

## Linear Genetic Programming for Prediction of Nickel Recovery from Spent Nickel Catalyst

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**Abstract: Problem statement:** In this study Linear Genetic Programming (LGP) and statistical regression are used in predicting Current Efficiency (CE) of Electro deposition cell used for recovery of nickel from spent nickel catalyst. **Approach:** The Nickel electro deposition from spent catalyst leachate solutions was studied to determine the effect of the operative conditions such as nickel concentration, temperature, current density and time on the CE of the unit cell. **Results:** For this purpose, LGP and regression models were calibrated with training sets and validated by testing sets. Additionally, the robustness of the proposed LGP and regression models were evaluated by experimental data, which are used neither in training nor at testing stage. The results showed that both techniques predicted the CE data in quite good agreement with the observed ones and the predictions of LGP are challenging. **Conclusion/Recommendations:** The performance of LGP, which was moderately better than statistical regression, is very promising and hence supports the use of LGP in simulating the electro deposition of Nickel from spent Nickel catalyst.

**Key words:** Linear programming, regression, modeling, spent nickel catalyst

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### INTRODUCTION

The demand for nickel has recently risen for its promising use in rechargeable batteries. The increase in the industrial demand for this metal has brought forth a steady growth in the need for refining of the metal. More importantly, the recovery of nickel from secondary sources such as spent catalyst could minimize landfill disposal and the waste of natural resources. In Egypt, large amount of Nickel catalysts are used by different industries resulting in the production of large amount of solid waste containing nickel which is the spent or deactivated catalysts. These catalysts contain from 14-24% nickel based on the production company and the process (Ghanem *et al.*, 2008). The catalysts have an active life of 3-5 years on the most after which it is deactivated and must be replaced. Therefore, over the last few years, tons and tons of spent reforming catalysts are being pulled up. The accumulating mass of spent catalysts is posing a great pollution problem for the concerned industry in particular and the country in general. Many researchers have studied the subject of recovering nickel from spent catalyst. The different studies dealt with the leaching of nickel from spent catalyst with acids and alkalis

(Amhad *et al.*, 1970; Singh, 1993). Some other researchers studied the nickel electro-winning (Khanna *et al.*, 2000) based on sulphate and/or chloride electrolyte. Rambla *et al.* (1999) and Alfantazi and Shakshouki (2002) Performed nickel electro winning using platinum catalyzed hydrogen diffusion anode and the effect of chloride and sulphate ions. Lupi and Pasquali (2003) and Zhou *et al.* (1997) Studied the electrolytic nickel recovery from lithium ion batteries. Nickel electro winning is performed from electrolytes containing  $\text{NiCl}_2$  and/or  $\text{NiSO}_4$  in the presence of high concentrations of  $\text{H}_3\text{BO}_3$ .

Recent years have seen an increase in the development of data modeling (using experimental data to build an input-output model that describes the response of process outputs to changes in inputs) using different techniques such as regression and linear genetic programming (McKay *et al.*, 1997). Statistical regression is a common approach to develop empirical process models (Seber, 2003), but the resulting models are accurate only within the ranges of data from which they are developed (Chan *et al.*, 2010). Genetic Programming (GP), on the other hand, can be used to generate models with higher order and interaction terms. One of the earliest users of GP in control was

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Mark Willis' Chemical Engineering group in Newcastle. e.g., they used GP to model flow in a plasticizing extruder (Willis *et al.*, 1997). They also modeled extruding food (McKay *et al.*, 2000). Domingos *et al.* (2005) worked on simulations of nuclear reactors (PWRs to be exact) to devise better ways of preventing xenon oscillations. In this study experimental data were collected to study the influence of the electrolyte composition and electrolysis conditions on Nickel recovery from spent catalyst leachate. Then statistical regression and linear genetic programming are used as simulating tools in predicting Current Efficiency (CE) of Electro deposition cell used for recovery of nickel from spent nickel catalyst.

### MATERIALS AND METHODS

The waste catalyst was supplied by Abo-Qir fertilizers Company, Alexandria, Egypt. Table 1 and Table 2 summarized the physical and chemical properties of the reformer catalyst.

Sulphoric acid, boric acid, nickel chloride and deionized water were used in all experiments. The purpose was to study the effect of concentration, temperature, current density and time on the on the cell performance.

**The cell set up:** Figure 1 shows a schematic representation of the electrolytic cell used for the electro-deposition of nickel and the accompanying electrical circuit.

The solid catalyst in the form of cylinder was crushed and grounded till size range 0.2-0.3 mm (Gaballah and Djona, 1994), the powder obtained is then dried at 110-120°C. About 50 g of dried catalyst powder was placed into a flask and 250 mL of sulphoric acid were added slowly to the content of the flask with boric acid and nickel chloride. The solution obtained from the leaching spent catalyst was collected which mainly contains nickel sulphate, boric acid and nickel chloride with pH equal 4 was placed in the cell. The stainless steel cathode was cleaned with emery study, washed and then weighed. The anode and cathode are connected as shown in Fig. 1. The current densities used ranged from 20-35 (mA cm<sup>-2</sup>). At the end of the experiment the cathode were removed, washed, dried and weighed. The amount of nickel deposited was determined and the current efficiency and deposition efficiency are then calculated. The efficiency of deposition was calculated as the mass of nickel deposited to the mass of nickel in the leachate liquor and is referred as percentage of recovery.

Table 1: Physical properties of the nickel reformer catalyst

Type	G1-26
Form	Cylindrical
Size "OD*H*ID" (mm)	16×16×8
Bulk density (kg L <sup>-1</sup> )	1.2
Crushing strength (kg cm <sup>-2</sup> )	40
Surface area (m <sup>2</sup> g <sup>-1</sup> )	10
Pore volume (c.c. g <sup>-1</sup> )	0.1

Table 2: Chemical properties of the Nickel reformer catalyst

NiO	18
CaO	8
SiO <sub>2</sub>	<0.2
Na <sub>2</sub> O	<0.2
K <sub>2</sub> O	<0.1
S	<0.02
Cl	<0.002
Al <sub>2</sub> O <sub>3</sub>	Balance

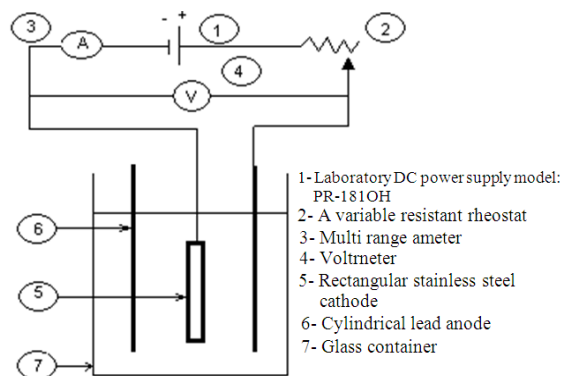


Fig. 1: The cell used

**Experimental design:** Many parameters affect performance of the ED cell. According to our previous study (Ghanem *et al.*, 2009), four parameters were selected. It is believed that they have the greatest effect on CE: Nickel concentration, temperature, current density and time. Four factors with their levels were studied based on the full factorial design:

- Temperature (T): 25, 40 and 60°C
- Concentration (C): 10, 20 and 24 ppm
- Current Density (CD): 18, 24 and 35 (mA cm<sup>-2</sup>)
- Time (t): 60, 120 and 180 min

CE defined as follows was used as a criterion of the cell performance:

$$CE = \frac{zFV_R C}{MIt} \times 100 \quad (1)$$

Where:

z = The number of electrons in the electrochemical reaction (2e<sup>-</sup>)

F = The Faraday constant (96,485 C mol<sup>-1</sup>)

- VR = The volume of solution (L)
- C = The nickel concentration in the electrolytic bath
- t = The time of electrolysis at s
- I = The current applied in A
- M = The molecular weight of nickel (g mol<sup>-1</sup>)

**Analytical method:** Concentration of nickel in the dilute compartment was measured at various operating conditions. In all experiments, atomic absorption was used to measure the amount of Nickel ions in solution.

**Linear genetic programming:** GP has been formulated originally as an evolutionary method for breeding programs using expressions from the functional programming language LISP (Kumara *et al.*, 2005). A new variant of linear GP (Brameier and Banzhaf, 2007) that uses sequences of instructions of an imperative programming language has been employed. More specifically, the method operates on genetic programs that are represented as linear sequences of C instructions. Genetic programs are prediction models that approximate an objective function  $f : I^n \rightarrow O^m$  with  $I^n$  denotes the input data space of dimension n and  $O^m$  is the m-dimensional output data space. In most cases there is only  $m = 1$  output. The objective function  $f$  itself represents the problem to be solved by GP. This function is unknown and defined only incompletely by a training set of input-output vectors:

$$T = \left\{ \left( \vec{i}, \vec{o} \right) \mid \vec{i} \in I' \subseteq I^n, \vec{o} \in O' \subseteq O^m, f(\vec{i}) = \vec{o} \right\}$$

On the contrary, genetic programs normally represent highly nonlinear solutions (Brameier and Banzhaf, 2001). The main differences to conventional tree-based GP or the graph-based data flow that results from a multiple usage of indexed variable (register) contents and the existence of structurally ineffective code (introns) (Brameier and Banzhaf, 2001). This concept was expanded to the Automatic Induction of Machine code by Genetic Programming technique (AIMGP), in which the solutions are directly computed as binary machine codes and executed without using an interpreter, thus in this way the computer program can evolve very quickly (Brameier and Banzhaf, 2001). Each individual program in LGP is represented by a variable-length sequence of simple C language instructions. These instructions operate on one or more Registers(R[i]) or Constants (C) from predefined sets. The LGP procedure is depicted in the flowchart of Fig. 2. The search is done by performing reproduction, crossover and mutation operations as follows:

**Reproduction:** The reproduction operation involves selecting a composite function from the current

population. In this research, we use tournament selection, where a number of individuals are randomly selected from the current population and the one with the highest fitness value is copied into the new population. On the contrary, genetic programs normally represent highly nonlinear solutions (Brameier and Banzhaf, 2001). The main differences to conventional tree-based GP or the graph-based data flow that results from a multiple usage of indexed variable (register) contents and the existence of structurally ineffective code (introns) (Brameier and Banzhaf, 2001). This concept was expanded to the Automatic Induction of Machine code by Genetic Programming technique (AIMGP), in which the solutions are directly computed as binary machine codes and executed without using an interpreter, thus in this way the computer program can evolve very quickly (Brameier and Banzhaf, 2001). Each individual program in LGP is represented by a variable-length sequence of simple C language instructions. These instructions operate on one or more Registers(R[i]) or Constants(C) from predefined sets. The LGP procedure is depicted in the flowchart of Fig. 2. The search is done by performing reproduction, crossover and mutation operations as follows:

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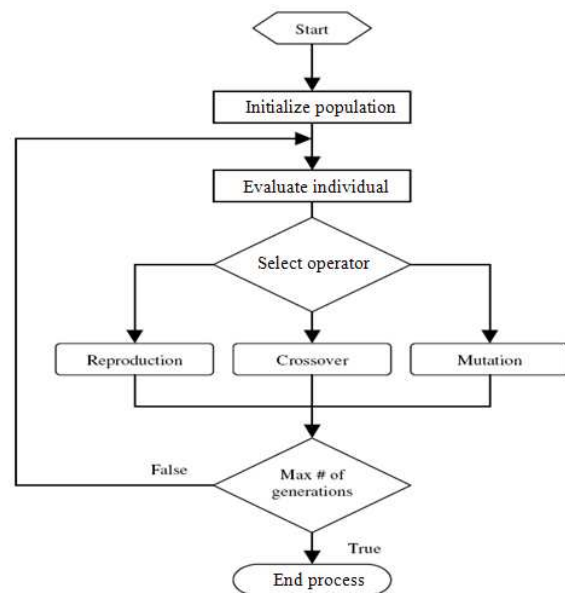


Fig. 2: Genetic programming flow chart

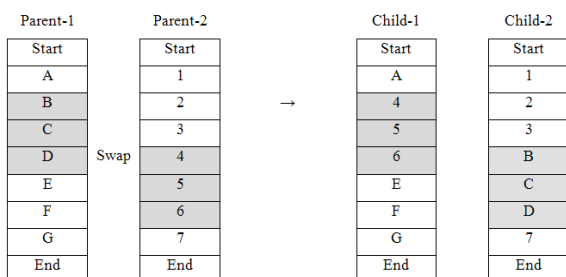


Fig. 3: Crossover operation in genetic programming

**Crossover:** To perform crossover, two code segments are selected on the basis of their fitness values. These two code segments are called parents. Sequence of instructions in each of these two parents is randomly selected and the two selected sequences with these two code segments are exchanged between the parents. In this way, two new composite functions, called offspring, are created. Figure 3 shows an example of crossover between two code segments.

**Mutation:** In order to avoid premature convergence, mutation is introduced to randomly change the structure of some of the individuals to help maintain the diversity of the population. Once a composite function is selected to perform a mutation operation, a code of one instruction in this function is randomly selected and then the code of this instruction is deleted. Another instruction code is randomly generated and this code replaces the previously deleted instruction code. The resulting new instruction represents a new composite function. This new composite function replaces the old one in the population.

## RESULTS

**Effect of initial nickel concentration:** Figure 4 shows the effect of changing the initial nickel concentration on the CE it was found that increasing the initial nickel concentration leads to increasing the CE.

**Effect of time of electro deposition:** The time of electro deposition was varied from 60-180 min (Fig. 5).

**Effect of current density:** Figure 6 show that the amount of nickel deposited slightly changes with changing current density. The amount deposited ranged from 1.03-1.07g in 1 h. Figure 7 shows that a high CE efficiency obtained at lower current density.

**Effect of temperature:** Figure 8 shows the effect of changing path temperature on current efficiency.

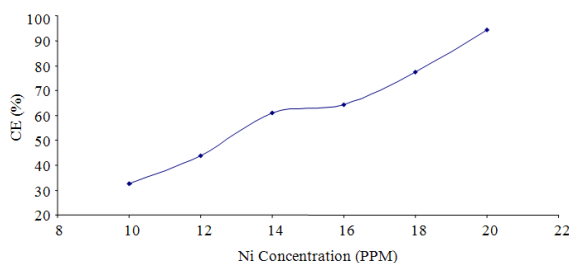


Fig. 4: Effect of changing nickel concentration

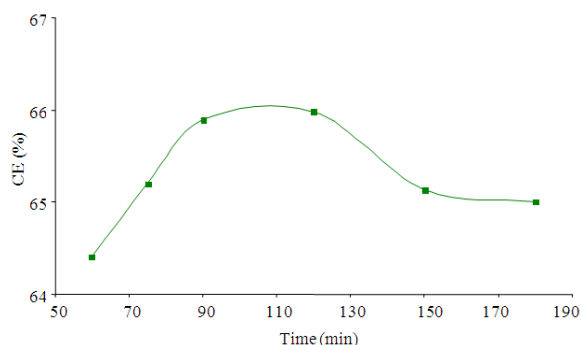


Fig. 5: Effect of changing time of electro deposition

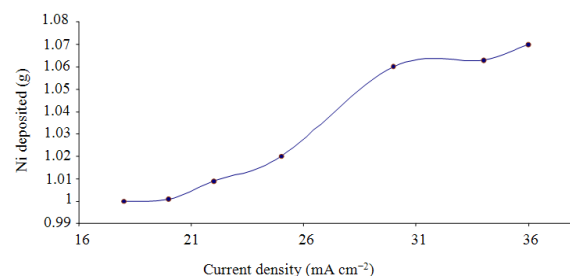


Fig. 6: Nickel deposited Vs current density

**Simulation using regression:** Numerical correlations using regression analysis for CE with Operating condition of the recovery process can presented in the following equation:

$$C.E = 46.87717 + 0.509382 * T - 0.00301 * t + 2.812682 * C - 2.34978 * CD$$

$$R^2 = 0.86$$

Where:

T = Temperature in °C

T = Time in min

C = Concentration of nickel in ppm

CD = The current density in mA cm<sup>-2</sup>

**Linear genetic programming simulation:** In this study, four basic arithmetic operators (+, -, \*, /) and some basic mathematical functions ( $\sqrt{\quad}$ , power) were utilized to get the optimum LGP formulation. A large number of generations are tested to find a formula with minimum error. First, the maximum size of each program was assigned as 256, starting with 64 instructions for program. The functional set and operational parameters used in LGP modeling during this study is given in Table 3. The program is run until there is no longer significant improvement in the performance.

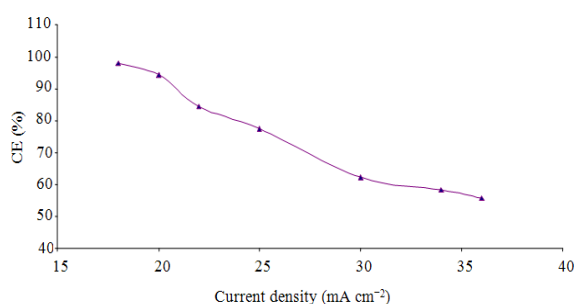


Fig. 7: Effect of changing current density

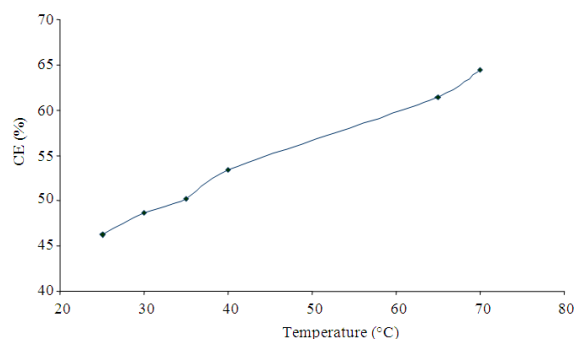


Fig. 8: Effect of changing temperature

Table 3: Parameters of the LGP model

Description of parameter	Setting of parameter
Function set	+, -, *, /, $\sqrt{\quad}$ , power
Population size	1100
Mutation frequency (%)	91
Crossover frequency (%)	64
Number of replication	10
Block mutation rate (%)	30
Instruction mutation rate (%)	30
Instruction data mutation rate (%)	40
Homologous crossover (%)	75
Program size	Initial 64, maximum 256

## DISCUSSION

Figure 4 shows that increasing the initial nickel concentration leads to increasing the CE. This is true since more  $\text{Ni}^{++}$  will reach the cathode leading to a higher amount of nickel deposited and as a result higher CE. This also agrees with Holm and O'Keefe (2000) who reported that a high CE is obtained with high concentration of nickel during electro-winning of nickel from sulphate solution.

Figure 5 shows that time slightly affect the CE % since after one hr the CE is 64.5% while after three hours the CE. 65.5%. So the electro deposition can be carried out for only 1 h. Figure 6 shows that the amount of nickel deposited slightly changes with changing current density. The amount deposited ranged from 1.03-1.07 g in 1 h. high CE efficiency obtained at lower current density. This is agreeing with Lubi and Pilone (2001). They varied the current density from 18-40 Ma  $\text{cm}^{-2}$  and reported that deposition of nickel is accompanied by hydrogen evolution. This can explain the highest CE observed using the lowest current density. This means that increasing current density had no effect on the electro deposition of nickel but another electrolytic process is taking place which is hydrogen evolution.

Figure 8 shows that increasing temperature leads to higher yields of nickel and better current efficiency. At higher temperatures, the mobility of ions increased and the viscosity of solution decreased leading to higher transfer of nickel ions from bulk of solution to the cathode surface. Therefore the current efficiency increased.

From the regression model, it was found that the concentration of nickel and the CD have the most significant effect on the CE while time has almost no significant effect on the CE These conclusions are in good agreement with the experimental results.

From the statistical results of LGP predictions, it can be concluded that LGP model performed better than linear regression. The interested readers may consult the corresponding author for free C codes of the LGP model.

The statistical results of model predictions for training and testing sets are given in Table 3. From the Table 3, it is clear that LGP model predicted the scour depth for both training and testing set with relatively lower error RMSE (0.035 and 0.067) and higher accuracy ( $R^2$ : 0.965 and 0.949), respectively. Further, the graph (Fig. 9) also proves the over performing of the LGP models compared to linear regression models.

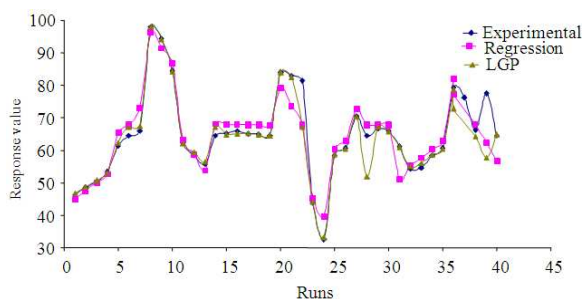


Fig. 9: Comparison between experimental, regression and LGP results

### CONCLUSION

Recovery of Nickel from spent Nickel catalyst leachate solution by electro deposition has been studied. The highest CE was 94% using 20 mA cm<sup>-2</sup>. The amount of nickel recovered increases by increasing the initial nickel concentration and temperature while the electro deposition can be carried out for only One hour since increasing the time more than one hour has no significant effect. Simulation of the electro deposition process using statistical regression was carried out and the model obtained showed that the concentration of nickel and the CD have the most significant effect on the CE while time has almost no significant effect on the CE These conclusions are in good agreement with the experimental results. Simulation of the electro deposition process using LGP carried out and the LGP model predicted the scour depth for both training and testing set with relatively lower error RMSE (0.035 and 0.067) and higher accuracy (R<sup>2</sup>: 0.965 and 0.949), respectively. And the results showed the over performing of the LGP models compared to linear regression models.

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