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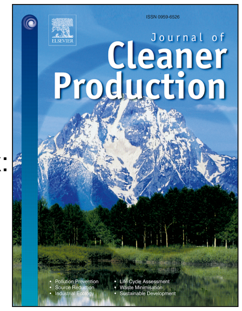

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Maximization of Extraction of Cadmium and Zinc during recycling of spent battery mix: An Application of combined Genetic Programming and Simulated Annealing approach

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Abstract

There are a number of government directives and regulations as well as many public schemes on the recycling of batteries, in spite of this; the quantity of batteries that are actually recycled is still very low. Current production capacity cannot meet projected demand of Lithium-ion batteries. To counter this, the reclamation and repurposing of metals like cadmium, Lithium and Zinc from used or spent batteries is the only viable scheme. This is both environmentally friendly and economically feasible. An alternative is the selective chemical leaching in the presence of Sulfuric acid and Sodium metabisulfite. In this paper, the effect of these chemicals as well as the solid-to-liquid ratio and time of retention is comprehensively studied. Experiments are designed for the recovery of Zinc and cadmium from the spend Lithium-ion batteries mix. To maximize the recovery of Zinc and cadmium, the combined genetic programming and simulated annealing approach is proposed. Genetic programming is used for the formulation of functional relationship between recovered metals Zinc and cadmium and the inputs (Solid/Liquid ratio, concentration of Sulfuric acid, mass of Sodium metabisulfite and retention time). The optimal input conditions determined using the simulated annealing algorithm includes Solid/Liquid ratio of 11.7%, 0.86 M Sulfuric acid, 0.56 g/g of Sodium metabisulfite and 45 minutes of retention time. Three dimensions surface analysis reveals that a lower value of Solid/Liquid ratio favours a better yield. The optimal conditions are validated using experiments. This confirms the efficacy of simulated annealing aided genetic programming techniques as well as the optimal conditions of the metal extraction.

Keywords: Spent battery mix; Metal recovery; Recycling; Genetic programming; Bioleaching

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1. Introduction

Cleaner energy storage systems such as Lithium ion batteries have propelled society to become more mobile and portable (Nemecek, 1994). Despite their considerable advantages, they still pose significant environmental and health hazards. Most of these hazards arise from improper disposal and storage of end-of-life batteries. Global production and consumption of batteries has increased disproportionately to present waste management measures, causing both short and long term issues.

Battery packs are extremely flexible in their design and use, being composed of individual smaller and usually identical cells (Battery pack, 2018). They have longer life span of 2-3 years, and their usage in electric vehicles have resulted in decreased environmental impact when compared to traditional internal combustion (IC) engine-run vehicles when compared using a life cycle assessment (Notter et al., 2010). Regulations have also been passed limiting the amount of dangerous chemicals in batteries, especially mercury (U.S. Environmental Protection Agency, 1997). The end product has become more environmentally friendly than before while being more efficient and resistant to failure. Lithium ion batteries have a very small environmental cost to bear (Boyden, 2014). Other battery chemistries like Ni-Cd pose more significant environmental threats and rising production levels imply higher rates of their consumption. Materials like Cadmium and Cobalt have very adverse effects on both health and the environment (World Health Organisation, 2010; Leyssens et al., 2017). There are several regulations in place limiting the use of these materials in most products. Their disposal and repurposing after they have reached their end-of-life is severely lacking (Official Journal of the European Union, 2006).

In some countries, upwards of 250,000 tonnes of batteries were deemed as waste in 2014 (Eurostat, 2018). In 2016, worldwide consumption of lithium for battery use was 77,821 metric tons of lithium carbonate equivalent (Statista, 2018a). Demand for the metal is projected to reach 422,614 metric tons of lithium carbonate equivalent (Statista, 2018b) by the year 2025. Producers are not currently capable of meeting this demand. In some countries, <2% of all lithium batteries are recycled while the rest are put to landfill (Boyden, 2014). This represents a high threat to public health (Rall & Pope, 1995) and the environment via the leakage of dangerous chemicals (Andresen & Küpper, 2012). It also represents a waste of reusable resources. There is a 30% decrease in overall cost by using recycled materials (Rabah et al., 2008).

Rising production demands can be alleviated by using materials from spent batteries that have undergone a set of recovery and extraction processes. Each process must start with the sorting of various batteries based on their chemical or energy contents (Tonteri et al., 2000). This can be done manually or through some degree of automation (Bernardes et al., 2004). The mix so obtained must undergo extraction to obtain metals like Li, Cd, Ni etc. which are used in further production. These extraction procedures are usually hydrometallurgical for lower value metals, but can also by pyrometallurgical, physical, chemical or biochemical in nature (Li et al., 2009). The set of production treatments depends on the battery chemistry in question (Wang, 2014). One must factor in transport and energy requirements to see the economic feasibility of recycling spent batteries (Niu et al., 2014).

Metal extraction via hydro and pyrometallurgical methods involves a heavy energy intake, as well as high security and pollution risks (Rocchetti, 2013). An alternative to this is chemical and biochemical methods of metal extraction. Bioleaching is one biochemical technique which uses bacteria (like *Acidithiobacillus ferrooxidans* for iron pyrites (Zhang et al., 2008) and *Penicillium citrinum* for low grade manganese ores (Acharya et al., 2002)) to react with the metal to yield soluble products. These soluble products then undergo further filtration to extract metal. This technique provides high yields but requires significant improvements before it can be considered commercially viable (Olson et al., 2003). Metal solubilisation via H_2SO_4 can be performed in a single step leaching process with yields of up to 81% for Cadmium, 96% of Cobalt, 94% of Manganese, 68% of Nickel and 99% of Zinc from a mix generated from spent batteries.

Previous research focussed on the use of response surface methodology (RSM) for modelling and optimizing the metal yields (Tanong et al., 2017). RSM is based on assumption of model structure followed by an estimation of coefficients in the model using optimization methods. This method works satisfactorily if the information about the system behavior is known. Actual engineering problems are often complex, multidimensional, and incomplete information. RSM is no longer suitable. Predictive modelling methods based on Artificial intelligence (AI) seems a better alternative. Among AI methods, evolutionary approach of genetic programming (GP) has the ability to automate the model structure and coefficients estimation resulting in the evolution of the best model (Woodward et al., 1999). The GP model has a free non-linear form that has the best fits. It can adapt to the system behaviour. A number of diverse applications for GP techniques have been found, which shows its effectiveness and efficacy to model the systems of any given complexity.

This study aims to propose a combination of GP and simulated annealing (SA) approach to maximize the recovery of Zinc and Cadmium. The specific works are listed as follows. Firstly, the effect of concentration of H_2SO_4, mass of $Na_2S_2O_5$ as well as the solid-to-liquid ratio and time of retention is comprehensively studied. Secondly, experiments are firstly designed for the recovery of Zinc and cadmium from the spend Lithium-ion batteries mix. Thirdly, GP is used for the formulation of functional relationship between recovered metals Zinc and cadmium and the inputs (Solid/Liquid ratio, concentration of Sulfuric acid, mass of Sodium metabisulfite and retention time). A comparative study between GP, the Box-Behnken model and analysis of variance (ANOVA) analysis has also been performed. Then, the optimal input conditions are determined and validated using experiments. Finally, conclusions are then drawn upon the efficacy of the proposed approach, as well upon metal extraction.

2. Research Problem Undertaken

This section discusses the research problem statement for the combined GP and SA approach for the study of chemical metal extraction from a spent battery mix. A disproportionate amount of spent batteries is not recycled, in spite of various public programmes for the same. Recycling spent batteries to recycle valuable metals is one way to reduce rising demands on production. Recycling batteries consists of sorting, metal extraction and reprocessing. Existing pyrometallurgical and hydrometallurgical extraction techniques require a high energy input while posing significant security and pollution risks. One alternative to the same would be to use chemical extraction using H_2SO_4 and $Na_2S_2O_5$ catalysed chemical leaching. The various parameters such as Solid/Liquid ratio, concentration of H_2SO_4, mass of $Na_2S_2O_5$ and retention time affecting the yield are interdependent to some degree and the appropriate amount of each is unknown that can result in maximization of Zn and Cd. The main problem undertaken in this study is to determine the optimum amount of Solid/Liquid ratio, concentration of H_2SO_4, mass of $Na_2S_2O_5$ and retention time resulting in maximum recovery of Zn and Cd from the spend Li-ion batteries mix. Depiction of the research problem statement is displayed as shown in Figure 1.

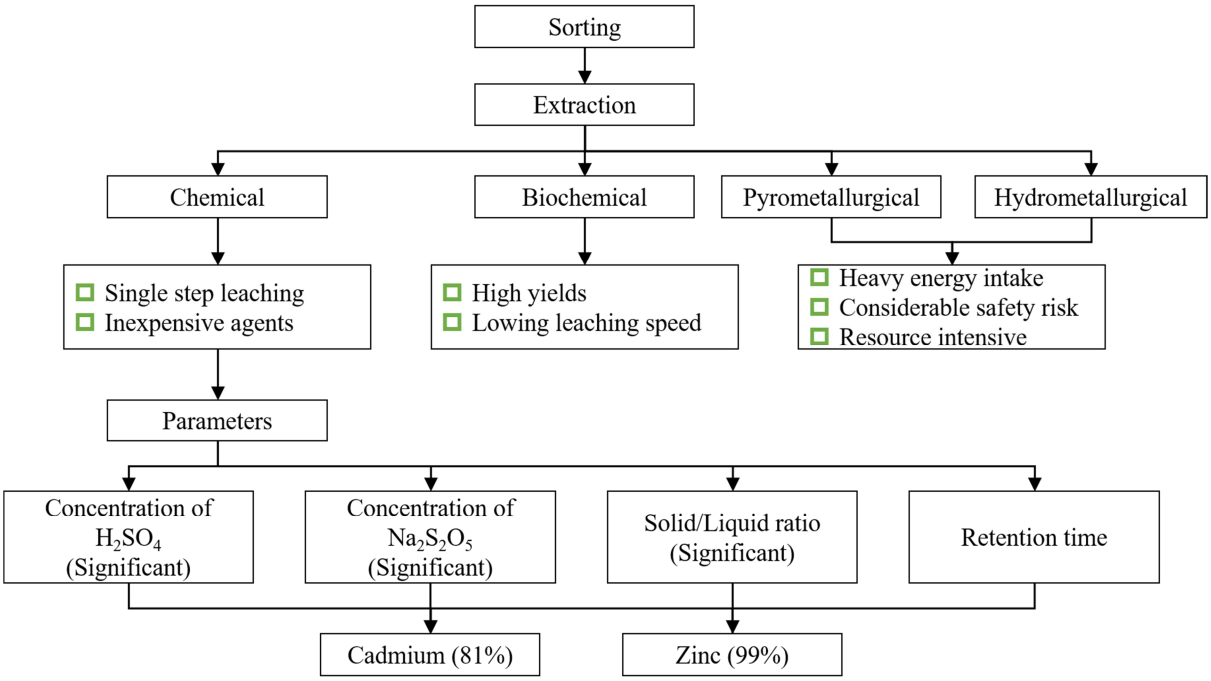


Figure 1. Depiction of the research problem statement

3. Design of Experiment

Samples of spent batteries were collected, manually disassembled and sorted according to the following concentrations (Tanong et al., 2017): 0.28% Li-ion, 0.80% lithium iron sulphide, 1.60% Ni-MH, 15% Zn-C, 14.3% Ni-Cd and 68% alkaline battery. The mix then underwent screening for alien particles including non-metallic components and other contaminants. Metallic composition of the resultant was then determined using inductively coupled plasma-atomic emission (ICP-AE) spectroscopy (Melville, 2014). The battery mix so obtained was then put through a series of experiments, each with different parameters.

Table 1: Box-Behnken optimised parameters

	Solid/Liquid ratio	**concentration of**	**mass of $Na_2S_2O_5$**	**Retention**

	(X_1)	**H_2SO_4 (X_2)**	**(X_3)**	**time(X_4)**
LOW	10%	0.5	0.45	15
MIDDLE	15%	1.0	0.60	30
HIGH	20%	1.5	0.75	45

The mix was added to an Erlenmeyer flask. The primary aim was to study the effects of H_2SO_4 and $Na_2S_2O_5$, as well as to study their interactions. These experiments involved the parameters: Solid/Liquid ratio (x_1), concentration of H_2SO_4 (x_2), mass of $Na_2S_2O_5$ (x_3) and the Retention time of the mixture (x_4). It was originally designed to be performed using a fractional Box-Behnken design, the discreteness of the parameter values. These values are listed in the Table 1. Based on the given range, the random data was generated to simulate data points, and additive white Gaussian noise was added. This was done in MATLAB 2016a as follows:

output_with_noise = awgn (output_without_noise, snr, 'measured') (1)

The code required to generate the data set is provided in Appendix A. It must be noted that for the data generated, snr value was always set to 2. This prevented too much distortion from the original data while also adding enough noise to simulate experimental error. Noise was added to the data to prevent it from overfit scenarios, as well as to simulate experimental error. The data set after the addition of noise with corresponding output is given in Appendix B.

Additive white Gaussian noise is usually added to data sets to simulate error functions. Gaussian noise is a statistical noise whose probability density function is normally distributed. This function is given by:

$$p_G(z) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(z-\mu)^2}{2\sigma^2}} \tag{2}$$

where z is the grey level, μ represents the mean and σ the mean. White Gaussian noise is a special case of the more general Gaussian noise, where values at any pair of times are identically distributed and statistically independent (Salam, 1998). The necessity of adding noise arises due to data biasing, which is a common pitfall in predictive models and inversion algorithms.

The ANOVA and Linear Regression analysis was then applied to the original dataset (Appendix A). This was done to fully compare the efficacies of all three methods in modelling the extraction of metal from a spent battery mix.

4. Genetic programming Approach

Genetic programming (Gandomi et al., 2015), an AI approach stem from the principle of Darwinian evolution i.e. "Survival of the fittest". The procedure involves randomly initialising candidate solutions, which are probabilistically chosen to reproduce basing on their fitness on the output data. Each generation has a fixed population size, where each member is one model. During the initialisation, the initial input and output sets (terminal set), the function space with which to compose model expressions (Figure 2), population and generation size and the number of genes (inheritable model information) need to be specified. Each model is usually represented as a tree as shown, but there have been techniques that use other approaches (Brameier & Banzhaf, 2011).

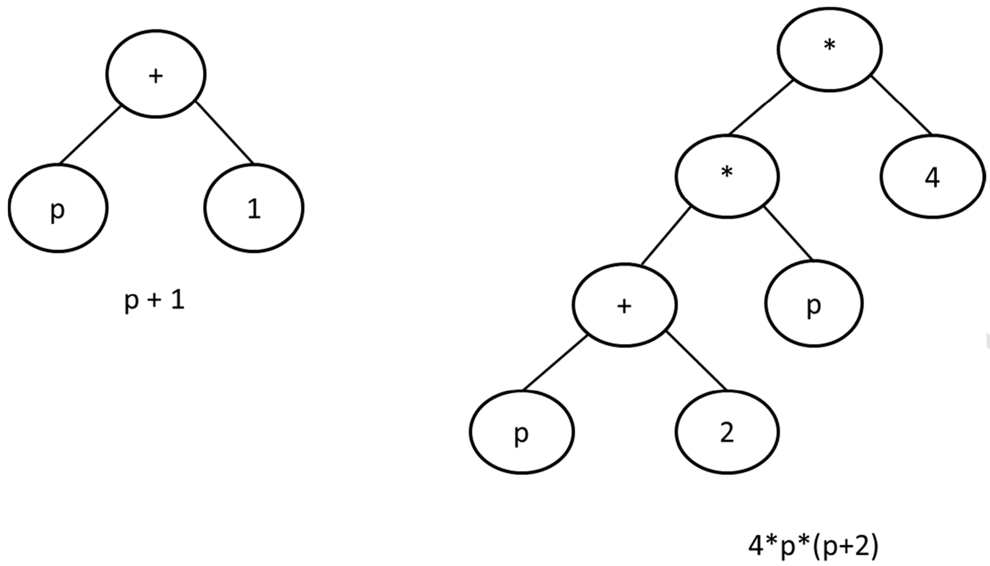


Figure 2. Internal representation of models in GP

Each model has a probability of being chosen for the so-called mating pool depending on their fitness ratio. This criterion of selection favours models that fit data better while also maintaining genetic diversity. Reproduction requires the model information from two candidate solutions, while mutation modifies the data of one model alone. Generally, mutation rates are kept very low.

The main advantage of using genetic programming is the lack of needing to specify specific model equations unlike other techniques (Huang et al., 2018; Garg et al., 2018). GP is able to generate free form equations depending on the functions available to it in the function space due to the nature of model reproduction. This allows for the modelling of highly non-linear models. Another consequence of the technique is also its data agnostic nature i.e. the approach is completely independent of the type of data, provided it is supplied in computable form. This opens it up to many problems, where the problem is reduced to finding a suitable representation for the data.

In this study, the model was trained on the data shown in Appendix B. The model was trained on 80% of the data, tested on 17% while the remaining 3% was used to validate it. Maximum tree depth was 9, while the population was limited to 100 for 65 generations. Best node count is depended on the data used, where it was 46 for the Cd dataset and 110 for the Zn. The crossover, mutation and direct reproduction probabilities were set to values of 0.85, 0.1 and 0.05 respectively. Performance evaluation was done using the common functions: Root Mean Square Error (RMSE) and Mean Absolute Percentage Error (MAPE). If the research problems are complex, other criteria such as performance index can be considered (Gandomi & Roke, 2015).

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N} \left| GP\_Yield_i - Actual\_Yield_i \right|^2}{N}} \quad (3)$$

$$MAPE\ (\%) = \frac{1}{n} \sum_i \left| \frac{GP\_Yield_i - Actual\_Yield_i}{Actual\_Yield_i} \right| \times 100 \quad (4)$$

where GP_Yield is the value predicted of ith data sample by the GP model, $Actual\_Yield_i$ is the actual value of ith data sample and N is the number of the training samples

5. Results and Discussions

5.1 Statistical modelling using linear regression and GP

The experimental output is dependent on the four correlated parameters: Solid/Liquid ratio (x_1), concentration of H_2SO_4 (x_2), Mass of $Na_2S_2O_5$ (x_3) and the Retention time of the mixture (x_4). The correlation matrix is given in Table 2. From Table 2, it can be seen that each parameter has some degree of correlation with each other. The linear modelling was unlikely to be successful. Multiple linear regression models were formulated from the data, and the results justify the findings of the correlation matrix. R Square (R^2) values (Table 3) for both outputs were found to be lower, indicating that a linear regression model is not accurate in prediction of outputs (Zn and Cd).

Table 2: Correlation matrix for input parameters and outputs

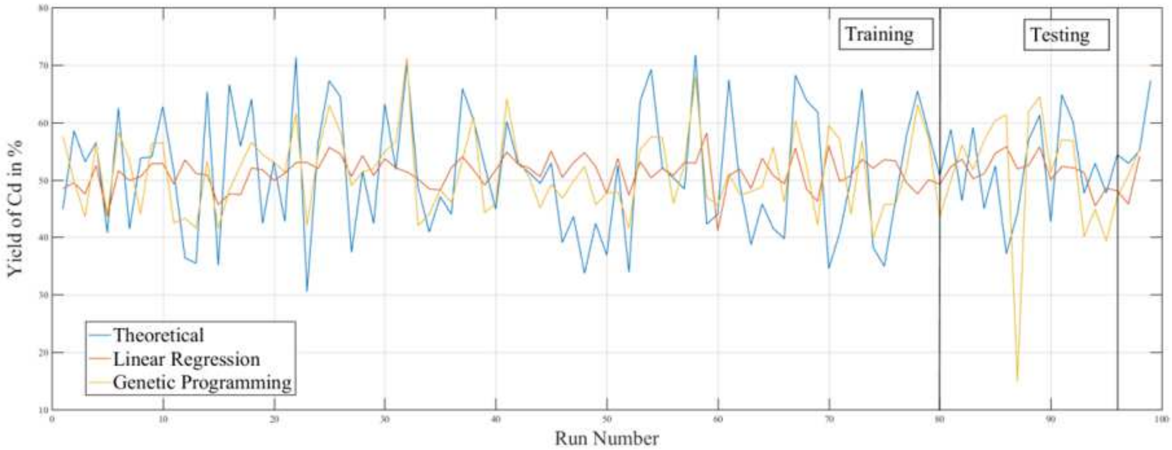
	Solid/Liquid ratio	H_2SO_4 conc.	$Na_2S_2O_5$ mass	Retention time	Y predicted for Zn	Y predicted for Cd
Solid/Liquid ratio	1					
H_2SO_4 conc.	0.02453313	1				
$Na_2S_2O_5$ mass	-0.045804544	-0.133175564	1			
Retention time	0.075367466	-0.020720113	0.076900659	1		
Y predicted - Zn	-0.252469158	0.153980331	-0.127701456	0.021473818	1	
Y predicted - Cd	-0.281479146	-0.134363065	0.118855452	0.158002348	0.158251496	1

Table 3: Results of Regression for Zn and Cd

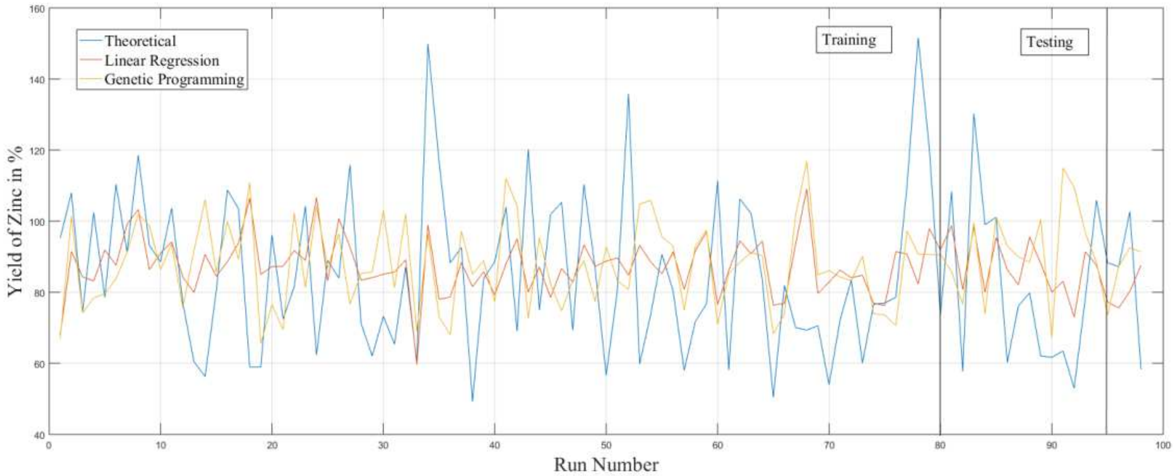
Regression Statistics		
Metrics	*Zn*	*Cd*
Multiple R	0.326388131	0.364153608
R^2	0.106529212	0.13260785
Adjusted R Square	0.068100361	0.095300661
Standard Error	7.761167586	7.384879928
Observations	98	98

In this perspective, the two GP models were constructed for the Zn and Cd yield respectively. Settings of GP was kept based on trial-and-error approach. The maximum number of genes was set to 6, and the node functions used were: TIMES, PLUS, MINUS, RDIVIDE, PLOG, SINE, COSINE, TAN, PLOG ($\log_e|x|$), PSQROOT ($\sqrt{|x|}$) and EXP. Tree depth was limited to 9. There were no limits on the number of nodes. 80% of the data was used for training, 17% for the testing and 3% for validation of the model. The best GP models are selected based on the minimum training error among all the runs.

The results as shown in Figure 3 and Table 4 shows that the GP models (Equations A1 and A2 given in Appendix C) performed better than other models such as linear regression and theoretical. This implies that the GP based metal yield models manages to closely capture dynamic involved in bioleaching process for active yields of metals.



(a)



(b)

Figure 3. Comparative Analysis of Theoretical, Linear regression and GP for (a) Cd and (b) Zn yields in %

Table 4: Performances of GP for Zn and Cd

Performance metrics	**Zn**	**Cd**
best training RMSE	8.69e-01	9.17e+00
best training MAPE	5.37e-01	1.65e+01
best test RMSE	1.42e+01	9.48e+00
best test MAPE	1.47e+01	1.55e+01

5.2 Effects of the inputs on Zn and Cd yield (%) (3-D analysis)

This section discusses the details on two dimensions (2-D) and three dimensions (3-D) analysis for evaluating the effect of inputs (Solid/Liquid ratio, concentration of H_2SO_4, mass of $Na_2S_2O_5$ and retention time) on the metals yields (%). 2-D analysis is performed by varying one given input while keeping other inputs at its mean value. 3-D analysis is performed by varying two inputs, while keeping others at its mean value.

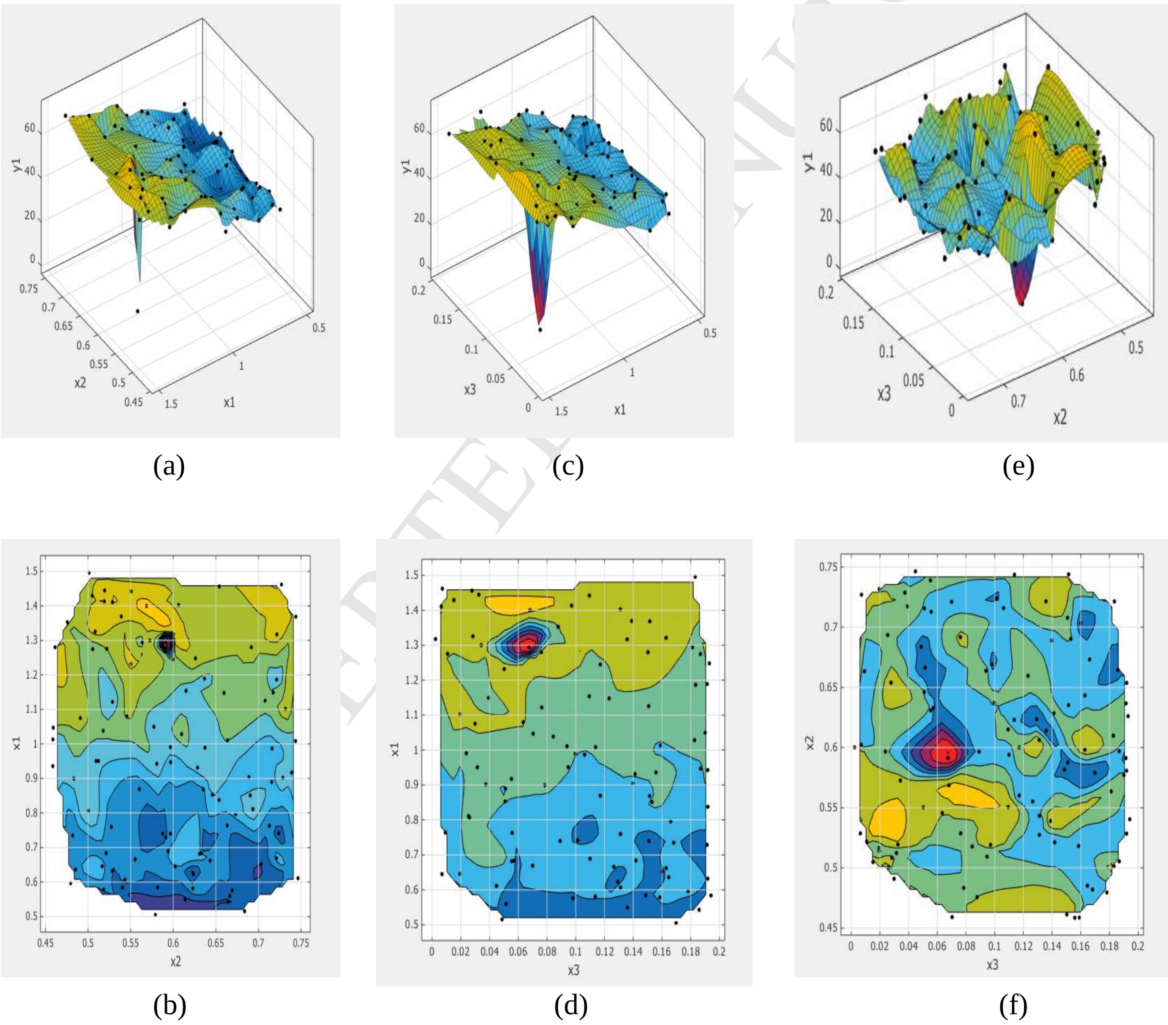


Figure 4. 3-D analysis investigating the effect of inputs on the yield of Cadmium. (a) Effect of the concentration of H_2SO_4 (x_1) and $Na_2S_2O_5$ (x_2) on yield of Cadmium, (b)Contour map of (a), (c) Effect of concentration of $Na_2S_2O_5$ (x_2) and Solid/Liquid ratio (x_3) on yield of Cadmium, (d) Contour map of (c), (e)Effect of the concentration of H_2SO_4 (x_1) and Solid/Liquid ratio (x_3) on yield of Cadmium, (f) Contour map of (e).

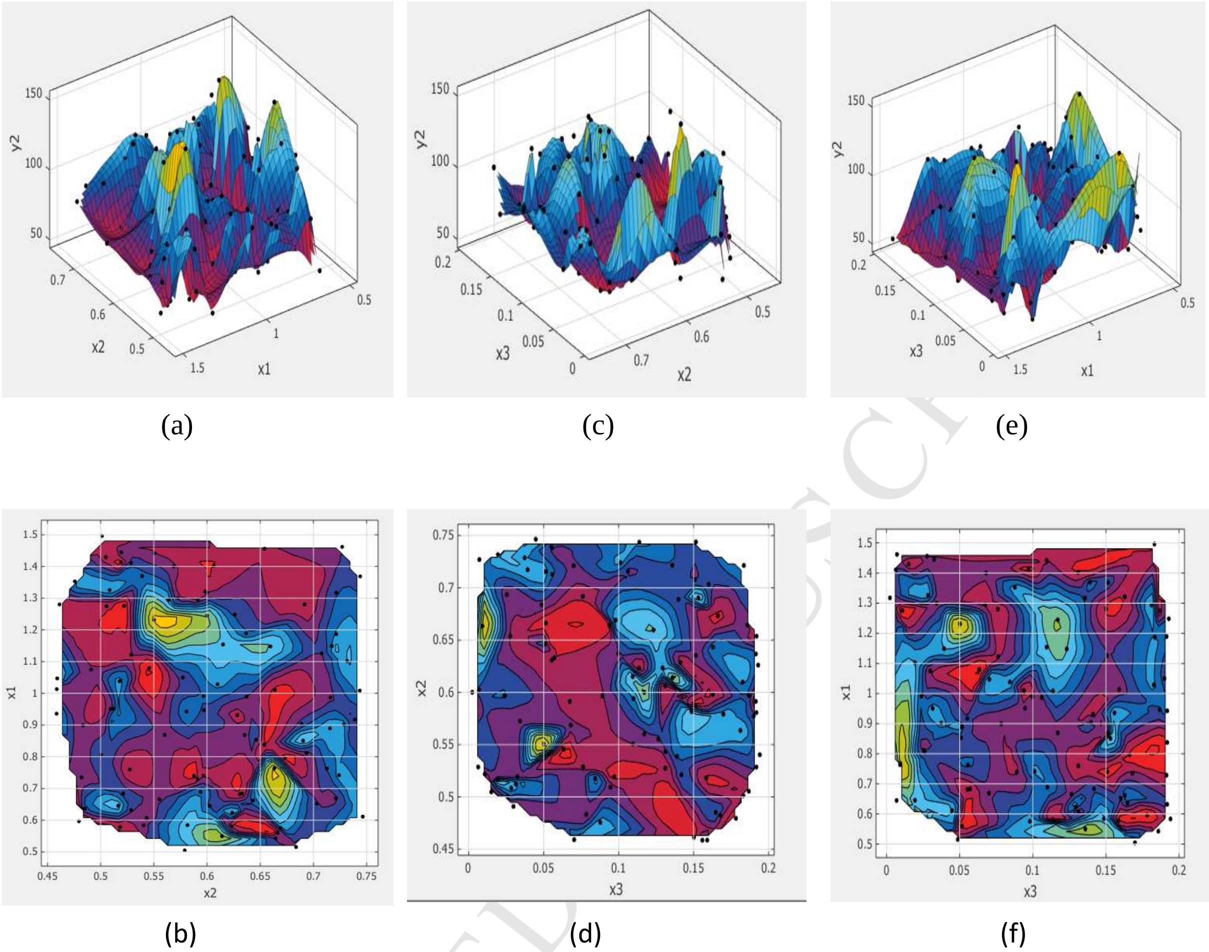


Figure 5. 3-D analysis investigating the effect of inputs on the yield of Zinc. (a) Effect of the concentration of H_2SO_4 (x_1) and $Na_2S_2O_5$ (x_2) on yield of Zinc, (b)Contour map of (a), (c) Effect of concentration of $Na_2S_2O_5$ (x_2) and Solid/Liquid ratio (x_3) on yield of Zinc, (d) Contour map of (c), (e)Effect of the concentration of H_2SO_4 (x_1) and Solid/Liquid ratio (x_3) on yield of Zinc, (f) Contour map of (e).

The above graphs indicate how variations in the input parameters affect the yield of metal. Common to both graphs is the trend of Solid/Liquid ratio; a lower value favours a better yield. This is especially evident in graph (e) for Zinc, where the highest value was observed at a lower value of S/L ratio. In graph (a) for Cadmium, a peak is observed in the same range, further confirming the trend. In the Cd graphs (c) and (e), peaks are seen near the optimal conditions for H_2SO_4 (1-1.5 M) and $Na_2S_2O_5$ (0.45 g/g).

5.3 Optimization for maximum yield (%) of Zn and Cd

For obtaining the maximum yield of Zn and Cd, the SA approach was used on the formulated GP models. SA is a heuristic minimalizing technique with roots in the annealing of metals. Optimising algorithms usually generate a random solution and compare it with the points in its neighbourhood. This can lead to the algorithm being fixed at local maxima/minima, which is undesirable. SA counters this by introducing the possibility of randomly moving to a 'worse' solution instead of a 'better' one. Details about this algorithm is given in (Kirkpatrick, 1983). It is essentially defined as follows:

1. Start at a high temperature value, with inputs defined. (Temperature is a controlling parameter which is iteratively scaled down)
2. Compute the cost of random input.
3. Compute the cost of random neighbour.
4. If cost of neighbour is lower, switch current point to neighbouring point. If the cost of neighbour is higher, switch basing on probability defined as:

$$P(switching) = e^{\frac{cost_{neighbour} - cost_{current}}{temperature}} \tag{5}$$

5. Scale temperature down, and rerun through steps 2-4 until arbitrary accuracy is reached.

This algorithm will act as a minimiser. Step 4 is modified to switch when cost of neighbour is higher, otherwise switch probabilistically as defined. This acts as a suitable maximising algorithm. Despite the stochastic nature of the algorithm, it is very effective.

In the run, the GP generated model for Zinc was used as the cost function to be optimised, temperature was set to $1.00*10^6$, alpha value as 0.88. SA algorithm was run 10 times, with 1000 iterations before the algorithm terminated each time. The optimal conditions obtained through this method are: Solid/Liquid ratio set to 11.7%, 0.86 M H_2SO_4, 0.56 g/g of $Na_2S_2O_5$ and 45 minutes of retention time. The obtained conditions comply well with experimental runs as mentioned in (Tanong et al., 2017). The computation of optimal points using SA aided GP programming is very effective in this case. The resilience of the algorithm to noise is also very good. The optimal conditions for the leaching of metals have been found and verified. This enables the further experimentation and development of this process. This also makes similar optimisation of other processes needed for battery recycling possible. This study focusses on the deterministic optimization. Uncertainty is inevitable in the chemical removal process (Zhang and Lam, 2015). Risk and reliability analysis can be investigated (Zhang et al., 2017a; 2017b). Intelligent disassembly problem (Yun et al., 2018) can be also solved using the present framework.

6. CONCLUSIONS

The present work proposes the comprehensive study to optimise the chemical metal leaching of valuable metals from a mix of spent batteries. The optimisation of the chemical metal leaching process has been carried out using combined using Combined genetic programming and simulated annealing Approach. Experiments were conducted to validate this approach. The optimal conditions obtained are: Solid/Liquid ratio = 11.7%, molarity of H_2SO_4 = 0.86 M, g/g of $Na_2S_2O_5$ = 0.56 g/g and 45 minutes of retention time for maximization of Zn and Cd from spend batteries. The obtained conditions comply well with experimental runs. This enables the further experimentation and development of this process. This also makes similar optimization of other processes needed for battery recycling possible. This technique for metal leaching can be recommended to help facilitate the recycling of batteries on a large scale. In this study, we chose an option to maximize the recovery of Zn and Cd. Researchers can also choose other metals. Since this study has only four input variables, the established GP models are acceptable. In the follow-up research, new models can be considered to accommodate higher dimensional problems. Future works shall emphasize on the incorporation of risk and reliability analysis on the chemical removal process having uncertainty and compare the performance to those conducted in the present study. The current framework can also be applied to solve intelligent disassembly problem of battery packs for electric vehicles.

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APPENDICES

APPENDIX A

```
%{
   this program generates data for experiment number 2.
   experiment 2 focuses on the effect of these parameters:
      1. solid/liquid ratio
      2. concentration of H2SO4
      3. mass of N2S2O5
      4.Retention time
%}

sl_ratio = rand(100,1)*0.2
conc_h2so4 = 0.5 + (1.5 - 0.5).*rand(100,1)
mass_na2s2o5 = 0.45 + (0.75 - 0.45).*rand (100,1)
retention_time = ceil (rand (100,1)*30 + 15)



% these functions return the yield of the respective metal as percentage value.

function [ y ] = CdRemovalYield ( x1, x2, x3, x4 )
% gives the yield percentage of Cadmium basing on the factors in the second experiment
 y = 36.9 - 25.4*x1 + 33.7*x2 - 5.27*x3 - 0.40*x4


function [ y ] = CoRemovalYield ( x1, x2, x3, x4 )
% gives the Cobalt removal yield from the second experiment
y = 37.7 - 21.1*x1 + 30.0*x2 - 2.05*x3 + 0.83*x4

function [ y ]= MnRemovalYield( x1, x2, x3, x4 )
% gives the yield percentage of Manganese basing on the factors of the second experiment
y = 68.2 - 15.7*x1 + 24.1*x2 - 0.47*x3 -0.50*x4 ...
   -0.56*(x1.*x3) - 0.04*(x1.*x4) -0.76*(x2.*x3) -1.02*(x2.*x4)...
   -2.19*(x3.*x4) - 1.08*(x1.*x1) - 10.3*(x2.*x2) ;

function [ y ] = NiRemovalYield( x1, x2, x3, x4 )
% gives the yield percentage of Nickel basing on the factors of the second experiment
y = 13.9 - 22.3*x1 + 28.3*x2 - 3.55*x3 + 2.21*x4 ...
   -4.38*(x1.*x4) -6.06*(x2.*x3) + 12.3*(x1.*x1) + 10.2*(x2.*x2) ...
   + 5.15*(x3.*x3) + 6.18*(x4.*x4) ;

function [ y ] = ZnRemovalYield( x1, x2, x3, x4 )
% gives the yield percentage of Zinc basing on the factors of the second experiment
y = 57.4 - 18.8*x1 + 29.8*x2 - 4,05*x3 - 0.90*x4 ...
   - 3.94*(x1.*x3) + 1.70*(x2.*x3) - 0.46*(x2.*x4) - 2.38*(x3.*x4) ...
   - 8.47*(x2.*x2);
```

APPENDIX B

The following table lists the noised input parameters and corresponding outputs.

Input parameters with noise				Recomputed output parameters				
Solid/Liquid ratio	H_2SO_4 conc.	$Na_2S_2O_5$ mass	Retention time	Cd Yield	Co Yield	Mn Yield	Ni Yield	Zn Yield
0.129	1.486	0.109	20	75.121	95.929	33.851	2579.407	95.252
0.109	1.900	0.418	23	86.775	110.649	-3.002	3405.929	107.987
0.218	0.843	0.749	18	48.626	71.795	22.662	2062.070	74.422
0.080	1.695	1.154	28	74.707	107.735	-57.142	4977.523	102.406
0.357	1.071	0.371	23	52.748	80.607	20.272	3330.526	78.585
0.081	1.963	1.194	24	85.112	112.361	-50.761	3700.174	110.383
0.169	1.376	0.808	28	63.494	96.974	-25.123	4948.675	91.204
0.085	2.240	1.306	19	95.730	116.184	-41.032	2378.939	118.539
0.067	1.375	0.697	30	65.865	101.010	-23.244	5684.370	93.119
0.092	1.238	0.342	37	59.695	102.920	-12.783	8586.772	88.575
0.101	1.751	0.048	23	83.875	107.077	22.054	3401.768	103.668
0.114	0.861	0.629	39	44.110	92.210	-28.897	9506.463	76.919
0.124	0.315	1.000	18	31.901	57.428	17.745	2053.255	60.459
0.161	0.198	0.476	27	26.162	61.667	22.396	4561.330	56.264
0.234	1.056	0.236	16	58.880	77.222	44.435	1649.550	80.454
0.174	1.968	0.470	22	87.527	110.367	-5.973	3122.400	108.778
0.134	1.764	0.130	16	85.868	100.814	34.857	1699.091	103.456
0.072	0.231	0.340	21	32.667	59.847	40.693	2783.539	58.933
0.093	0.246	0.614	20	31.590	58.457	29.621	2526.390	58.980
0.133	1.517	0.692	24	71.402	98.907	-7.827	3669.964	96.107
0.095	0.701	0.527	21	46.915	73.060	28.125	2797.058	72.490
0.031	0.964	0.881	20	55.953	80.757	12.030	2559.198	81.541
0.057	1.742	0.619	33	77.683	114.866	-43.004	6880.838	104.227
0.265	0.467	1.557	43	20.507	78.622	-117.629	11498.739	62.340
0.060	1.231	0.821	44	54.953	108.216	-76.310	12107.570	88.970
0.018	1.038	1.449	22	54.988	83.751	-24.128	3088.408	83.995
0.092	2.153	0.310	30	93.469	124.598	-31.094	5731.669	115.814
0.048	0.626	0.987	27	40.787	75.865	-11.646	4591.578	71.164
0.184	0.407	0.560	31	30.590	70.607	6.294	6004.229	62.066
0.040	0.690	0.951	23	44.943	74.711	2.710	3350.798	73.224
0.069	0.446	0.769	18	38.944	63.004	27.607	2062.010	65.410
0.110	1.199	0.487	29	60.337	94.412	-1.159	5303.610	87.054
0.119	0.304	0.012	21	35.646	61.704	54.993	2781.701	60.214
0.031	3.258	0.107	16	138.969	147.868	-28.367	1826.607	149.926
0.275	2.300	0.659	44	86.357	136.071	-126.013	12127.200	116.774
0.119	1.245	1.227	28	58.175	93.270	-46.409	4949.050	88.270
0.096	1.375	0.543	44	60.361	112.350	-56.685	12108.474	92.588
0.224	0.001	0.846	38	11.586	62.810	-25.645	8980.915	49.218
0.150	1.116	0.972	16	59.187	79.312	18.151	1656.906	83.846
0.110	1.250	0.033	36	61.668	102.702	13.739	8134.095	88.593
0.028	1.714	0.842	37	74.725	117.524	-74.178	8621.401	103.962

0.106	0.593	0.238	35	38.914	81.794	19.931	7662.146	69.057
0.099	2.304	1.246	32	92.661	128.735	-114.048	6502.443	120.198
0.124	0.803	0.606	22	48.814	76.187	19.950	3065.237	74.992
0.061	1.664	1.275	39	69.115	116.097	-118.172	9554.430	101.849
0.075	1.789	0.446	22	84.129	107.131	3.591	3122.648	105.301
0.091	0.591	0.914	27	38.915	74.069	-7.398	4584.248	69.323
0.028	1.928	0.437	42	82.067	128.919	-68.711	11089.308	110.332
0.145	1.215	0.508	41	55.074	104.069	-37.912	10509.233	86.871
0.254	0.267	0.022	28	28.127	63.544	46.534	4892.996	56.582
0.029	0.927	0.751	25	53.445	84.108	3.059	3958.895	80.479
0.218	2.904	0.554	34	112.694	147.293	-112.972	7354.361	135.831
0.146	0.308	0.160	43	25.525	79.220	21.842	11514.216	59.832
0.236	0.835	0.333	43	40.064	92.756	-12.944	11515.057	73.826
0.118	1.324	0.726	33	61.511	100.846	-34.652	6847.061	90.651
0.104	0.973	0.230	25	55.838	84.976	29.937	3953.409	80.443
0.187	0.273	0.949	41	19.938	74.020	-47.166	10463.898	58.012
0.072	0.662	0.414	29	43.590	79.254	17.647	5285.578	71.766
0.012	0.789	1.354	43	38.854	94.036	-104.504	11560.116	76.692
0.342	2.163	0.085	16	94.252	108.473	19.973	1708.727	111.420
0.192	0.279	0.589	31	25.930	66.549	6.054	5998.885	58.112
0.088	1.828	0.115	37	80.880	121.174	-20.765	8624.208	106.237
0.124	1.715	0.103	21	82.604	103.752	25.037	2848.876	102.174
0.007	1.022	0.394	26	58.705	89.000	18.916	4284.668	83.739
0.163	0.004	0.898	21	19.757	49.965	13.179	2768.457	50.450
0.208	1.089	1.179	25	52.089	84.303	-27.792	3942.826	81.932
0.004	0.560	0.891	28	39.791	75.843	-7.024	4937.326	70.026
0.255	0.696	0.422	32	38.861	78.900	6.835	6394.624	69.351
0.274	0.749	0.925	17	43.505	66.599	18.904	1835.763	70.564
0.031	0.039	0.865	32	20.068	63.004	-9.772	6409.579	53.982
0.172	0.744	0.660	24	44.515	74.949	11.874	3628.693	72.329
0.127	1.089	0.901	23	56.436	84.942	-3.568	3356.498	83.473
0.059	0.262	0.878	23	30.412	61.612	10.351	3334.358	60.103
0.023	0.793	0.001	20	55.059	77.624	54.240	2556.489	76.619
0.164	0.895	1.015	44	39.934	95.517	-82.561	12070.332	76.976
0.126	0.923	0.288	44	45.701	98.674	-12.120	12081.383	78.550
0.191	2.017	0.210	40	82.923	126.957	-49.582	10048.589	109.923
0.219	3.432	0.817	37	127.909	165.091	-191.090	8717.180	151.572
0.064	2.295	0.479	21	101.675	121.633	-14.550	2889.876	120.574
0.191	0.801	0.519	26	45.913	78.219	13.280	4249.760	73.681
0.031	1.863	1.020	21	85.122	108.276	-22.422	2860.472	108.336
0.037	0.168	1.151	16	29.146	52.871	19.555	1634.550	57.703
0.067	2.621	0.490	25	110.947	134.666	-47.926	4058.834	130.251
0.165	1.635	0.860	36	68.874	111.383	-70.176	8139.116	99.019
0.004	1.603	0.495	34	74.625	112.923	-29.991	7298.754	101.105
0.022	0.243	0.727	32	27.905	69.600	-2.270	6415.985	60.231
0.128	0.845	0.866	29	45.939	82.625	-16.490	5283.943	76.157
0.038	0.910	0.263	24	55.627	83.589	32.559	3653.979	79.813
0.010	0.298	0.654	31	30.839	70.817	4.484	6027.783	62.093
0.270	0.449	0.913	38	25.143	75.126	-41.082	8984.998	61.691

0.177	0.450	0.755	38	28.382	77.449	-26.031	9001.798	63.472
0.208	0.117	1.406	33	14.965	61.342	-55.705	6790.325	52.989
0.159	0.934	0.190	38	48.125	93.507	7.661	9025.803	78.238
0.136	0.134	0.228	35	22.764	67.436	29.002	7641.358	54.839
0.227	1.906	0.303	19	86.151	105.224	13.240	2350.387	105.918
0.252	1.330	1.009	31	57.608	95.950	-50.035	6031.528	88.303
0.246	1.289	1.118	26	57.786	90.456	-34.638	4263.425	87.176
0.087	0.989	0.533	18	58.018	79.387	31.692	2081.643	81.241
0.266	1.821	1.000	20	78.217	101.247	-19.481	2577.601	102.646
0.017	0.175	0.647	21	30.563	58.700	27.442	2788.232	58.299

APPENDIX C

The model equations for Zinc yield in % is:

$$y_{Zn} = 97.867 + (132.4566) * \left(\frac{x1}{(x3) * \left(\left(\frac{x4}{(-0.731135)}\right) + (\sin((-29.969512)))\right)}\right) + (-9.4264) * ((x3) + (x3)) + (0.45898) * \left(\left(\frac{\sin(\exp(((x3) + ((9.440839))) + (\cos(x1))))}{x3}\right) *\right.$$

$$\left.\left(\left(\tan(((x4) * (x1)) * (\cos(x1)))\right) - \left(\left(\frac{\cos(x3)}{\frac{x4}{x1}}\right) * \left(((x4) + (x1)) * (plog(x3))\right)\right)\right)\right) + (-0.00097061) * \left(\left(((x3) + (x4)) + (plog(x2))\right) -\right.$$

$$\left.\left(\left((x3) * \left(\left(\frac{x4}{x3}\right) + \left(\frac{x4}{x1}\right)\right)\right) * (\tan(x3))\right)\right) + (8.3473) * \left(\frac{\frac{\frac{\sin(\sin(x1))}{\tan\left(\frac{(x1)+(x1)}{(x3)*(x2)}\right)}}{\exp\left(\sin\left(\left(\frac{x1}{x3}\right) + (\tan((2.478800)))\right)\right)}}{\tan\left(\left(\frac{\exp(\exp((-1.608622)))}{(plog(x1)) * (\sin((-21.211251)))}\right) + \left(plog\left(\left(\frac{x4}{(-0.731135)}\right) + (\sin((-29.969512)))\right)\right)\right)}\right) + (0.18178) *$$

$$\left(\frac{x1}{\frac{\frac{\sin(\sin(\tan(x1)))}{\tan\left(\frac{(x1)+(x1)}{(x3)*(x2)}\right)}}{\exp\left(\sin\left(\left(\frac{x1}{x3}\right) + (\tan((2.478800)))\right)\right)}}\right) \quad \text{(A1)}$$

and that for Cadmium yield in % is:

$$y_{Cd} =$$

$$41.7032 + (-4.3431) *$$

$$\left(\sin\left(\left(\left((square((-23.617098))) * (x2)\right) - \left((psqroot(x4)) - (\sin((5.202223)))\right)\right) - \left((psqroot(x4)) - (\sin((5.202223)))\right)\right)\right) + (0.0024014) *$$

$$\left((square(x2)) + \left(\left((\cos(square(x4))) + \left(\frac{x4}{\cos(square(x4))}\right)\right) - (x4)\right)\right) + (4.5706) * \left(\left((\cos(\exp(\cos(square(x4))))) * (\sin(x4))\right) - (\sin(x1))\right) +$$

$$(-2.9824) * \left(\sin\left((\sin(psqroot(x2))) * \left(\frac{x4}{\cos(square(x4))}\right)\right)\right) + (3.5601) *$$

$$\left((square(\sin(x4))) - \left((\cos(\exp(\cos(square(x4))))) - \left(\exp\left(psqroot(psqroot(x3))\right)\right)\right)\right) + (0.0054371) * \left(\left(\sin\left((\sin(psqroot(x2))) *\right.\right.\right.$$

$$\left.\left.\left.\left(\frac{x4}{\cos(square(x4))}\right)\right)\right) - \left((\sin(x4)) * \left((square(x4)) + (square(x2))\right)\right)\right) \quad \text{(A2)}$$

Problem of Maximum Extraction of Cadmium and Zinc from spent battery mix is undertaken

Combined Genetic Programming and Simulated Annealing approach is proposed

Genetic programming models fits the cadmium and zinc output very well

Simulated annealing optimizes the Genetic programming model to obtain the optimum inputs