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Original Article

# Fuzzy optimization for the removal of uranium from mine water using batch electrocoagulation: A case study

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This research presents a case study on the remediation of a radioactive waste (uranium: U) utilizing a multi-objective fuzzy optimization in an electrocoagulation process for the iron-stainless steel and aluminum-stainless steel anode/cathode systems. The incorporation of the cumulative uncertainty of result, operational cost and energy consumption are essential key elements in determining the feasibility of the developed model equations in satisfying specific maximum contaminant level (MCL) required by stringent environmental regulations worldwide. Pareto-optimal solutions showed that the iron system (0 μg/L U: 492 USD/g-U) outperformed the aluminum system (96 μg/L U: 747 USD/g-U) in terms of the retained uranium concentration and energy consumption. Thus, the iron system was further carried out in a multi-objective analysis due to its feasibility in satisfying various uranium standard regulatory limits. Based on the 30 μg/L MCL, the decision-making process via fuzzy logic showed an overall satisfaction of 6.1% at a treatment time and current density of 101.6 min and 59.9 $mA/cm^2$, respectively. The fuzzy optimal solution reveals the following: uranium concentration – 5 μg/L, cumulative uncertainty – 25 μg/L, energy consumption – 461.7 kWh/g-U and operational cost based on electricity cost in the United States – 60.0 USD/g-U, South Korea – 55.4 USD/g-U and Finland – 78.5 USD/g-U.


## 1. Introduction

In recent times, nuclear fuel has proven to be a viable alternative source of energy due to its high power generation, environmentally-benign nature, high efficiency, sustainability and cost-effectiveness [1]. Due to the increasing demand in energy worldwide, the continuous development of nuclear technology has generated a significant amount of radioactive wastes [2]. In the past years, uranium has been considered as an essential energy component in the nuclear technique [3]. Uranium is a naturally-occurring radionuclide characterized by its high biological and chemical toxicity, radioactivity, non-degradability and long half-life [4]. In the environment, uranium is naturally found in various chemical forms (+2, +3, +4, +5, +6) where U(VI) is the most common valence state of uranium found in water under oxidizing conditions [5]. Anthropogenic activities that release uranium include coal combustion, utilization of phosphate fertilizers, ore mining, nuclear fuel combustion and manufacture of weapons [6]. Uranium has no essential biological function in living organisms wherein radionuclide exposure has been determined to cause serious diseases including liver and kidney damage, muscle cramp, and lung insufficiency [5]. The US Environmental Protection Agency has classified uranium to be a human carcinogen belonging to Group A. The World Health Organization and US Environmental Protection Agency have set the maximum contaminant level in drinking water at 30 μg/L [1].

Numerous physico-chemical technologies have been applied in the separation and removal of uranium from wastewater that include in situ bioremediation [7], adsorption [2], emulsion liquid membrane [8], nanofiltration [9] and electrocoagulation [10]. Electrocoagulation is a process where coagulants are generated in situ via dissolution of the metal sacrificial electrodes [11]. The method involves application of a direct electric current of low-voltage, which produces a high ionic charge that destabilizes the target contaminants present in the aqueous environment [12]. Electrocoagulation has several attractive features including low dissolved solids, capacity to accurately control the rapid generation

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of coagulants, low volume of sludge production and non-requirement of supplementary chemicals and microorganisms [13]. Moreover, electrochemical methods are known to be highly selective, simple to operate and flexible that utilize compact equipment systems of high energy efficiency [14].

Previous studies have reported on the use of electrocoagulation in the treatment of numerous types of wastewater generated by textile processing [15], paper and pulp industry [16], polymer production [17], alcohol distillery [18], mine water [10] and chemical polishing [19]. The previous work of Nariyan et al. [10] investigated the performance of electrocoagulation in removing uranium from mine water. Parameters such as reaction time, current density and electrode type were optimized using response surface methodology (RSM) via the central composite design. At a current density of 70 mA/cm$^2$ and reaction time of 120 min, the optimum removal efficiency of 99.7% and 97.7% was attained using an iron-stainless steel electrode and aluminum-stainless steel electrode, respectively.

RSM, which is comprised of practical mathematical techniques, is applied in the improvement, enhancement and optimization of complex systems [20]. RSM is employed in the assessment of the effect of different independent parameters and its interactions on a system response, which is typically disregarded in a one-factor design [21]. It is a multivariate statistical design method that determines the relation between parameters and response within the total variable scope [22]. In addition, the operating parameters and regression model are derived from the quantitative data provided by the designed experiments [23]. The popularity of RSM is due to its numerous benefits that includes requirement of lesser experimental runs, generation of surface contours for analyzing the parameters' interactions and utilization of very minimal resources and time [24]. However, RSM involves an uni-model objective function, which only explores local optimization. It also does not account for the operating cost and cumulative error of uncertainty that are significant in practical applications for the scale-up and design of a pilot system [25].

Fuzzy logic model is a technique that develops indices that would express human experiences and thoughts derived from subjective, ambiguous information [26]. In systems engineering, numerical function modelling is obtained using fuzzy logic via arithmetical calculations acquired from fuzzy rules [27]. The fuzzy logic system has gained attention due to its adaptability to gain control over non-linear methods, development of control structures with multiple input and output and simple controller standardization [26]. In addition, uncertainties are adequately handled by fuzzy logic where the input/output correlation is inferred using relatively simple mathematical equations in linguistics terms such as "if-then" rules [28]. Several applications of the fuzzy logic system include environmental management [29], wastewater treatment systems [30], water quality modelling [31] and air pollution [32]. However, there are no published studies on the application of fuzzy logic in evaluating the feasibility of electrocoagulation in uranium removal from mine water. The relationship between uranium removal efficiency and operating cost using electrocoagulation has not yet been reported, which is significant in the evaluation of the viability of electrocoagulation for environmental and practical application.

In the present work, fuzzy logic model was employed in multi-objective optimization to assess the most satisfactory condition that would provide the lowest uranium concentration retained at a minimum cost and energy. The effect of operating conditions such as type of electrode (aluminum-stainless steel electrode, iron-stainless steel electrode) and treatment time in relation to the operating costs and retained uranium concentration was investigated. The energy consumption was determined as basis of the operational cost of the electrocoagulation process.

## 2. Methodology

### 2.1. Experimental method

The mine water samples were obtained from the Pyhäsalmi mine (Finland) and were preserved at −20 °C. Table 1(a) lists the detailed characteristics of the mine water. The experimental set-up was comprised of anode/cathode combination (iron-stainless steel, aluminum-stainless steel) in a 500-mL beaker where mine water was stirred at 200 rpm under a voltage of 40 V and electrical current of 5 A. The electrode dimensions are 70 × 50 mm where a distance of 5 mm between the cathode and anode was maintained throughout the entire duration of the study. The current density (10 mA/cm$^2$ to 70 mA/cm$^2$) and time (1 min–120 min) were varied where direct power supply was used to deliver the current density (GW INSTEK psp-405). The pH, redox-potential and uranium concentration were measured using pH meter (pHC101), ORP meter (intelliCAL™ MTC101) and inductively coupled plasma mass spectrometer (ICP-MS, RA3000, detection limit: less than 1 μg/L), respectively.

### 2.2. Fuzzy multi-objective optimization method

The Pareto set with fuzzy mathematical programming was employed in the optimization study to provide the most probable operating conditions of energy consumption and total uranium concentration in account of its cumulative uncertainty of results. For the process parameters (treatment time and current density) of uranium treatment in electrocoagulation, the process parameters and the electricity prices [33] are listed in Table 1(b) and (c). Moreover, Table 2 shows the data derived from the study of Nariyan et al. [10] with the additional incorporation of the cumulative uncertainty of results. The estimation of the reasoning approach was achieved using a linguistic value where the true value ranges from 0 (completely false) to 1 (completely true) [34]. As shown in Fig. S1, the fuzzy system algorithm of the present work was illustrated in a flowchart. Optimization studies were carried out via Lingo 18.0 (Lindo Systems, Chicago, IL, USA). In non-linear programming, a global optimizer was used by the application software. All the symbols used in this study are described in Table S1. On the other hand, the specific equations utilized in this research are listed in Table 3.

#### 2.2.1. Determination of boundary limits through the Pareto optimality

The usage of the Pareto analysis has the advantage in the early stage of the decision analysis. When there are numerous competing practical solutions, decision-making strategies employ the Pareto analysis in picking a definite amount of courses that would bring about substantial overall effect [35]. The application of the Pareto set was attributed to its capability in defining the initial Pareto optimal solution under various objective functions [36]. Initially, the precise values of boundary limits was obtained using the Pareto method for the energy consumption and retained uranium concentration related to its operational cost. The multi-objective fuzzy optimization applies the boundary limits. Eq. (1) shows the first objective function in which $E$ is minimized to determine the lower boundary limit of the energy consumption and the upper boundary limit of the overall uranium concentration retained after the electrocoagulation process. On the other hand, the second objective function in Eq. (2) is minimized in order to determine the lower boundary limit of the uranium concentration that accounts its cumulative uncertainty of the response in the treatment process and

**Table 1**
(a) Mine water characteristics, (b) factors affecting electrocoagulation and (c) the operational cost in terms of electrical energy consumption.

| (a) Mine water [10] | | (b) Process parameters | | (c) Energy consumption cost [33] | |
|---|---|---|---|---|---|
| Characteristics | | Variables | Range | Countries | Electricity prices (USD/kWh) |
| Uranium (μg/L) | 620 | Treatment time ($X_1$: min) | 0–120 | United States | 0.13 |
| pH | 2.68 | Current density ($X_2$: mA/cm$^2$) | 10–70 | South Korea | 0.12 |
| Color | Yellow-brownish | | | Finland | 0.17 |
| Redox (mV) | 467 | | | | |

**Table 2**
CCF design matrix for uranium removal by electrocoagulation with an iron–stainless steel and Al-stainless steel anode/cathode system.

| Run | Treatment time ($X_1$: min) | Current density ($X_2$: mA/cm$^2$) | Uranium concentration (μg/L) | | | |
|---|---|---|---|---|---|---|
| | | | Fe-stainless steel | Cumulative uncertainty[a] | Al-stainless steel | Cumulative uncertainty[a] |
| 1 | 120 | 10 | 406 | ±35 | 611 | ±30 |
| 2 | 120 | 40 | 13 | ±14 | 537 | ±64 |
| 2 | 120 | 70 | 2 | ±23 | 14 | ±10 |
| 3 | 60 | 10 | 579 | ±73 | 620 | ±23 |
| 4 | 60 | 40 | 305 | ±6 | 620 | ±10 |
| 5 | 60 | 70 | 16 | ±44 | 538 | ±44 |
| 6 | 0 | 10 | 620 | ±11 | 620 | ±47 |
| 7 | 0 | 40 | 620 | ±48 | 620 | ±6 |
| 8 | 0 | 70 | 620 | ±19 | 620 | ±27 |

[a] Calculated cumulative uncertainty of the results.

**Table 3**
List of equations.

| | | | |
|---|---|---|---|
| (1) | $\min E$ | (20) | $OC_1 = 0.13E_{Fe}$ |
| (2) | $\min Y + W_Y$ | (21) | $OC_2 = 0.12E_{Fe}$ |
| (3) | $E = \frac{V \cdot I \cdot \Delta t}{1000 \cdot v \cdot \Delta U}$ | (22) | $OC_3 = 0.17E_{Fe}$ |
| (4) | $Y = \beta_0 + \sum_{i=2}^{2} \beta_i X_i + \sum_{i=1}^{2} \sum_{j=i+1}^{2} \beta_{ij} X_i X_j + \sum_{i=2}^{2} \beta_{ii} X_i^2$ | (23) | $Y_{Fe} \geq 0$ |
| (5) | $W_Y = \left( \sum_{i=2}^{2} \frac{\partial Y}{\partial X_i} W_{X_i} \right)^{\frac{1}{2}}$ | (24) | $0 \leq X_1 \leq 120$ |
| (6) | $C_l = E(b_l)$ | (25) | $10 \leq X_2 \leq 70$ |
| (7) | $Y \geq 0$ | (26) | $Y_{Al} = 473.82 + 4.34X_1 + 9.05X_2 - 0.08X_1X_2 - 0.02X_1X_1 - 0.10X_2X_2$ |
| (8) | $X_i^L \leq X_i \leq X_i^U$ | (27) | $W_{Y_{Al}} = \sqrt{\left( \frac{\partial Y_{Al}}{\partial X_1} W_{X_1} \right)^2 + \left( \frac{\partial Y_{Al}}{\partial X_2} W_{X_2} \right)^2}$ |
| (9) | $\max \mu_O \leq \mu_k$ | (28) | $\frac{\partial Y_{Al}}{\partial X_1} = 4.34 - 0.08X_2 - 0.05X_1$ |
| (10) | $\mu_1 = \frac{E^U - E}{E^U - E^L}$ | (29) | $\frac{\partial Y_{Al}}{\partial X_2} = 9.05 - 0.08X_1 - 0.20X_2$ |
| (11) | $\mu_2 = \frac{(Y + W_Y)^U - (Y + W_Y)}{(Y + W_Y)^U - (Y + W_Y)^L}$ | (30) | $E_{Al} = \frac{140X_1X_2}{3(620 - Y_{Al})}$ |
| (12) | $(Y + W_Y) \leq MCL$ | (31) | $OC_1 = 0.13E_{Al}$ |
| (13) | $0 \leq Y \leq MCL$ | (32) | $OC_2 = 0.12E_{Al}$ |
| (14) | $\mu^L \leq \mu_O \leq \mu^U$ | (33) | $OC_3 = 0.17E_{Al}$ |
| (15) | $Y_{Fe} = 768.43 - 4.42X_1 - 7.46X_2 - 0.06X_1X_2 + 0.02X_1X_1 + 0.07X_2X_2$ | (34) | $\mu_O \leq \mu_1 \& \mu_2$ |
| (16) | $W_{Y_{Fe}} = \sqrt{\left( \frac{\partial Y_{Fe}}{\partial X_1} W_{X_1} \right)^2 + \left( \frac{\partial Y_{Fe}}{\partial X_2} W_{X_2} \right)^2}$ | (35) | $\mu_1 = \frac{491.8 - E_{Fe}}{491.8}$ |
| (17) | $\frac{\partial Y_{Fe}}{\partial X_1} = -4.42 - 0.06X_2 - 0.04X_1$ | (36) | $\mu_2 = \frac{620 - (Y_{Fe} + W_{Y_{Fe}})}{620}$ |
| (18) | $\frac{\partial Y_{Fe}}{\partial X_2} = -7.46 - 0.06X_1 + 0.14X_2$ | (37) | $0 \leq \mu_O \leq 1$ |
| (19) | $E_{Fe} = \frac{140X_1X_2}{3(620 - Y_{Fe})}$ | | |

the upper boundary limit of the energy consumption.

In order to satisfy the objective functions, the constraints were subjected to Eq. (3) – (8). Eq. (3) shows the energy consumption associated with voltage, current, duration time, reacting volume and uranium removal. The responses for uranium removal with respect to the treatment time and current density follows the response surface equation in Eq. (4). Eq. (5) was used to account the cumulative uncertainty of response for uranium removal. The operational cost based on the electrical cost of energy consumption is shown in Eq. (6). The feasible regions of the response and variables tested in this case study are specified in Eqs. (7) and (8), respectively.

### 2.2.2. Multi-objective decision making through fuzzy logic

From the set of Pareto optimal solutions, the best solution was selected using a multi-criteria analysis [36]. This is due to the disadvantage of the Pareto set analysis in defining the specific optimum compromise solution of multiple objective functions. Thus, the fuzzy mathematical programming method was interjected to address the disadvantage of the resulting Pareto set in order to appropriately determine a solution to the multi-objective decision-making problem [37]. The overall satisfaction degree was maximized by utilizing the idea of max-min aggregation where the degree of satisfaction was concurrently optimized [37,38]. Moreover, the degree of satisfaction for the energy consumption and the retained uranium content in account of its cumulative uncertainty of the results must satisfy the overall satisfaction shown in Eq. (9).

The constraints used to attain the fuzzy goal is given in Eq. (10)–(14). The linear membership function for minimizing the energy consumption and overall retained uranium concentration are shown in Eqs. (10) and (11), respectively. A limiting constraint in the aspect of a specified maximum contaminant level with respect to the retained uranium concentration in account of its cumulative uncertainty of results is defined in Eq. (12). If the constraint of Eq. (12) is infeasible, then Eq. (13) would be utilized to determine the retained uranium content with respect to the maximum contaminant limit. Eq. (14) pertains to the level of satisfaction that could only be adjusted within its feasible region.

## 3. Results and discussion

### 3.1. Comparison of uranium removal and operational cost on treatment time

The time of electrolysis is related to the charge loading effect that influences the performance of electrocoagulation [39]. The specific effect of treatment time at a constant current density of 70 mA/cm$^2$ towards the capacity of uranium removal and its respective operational costing are illustrated in Fig. 1.

The trend from the treatment time of 0–120 min showed that uranium is effectively removed throughout a higher treatment time interval. For the iron-stainless steel system shown in Fig. 1(a), the uranium concentration nearly reached a complete removal attaining 9 μg/L at a treatment time of 90 min. On the other hand, the aluminum-stainless steel system in Fig. 1(b) showed the lowest uranium concentration at 96 μg/L that was retained at the maximum treatment time of 120 min for the given set of process parameters for this case study. The removal of uranium content is associated with the critical generation of metal ions that enables the formation of floc at a rapid pace [40]. The electrocoagulation process neutralizes the negatively charged colloidal particles through the collisions of the iron and aluminum cations [41]. Throughout the whole treatment duration, results exhibited that the iron-stainless steel electrode showed a higher uranium removal rate as compared to the aluminum-stainless steel electrode. This is due to the interaction of uranium towards the iron-stainless steel electrode that promotes a hematite-$UO_2^{2+}$ interaction, while the use of the aluminum-stainless steel electrode tends to have a repulsive effect between the $Al^{3+}$ and $UO_2^{2+}$ species [10].

In terms of the operational cost based on the energy consumption, the iron-stainless steel system (Fig. 1(a)) observed an incremental energy consumption trend from 362.9 kWh/g-U to 632.3 kWh/g-U at 30 min–120 min, respectively. This is similarly reported in the study of Hansen et al. [42] where higher treatment time concurrently gives rise to the energy per amount of contaminant removed in the electrocoagulation process. Thus, the operational cost in terms of the electricity prices at the end of the treatment time ensued values of 82.2 USD/g-U (United States), 75.9 USD/g-U (South Korea) and 107.5 USD/g-U (Finland). Conversely, the aluminum-stainless steel system (Fig. 1(b)) gave a decreasing trend in the energy consumption from 1645.0 kWh/g-U to 747.4 kWh/g-U at its corresponding treatment time from 30 min to 120 min. At a treatment time of 30 min, the operational cost according to electrical costs per country indicated the values of 97.2 USD/g-U (United States), 89.7 USD/g-U (South Korea) and 127.1 USD/g-U (Finland). Based on the energy consumption principle in Eq. (3), a simultaneous decline with both energy utilization and uranium concentration at increasing treatment time indicates an inefficiency in the electrocoagulation process. This implies that a lower treatment time requires more energy consumed in the aluminum-stainless steel system due to achieving minimal uranium removal.

### 3.2. Comparison of uranium removal and operational cost on current density

The current density is an essential parameter in order to control

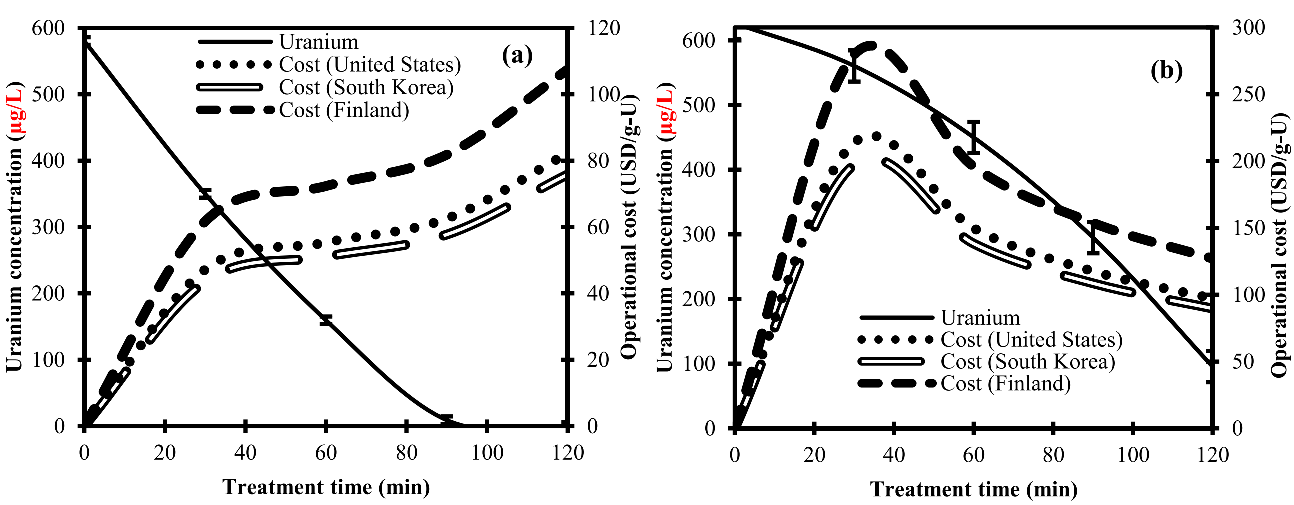


**Fig. 1.** Electrocoagulation removal of uranium and its operational cost at varying treatment time using (a) iron stainless steel and (b) aluminum stainless steel anode/cathode systems.

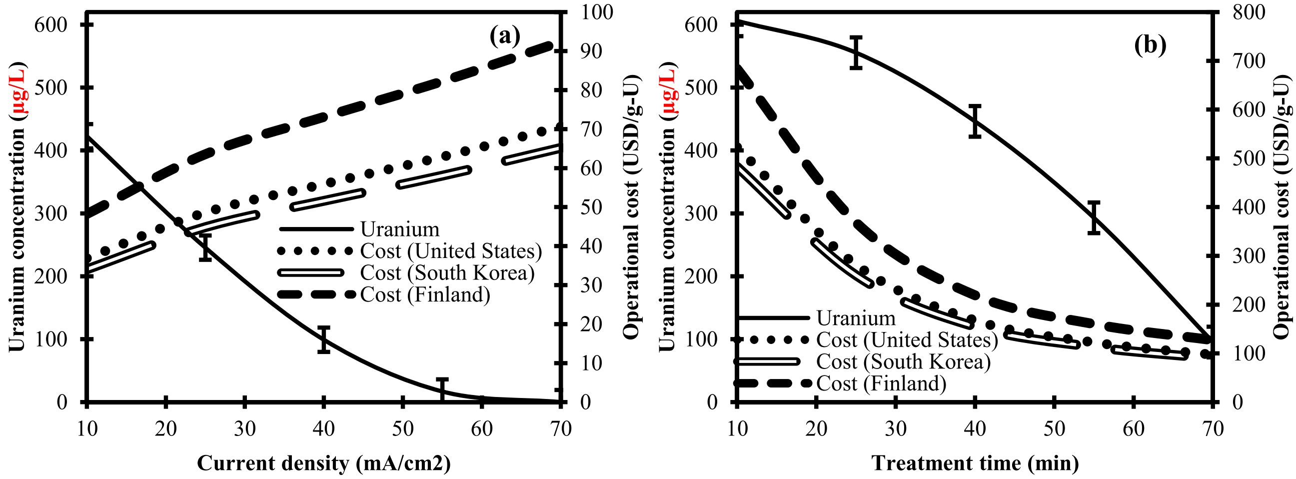


**Fig. 2.** Electrocoagulation removal of uranium and its operational cost at varying current density using (a) iron stainless steel and (b) aluminum stainless steel anode/cathode systems.

the reaction rate towards the regulation of bubble production and coagulant production rate that influences the growth of flocs in an electro-coagulator setup [43]. The combinations of iron-stainless steel and aluminum-stainless steel anode/cathode were tested and compared with respect to uranium removal and its corresponding operational cost at varying current densities in Fig. 2. A constant reaction time of 120 min was set for the comparative assessment in the electrocoagulation treatment process.

Higher uranium removal was observed at the increasing current density level in the electrocoagulation process. This is attributed to a high generation of ions leading to the pollutant molecules being destabilized at a higher current density, which would then result to the agglomeration of induced flocs [44]. At the current densities of 10 mA/cm$^2$ to 70 mA/cm$^2$, the iron-stainless steel (Fig. 2(a)) and aluminum-stainless steel (Fig. 2(b)) combinations in the electrocoagulation process showed uranium removal of 27.2%–100% and 3.3%–84.8%, respectively. The removal of uranium is observed to be higher in the iron electrode than that of the aluminum electrode throughout the tested current density range. This is due to the iron electrode ($Fe^{3+}$: 10 μm–30 μm) being able to release larger cations than the aluminum electrode ($Al^{3+}$: 0.05 μm–1 μm), which results to higher uranium removal efficiencies [45]. Based on the solubility diagram presented in the study of Nariyan et al. [10], its redox potential-pH diagram supports that the iron electrode decreases the redox potential and increases the pH greater than the aluminum electrode that explains the superiority of iron over aluminum in terms of uranium removal.

The iron-stainless steel combination (Fig. 2(a)) exhibited increasing energy consumption of 283.7 kWh/g-U to 373.9 kWh/g-U at its corresponding applied current densities of 10 mA/cm$^2$ to 70 mA/cm$^2$. In the electrochemical reactions of the formation of iron hydroxides, the removal of uranium in this system needs a minimum energy requirement in order to overcome the resistances in the aspect of over potential, concentration polarization and Nernst potential [42]. As a result, the operational cost upon the complete removal of uranium at a current density of 70 mA/cm$^2$ based on electricity cost in the United States, South Korea and Finland are 70.6 USD/g-U, 65.2 USD/g-U and 92.3 USD/g-U, respectively. On the other hand, the aluminum-stainless steel combination (Fig. 2(b)) showed a substantially high energy consumption from an initial of 4021.6 kWh/g-U to 747.4 kWh/g-U at its respective current densities of 10 mA/cm$^2$ to 70 mA/cm$^2$. The trend in the operational cost at its initial current density (10 mA/cm$^2$) based on the electricity costing in the countries of the United States, South Korea and Finland resulted to the highest costs of 522.8 USD/g-U, 482.6 USD/g-U and 683.7 USD/g-U, respectively. This trend could be explained in the theoretical definition of the energy consumption in Eq. (3). A high energy consumption at low current density indicates an inadequate degree of uranium removal. Conversely, higher current density would result to lower energy consumption due to a more efficient removal of the uranium content. In terms of the initial capital cost associated with the sacrificial material, the iron electrode has an advantage over the aluminum electrode due to having a lower purchasing cost. This supports the efficient use of iron electrodes in having a high uranium removal capacity at a lower cost.

### 3.3. *Pareto optimality analysis*

The boundary limits (high and low) of the retained uranium concentration in the account of the cumulative uncertainty of the response and the energy consumption of the electrocoagulation process in the application of iron-stainless steel and aluminum-stainless steel systems are determined through the Pareto set analysis. The resulting Pareto efficiency of the systems is based on the equation generated from the central composite face-centered design (CCFD) in the response surface methodology for the electrocoagulation process. This in turn is beneficial for the selection of the preference criterion in the latter part of the decision-making analysis. In this case study, non-linear model equations are utilized in the optimization analysis. This would lead to the presence of multiple local optimums that would not directly give the essential global optimum to achieve its best possible solution in the given constraints of the objective function [47]. Therefore, the Lingo software is used for its global optimizer option enabling to reach a global optimal solution.

#### 3.3.1. *Pareto analysis in the case of the iron-stainless steel electrode combination*

The iron-stainless steel system for uranium removal in the Pareto analysis is portrayed in Eq. (15) – (25). The generated and validated model equation in Eq. (15) is derived from Nariyan et al. [10] as the basis to describe the retention of uranium concentration as part of the case study of the batch electrocoagulation process. Eq. (16) indicates the cumulative uncertainty that is essential towards the response. This is determined through the partial derivatives with respect to treatment time and current density as shown in Eqs. (17) and (18), respectively. A vital parameter in the Pareto optimal analysis is the energy consumption of the electrocoagulation process in Eq. (19). The energy consumption is then used to calculate the operational cost based on the electricity cost in the United States, South Korea and Finland as shown in Eq. (20) − (22), respectively. The boundary limits in Eq. (23) − (25) refers to the uranium concentration and variables of treatment time and current density, respectively, generated on the basis of a CCFD.

The global optimum in the iron-stainless steel system after minimizing the objective function of the sum of Eqs. (15) and (16) subjected to Eq. (17) − (25) have achieved a complete uranium removal with a cumulative error of uncertainty of 15 μg/L U. Results indicated that the associated energy consumption to attain the global minimum value for uranium content is 491.8 kWh/g-U, which is designated as its upper boundary limit. This is equivalent to the operational costs of 63.1 USD/g-U, 58.2 USD/g-U and 82.5 USD/g-U based on the electricity cost in the United States, South Korea and Finland, respectively. The parametric conditions following the minimum uranium concentration are the treatment time of 92.1 min and current density of 70.0 mA/cm$^2$.

Additionally, a separate objective function is set to minimize the energy consumption of the electrocoagulation process based on the process variables in Eq. (19) subjected to the constraints of Eq. (15) − (18) and (20)–(25). This is needed in order to determine the lower and upper boundary limits of energy consumption and uranium concentration, respectively, essential for the Pareto optimality analysis. The global optimum values led to the absence of energy consumption and an unreacted uranium concentration of 620 μg/L. This is due to the lower limit of 0 min at the treatment time set in the model equation. The absence of the treatment time would also lead to generating no operational cost.

Fig. 3(a)-(c) are the graphical illustration for the Pareto optimality that comprises the identified boundary limits utilizing the results of the following: (1) least possible retention of uranium concentration and its respective cumulative uncertainty of the results and (2) the minimized energy consumption in relation to its corresponding operational cost. The results of the Pareto analysis demonstrate that a lower overall retained uranium content would render a higher energy consumption with its corresponding operational cost in the electrocoagulation process using an iron-stainless steel anode/cathode combination. This trend is attributed to a higher operating condition that essentially adds to greater energy consumption translating to higher operational cost to successfully remove the radioactive uranium material. Larger uranium removal efficiencies with higher parameters of electrocoagulation time and current density can be explained accordingly in Faraday's law [46]. This is wherein higher amounts of iron cations are released from the anode into the water that functions as a coagulant to promote the removal of the anionic target material at greater parametric levels [40].

### 3.3.2. Pareto analysis in the case of the aluminum-stainless steel electrode combination

Eqs. (26) − (33) are for the Pareto set analysis of the aluminum-stainless steel system for uranium removal. The uranium removal model equation in Eq. (26) for this case study is derived from Nariyan et al. [10] that simulates the electrocoagulation process in the aluminum-stainless system. The cumulative uncertainty of the experiment is included in Eq. (27) to account for errors associated to the accuracy in the variables through its partial derivatives in Eqs. (28) and (29). Eq. (30) describes the energy consumption integrated in the Pareto analysis for uranium removal in a batch electrocoagulation process. Furthermore, the operational costs are calculated based on electrical costing in the United States, South Korea and Finland in Eq. (31) − (33), respectively. Similar boundary limits are utilized in Eqs. (24) and (25) for the variables used in the CCFD.

The global optimum values for minimizing the objective function in Eqs. (26) and (27) subjected to the constraints of Eqs. (24), (25) and (28) − (33) are used to simultaneously determine the lowest possible retained uranium concentration with its respective energy consumption value. The results led to 95.5 μg/L uranium concentration with a cumulative uncertainty of experimental result of 73.2 μg/L. Furthermore, the corresponding energy consumption of 747.4 kWh/g-U is associated towards the global minimum value of the retained uranium concentration in the aluminum-stainless steel system. The corresponding operational costing based on the electrical cost in the United States, South Korea and Finland resulted to 97.2 USD/g-U, 89.7 USD/g-U and 127.1 USD/g-U. This condition is attained at the parametric conditions of 120 min and 70.0 mA/cm$^2$ for the treatment time and current density, respectively.

The detailed representation of the Pareto optimality plot in Fig. 3(d)–(f) shows the relationship of uranium removal and energy consumption with its corresponding operational cost in the aluminum-stainless steel system. Results indicated a clear inefficiency in utilizing the aluminum electrode. This is due to a small reduction in the overall uranium content that correlates to high operational cost and energy consumption. Moreover, the results in-line with the energy consumption in the electrocoagulation setup are consistent with the study of Nariyan et al. [48], wherein the aluminum-stainless steel system was found to have a higher energy demand than in the iron-stainless steel system. It was therefore determined that iron is the best anode to be utilized in the remediation of uranium due to simultaneously being less energy intensive, cheaper operational cost and superior removal capacity over the aluminum anode. Moreover, the global minimum uranium content generated by the aluminum-stainless steel system was not able to satisfy any of the standard regulatory limits for the radioactive waste contaminant set by various organizations worldwide. Thus, only the iron-stainless steel system used for the electrocoagulation process is feasible to be subjected to the fuzzy multi-objective optimization to determine the optimal uranium removal efficiency at the most economic cost.

### 3.4. Multi-objective decision making through fuzzy optimization

The decision making strategy in this case study carried out fuzzy optimization on the electrocoagulation setup for the iron-stainless steel combination. This consists of a main objective function subjected for global optimization in Eq. (34). Moreover, the fuzzy constraints utilized to reach the fuzzy goals are subjected to Eq. (35) − (37). A linear membership function is utilized in the decision making process for its capacity to estimate any non-linear equations and its favorability to describe subjective preferences with regard to the objective uncertainty [49]. Therefore, the linear membership function upon the minimization of the energy consumption (Fig. 4(a)) and reducing the overall uranium concentration for a specified maximum contaminant level (Fig. 4(b)) are defined in Eqs. (35) and (36), respectively. The level of satisfaction for both the uranium content with its respective cumulative uncertainty of the result and energy consumption must be able to satisfy the overall degree of satisfaction in Eq. (34). The given sets of boundary limits are incorporated from the Pareto optimality analysis as its basis for the case study in the multi-objective fuzzy optimization. The quantified energy consumption limits of 491.8 kWh/g-U (upper bound) and 0 kWh/g-U (lower bound) are attained from the minimized retained overall uranium content and consumed energy from the electrocoagulation process,

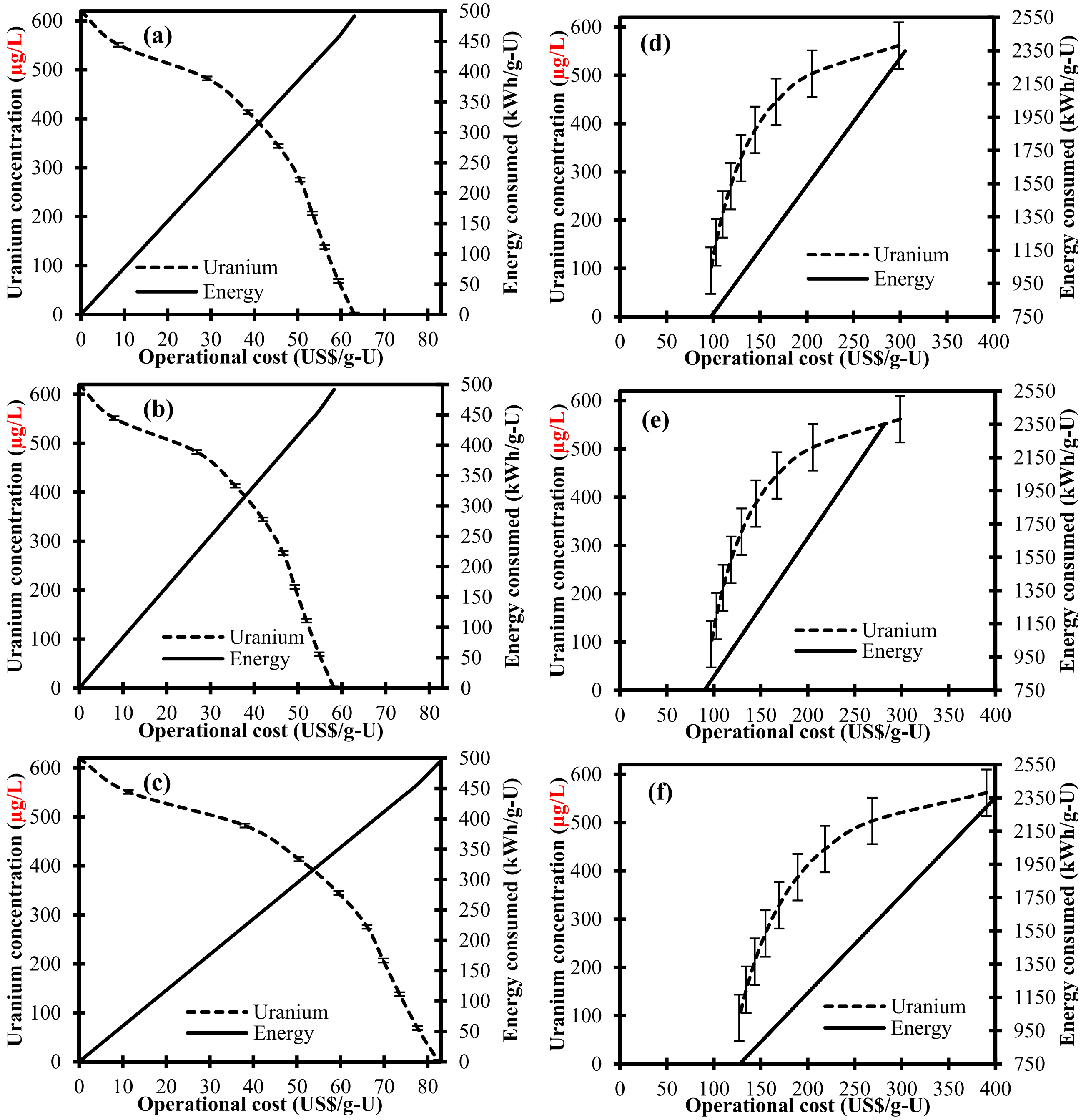


**Fig. 3.** Operational cost Pareto plot of uranium removal and energy consumption for iron stainless steel anode/cathode system in terms of the currency at the (a) United States, (b) South Korea and (c) Finland and for aluminum stainless steel anode/cathode system in terms of the currency at the (d) United States, (e) South Korea and (f) Finland.

respectively. Moreover, the upper boundary of the total uranium content retained at 620 μg/L is derived from the least possible energy consumption in the electrocoagulation treatment. On the other hand, the lower boundary of the retained uranium concentration of 0 μg/L is obtained from the maximum removal capacity of the iron-stainless steel system. In Eq. (37), the numerical implications of "0" and "1" refers to the dissatisfaction and satisfaction ratings, respectively. Additional constraints in Eq. (15) – (25) are also subjected in the case study of the multi-objective fuzzy optimization process.

The results on the feasibility of the generated model with regards to the fuzzy analysis in this case study are listed in Table 4. Different maximum contaminant level were set to satisfy the stringent environmental regulation from various organizations [10,50]. Results indicated that the total uranium concentration (with the cumulative uncertainty of result) of the iron-stainless steel anode/cathode combination model are only feasible with the maximum contaminant levels from 20 μg/L – 70 μg/L, while lower uranium concentration standard in the European Union and World Health Organization are infeasible. This implies that the iron-stainless steel system is not capable of handling lower amounts of uranium (≤15 μg/L) with respect to the possible instrumental error affecting the accuracy obtained from the process parameters. However, the concentrations of 10 μg/L and 15 μg/L are

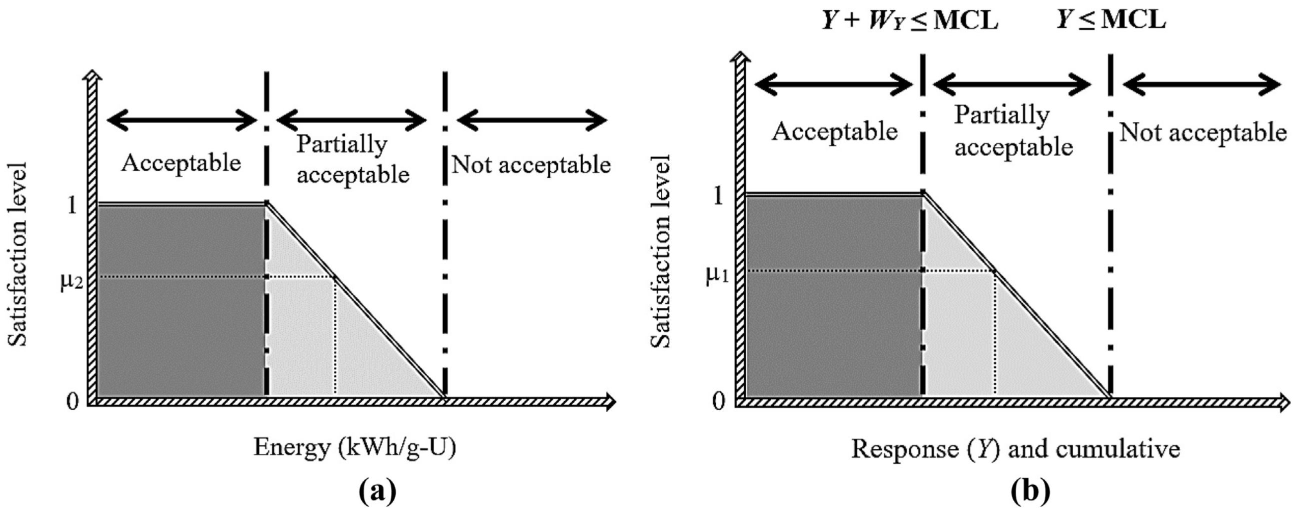


**Fig. 4.** Linear membership function for the (a) energy consumption and (b) response in account with the uncertainty of results.

**Table 4**
Uranium removal feasibility by electrocoagulation in the frame of an iron–stainless steel anode/cathode system based on various maximum contaminant limit.

| Organization | Maximum contaminant level (μg/L) [10,50] | $Y_{Fe}+W_{Y_{Fe}}$ | $Y_{Fe}$ |
|---|---|---|---|
| Brazil Environmental Agency | 20 | Feasible | – |
| Department of Water Affairs and Forestry | 70 | Feasible | – |
| European Union | 10 | Infeasible | Feasible |
| Finland | 30 | Feasible | – |
| Health Canada | 20 | Feasible | – |
| Ministry of Environment in Korea | 30 | Feasible | – |
| National Health and Medical Research Council in Australia | 20 | Feasible | – |
| United States Environmental Protection Agency | 30 | Feasible | – |
| World Health Organization | 15 | Infeasible | Feasible |

**Table 5**
Fuzzy optimal solutions based on various maximum contaminant level on the iron–stainless steel anode/cathode system.

| Parameters | Unit | Feasible at $Y_{Fe}+W_{Y_{Fe}}$ | | | Feasible at $Y_{Fe}$ | |
|---|---|---|---|---|---|---|
| | | MCL:20 μg/L | MCL:30 μg/L | MCL:70 μg/L | MCL:10 μg/L | MCL:15 μg/L |
| $\mu_{Overall}$ | % | 4.0 | 6.1 | 11.6 | 7.0 | 7.7 |
| $\mu_1$ | % | 96.8 | 95.2 | 88.7 | 94.0 | 93.2 |
| $\mu_2$ | % | 4.0 | 6.1 | 11.6 | 7.0 | 7.7 |
| Uranium concentration ($Y_{Fe}$) | μg/L | 0 | 5 | 44 | 10 | 15 |
| Cumulative uncertainty ($W_{Y_{Fe}}$) | μg/L | 20 | 25 | 26 | 27 | 27 |
| Overall ($Y_{Fe}+W_{Y_{Fe}}$) | μg/L | 20 | 30 | 70 | 37 | 42 |
| Energy consumption | kWh/g-U | 472.2 | 461.7 | 434.5 | 457.4 | 453.8 |
| Operational cost (United States) | USD/g-U | 61.4 | 60.0 | 56.5 | 59.5 | 59.0 |
| Operational cost (South Korea) | USD/g-U | 56.7 | 55.4 | 52.1 | 54.9 | 54.5 |
| Operational cost (Finland) | USD/g-U | 80.3 | 78.5 | 73.9 | 77.8 | 77.2 |
| Treatment time | min | 96.4 | 101.6 | 95.2 | 104.1 | 103.3 |
| Current density | mA/cm$^2$ | 65.0 | 59.9 | 56.3 | 57.4 | 56.9 |

only feasible without accounting the cumulative uncertainty of the result for uranium removal. This in turn is only partially acceptable in the fuzzy analysis (Fig. 4(b)).

Table 5 summarizes the global optimum results obtained from the simulated fuzzy mathematical programming optimization approach. The results indicated that the acceptable maximum $\mu_O$ values for the maximum contaminant level for uranium at 20 μg/L, 30 μg/L and 70 μg/L are 4.0%, 6.1% and 11.6%, respectively, while the partially acceptable maximum $\mu_O$ values for the maximum uranium content for 10 μg/L and 15 μg/L are 7.0% and 7.7%, respectively. The overall degree, $\mu_O$, indicates that the aggregated satisfaction rating is able to achieve the fuzzy goals subjected to its fuzzy constraints. This is essential towards deriving optimum results in the multi-objective functions against other optimization methods that only deals with uni-objective models. Furthermore, the corresponding values of $\mu_O$ in the fuzzy optimization technique are attributed to the appropriate allocation scheme that attains a compromise solution for the best combination in the removal of uranium at the least possible consumed amount of energy. Thus, the application of the fuzzy theory would be favorable towards the most practical and economic operational cost in the electrocoagulation process using the iron-stainless steel anode/cathode combination. The optimal fuzzy solution is obtained from a simultaneous minimization of the total uranium concentration and energy consumption resulting in the optimized operating parametric conditions. To be specific, the optimum parameters at 101.6 min and 59.9 mA/cm$^2$ to satisfy the

30 μg/L ($Y_{Fe}$= 5 μg/L and $W_{Y_{Fe}}$ = 25 μg/L) total uranium concentration resulted to the operational costs of 60.0 USD/g-U, 55.4 USD/g-U and 78.5 USD/g-U according to the electrical costs and regulatory limits in the United States, South Korea and Finland, respectively. An allocated energy consumption of 461.7 kWh/g-U was able to satisfy the set objective function for the decision-making analysis. In general, all the set maximum contaminant levels in various organizations are only able to be satisfied at the treatment time and current density above its middle range. This is consistently in-line with Faraday's law in which a higher time and current are able enhance the metal cation production rate in the cathode solution [40]. The metal cation in the system can precipitate to a polymerized amorphous hydroxides that are favorable to remove the contaminant through sweep coagulation [51]. Thus, the generated global fuzzy optimum solutions and its respective conditions in this case study are able to effectively satisfy the given uranium concentration limit. The incorporation of operational cost through the energy consumption and the cumulative uncertainty of the response criteria has successfully drawn out essential compromise results in the decision-making process.

### *3.5. Comparison to previous results, implications and future works*

In the previous study of Nariyan et al. [10], the optimum parameters of 70 mA/cm$^2$ and 120 min in the iron-stainless steel attained a computed cost of 82.2 USD/g-U. As compared to the current study, the compromise solution to the adherence with the uranium standards at 20 μg/L, 30 μg/L and 70 μg/L at the least possible cost resulted in being cheaper by 25.3% (61.4 USD/g-U), 27.0% (60.0 USD/g-U) and 31.3% (56.5 USD/g-U), respectively. Thus, the application of the fuzzy theory could draw out a cost-effective output towards avoiding high environmental impact of uranium in the effluents at a more economical cost.

In-line with the possible handling and management requirements of the radioactive sludge, this would include several steps that can affect the expenditure cost [52]. The sludge would typically undergo a dewatering process. The dewatered sludge with its filter bags would be stored prior to its disposal via forklift with a drum attachment. The disposal of the dewatered sludge involves the common mixture of a chemical stabilizing material in the drum. These drums would then be stored in an environmentally appropriate storage facility. The drummed PPE would then be further screened for radioactivity and disposed according to the designated standard regulatory requirements. Thus, the future outlook of this research can include an elaborated overall expenditure cost analysis for possible up-scale basis in the uranium removal through the electrocoagulation process.

## 4. Conclusions

In this research, a decision-making analysis was done for the remediation of radioactive waste in the case study for the reduction of uranium concentration in a batch electrocoagulation process through the multi-objective fuzzy optimization analysis. A systematic approach by the incorporation of the cost criteria through the energy consumption and the cumulative uncertainty of retained uranium concentration were done to realize the optimal solutions. The lower and upper boundary limits of the iron-stainless steel system (energy consumption: 0–491.8 kWh/g-U and total uranium concentration: 0–620 μg/L) were determined through the various combinations in the parameters of treatment time and current density in the Pareto set analysis. For the aluminum-stainless steel combination, it was found out that the global minimum uranium concentration was not able to satisfy any of the stringent environmental standards. Therefore, only the iron-stainless steel anode/cathode combination model was feasible towards the decision-making analysis using fuzzy optimization. The innovative utilization of fuzzy logic indicated the overall maximum satisfaction ranging from 4.0% to 11.6%. This attained a properly allocated compromise solution in terms of the uranium removal at the least possible consumed energy in the electrocoagulation setup. Thus, the criteria utilized in this study have proven to be effective in properly achieving the uranium standards in various organizations in consideration of the energy consumption. This in turn could serve as a pioneer in succeeding optimization applications that have multiple criteria requirements.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2019.12.016.

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