzylogic
T2FS	Type-2 fuzzy system
BBD	Box-Behnken Design
RMSE	Root Mean Square Error
ANOVA	Analysis of Variance
T2FLS	Type-2 Fuzzy Logic System
ANN	Artificial Neural Network

metal ions (Cd^{2+} , Zn^{2+} , Cu^{2+}) can be treated by MEUF. Micellar-enhanced ultrafiltration is a separation method which combines high selectivity of reverse-osmosis and high flux of ultrafiltration for modification of this process. The combination of surfactants and membranes in separation of dilute polluted water has led to the development of novel techniques or enhancement of existing techniques. In MEUF, micelles of ionic surfactant are introduced into aqueous solution to bind ions on the surface of the opposite-charged micelles via electrostatic interactions [41]. Then they can be retained by an ultrafiltration membrane with pore sizes smaller than micelle sizes [12]. As a result, the metal ions associated with micelles are removed effectively. The major advantages of applying of this MEUF process are: (i) Low energy requirement, (ii) Low pressure driven, (iii) High removal efficiency, (iv) Required small space for its high packing density and also (v) It has the potential to remove and recycle heavy metals from wastewater [12]. Most of the MEUF studies are performed using synthetic surfactant for the removal of heavy metals or organic compounds from wastewater [10,52,24,48,50]. Since synthetic surfactant has relatively high CMC, large quantities of surfactant must be used for an effective separation. This results to an increase in concentration polarization and membrane fouling leading to decrease in process throughput [50].

Conventionally, the study of MEUF has been conducted using the one variable at a time approach, i.e., a single factor is varied while all other factors are kept unchanged for a particular set of experiments. Such conventional or classical methods of experimentation usually lead to a low efficiency in optimization issues because it involves many experimental runs, which are time consuming, ignore interactions effects between the considered parameters of the process [13,18]. The application of statistical experimental design for membrane systems seems to be the best methodology for process control and optimization. The present study reports data of a metal-polluted wastewater model in order to evaluate the efficiency of MEUF for the removal of Pb^{2+} from aqueous solutions. The factors that have been considered are operative pressure difference, membrane molecular weight cut-off, molar ratio of surfactant to the solute, kind of surfactant and additives, pH, ionic strength, etc.

In this investigation, MEUF process using Box-Behnken as an experimental design was performed and the ability of FIS was evaluated to modelling and simulation. In this study, Box-Behnken designs (BBD) were applied as experimental design strategies to investigate MEUF process performance c.f Huang et al. [16]. Basically in statistics, Box-Behnken designs are experimental designs for response surface methodology (RSM) devised by George E. P. Box and Donald Behnken. Ferreir et al. (2007) described the fundamentals, advantages and limitations of the Box-Behnken design (BBD) for different optimization of analytical methods. They established also a assessment between this design and composite central, three-level full factorial and Doehlert designs. A lucid study on factors and responses concerned during the optimization of analytical systems was presented. One of the main advantages of Box-Behnken design matrix is that it does not contain combinations for which all factors are simultaneously at their highest or

lowest levels. So this design is useful to avoid experiments performed under extreme conditions [26]. Box-Behnken designs include all spherical designs and require factors to be run at only three levels which are main advantages of this particular design [53]. The main factors behind the application of this design are:

- Each independent variable is placed at one of three equally spaced values which are generally coded as -1 , 0 and $+1$.
- The design should be sufficient to fit a quadratic model i.e. one containing squared terms, products of two factors, linear terms and an intercept.
- The ratio of the numbers of experimental points to the number of coefficients in the quadratic model should be reasonable (1.5 to 2.6).

Response surface methodology (RSM) is a collection of mathematical and statistical techniques that can be used for studying the effects of several factors at different level and their influence on each other. The main objective of using RSM is to optimize the response based on the factors investigated [36,38]. The RSM study begins with a definition of a problem concerning which response is to be measured, how it is to be measured, which variables are to be explored and the experiment plan is then designed. Major benefits of this response methodology are: (i) Reduction of number of experiments needed rather than a full experimental design at the same level, (ii) It allows the effects of a factor to be estimated at several levels of other factors, (iii) It helps to obtain the surface contour that provides a good way for envisioning the parameter interaction [9]. The Design Expert Version 6.0 software was used to develop the experimental plan for RSM. This software is also used to analyze the data collected by performing analysis of variance (ANOVA). If the model looks good, then the three-dimensional graphs and contour plots would be plotted for interpretation. In brief, a good model must be significant and the lack-of-fit must be insignificant. The various coefficient of determination, R^2 -values should be close to 1.

Fuzzy inference system is one of the most efficient computational methods rather than other analytical and statistical techniques. Since lead removal system and technologies are quite intricate and uncertain, they can widely be applied for modelling of different components in this sector, because they can study new patterns which were not previously available in the trained data sets and they can also apprise knowledge over time as long as more training data sets are provided [27]. In the context of environmental management, the use of the fuzzy logic method is strongly suggested. Khoshnevisan et al. [27] estimated the yield of greenhouse strawberry with the help of adaptive neuro-fuzzy inference system (ANFIS). Ana et al. [5] suggested an integrated recycling approach for GFRP pultrusion wastes using fuzzy logic. Sami et al. [45] had done a case study in cane farms in Iran using fuzzy logic. Khoshnevisan et al. [29] predicted the potato yield based on energy inputs by means of ANFIS. Alavi et al. [4] had determined the quality of Mozafati dates using Mamdani fuzzy inference system. Hosoz et al. [14] predicted the performance of a refrigeration system with the help of ANFIS. A technique introduced by Wua et al. [51] for the measurement

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