Intelligent optimization of process conditions for maximum metal recovery from spent zinc-manganese batteries

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Abstract: By 2025, 2 million metric tons of batteries must be recycled. Among these batteries, the spent Zinc-Manganese batteries poses a serious threat to environment due to toxic heavy metals. These metals are toxic but at same time vital for various industrial applications. These metals are generally recovered by physical-chemical process which are highly energy intensive and polluting. An eco-friendly recycling process has to be explored to tackle such issue. The bioleaching is one such eco-friendly recycling method. The objective of this work is to optimize the process parameters of bioleaching method, so as to make this process commercially viable. The optimization of this process is done through statistical based automated neural network intelligent optimization approach. The formulated models were inline with the complex behaviour of bioleaching process. The training and validation performance of the models were near to 1. The parametric, global sensitivity and interaction analysis was undertaken for understanding the relationship between different parameters and its affect on the metal yield. The optimum values of process parameters were determined for maximizing the metal yield.

Keywords: Recycling methods; bioleaching process; Intelligent Optimization methods;
because of its compact design and high energy capacity[4-5]. The cells used in battery packs are Li, Ni-Cd and Zn-Mn [5].

For the waste treatment of the spent zinc-manganese batteries (ZMB), the removal and recovery of Zn and Mn is very essential [12-15]. Spent ZMBs are the major chunk of the overall spent batteries, because of its household application. The Zn and Mn are roughly in range of ~14-29% and ~27-44% respectively in the used batteries[8-9]. To recycle this batteries there are many commercial process available, some of the examples of this commercially viable process are pyrometallurgy and hydrometallurgy[9], these process come under physical-chemical domain. This category of processes are very energy abusive, polluting and the working conditions associated with this process are risky for the on-site workers. To tackle this problem, an eco-friendly avenue has been explored. One of such promising method is bioleaching. It is less energy demanding, and has non-existent pollution[15-18]. This process is not commercially viable compared to the available process in use. Bioleaching process has less yield compared to hydrometallurgy. For this green technology method, the metal recovery is dependent on substrate concentration (SC), initiating pH level, incubation temperature(T), and pulp density (PD) of the mix [18-19]. The leaching period is also a very important parameter from economic point of view, direct methods are having less leaching period compared to that of indirect process [14-19]. But, for that case the pulp density is severely affected due to the toxicity of the mix. The range of ~1-10% is desirable PD range, near higher end of this range there is drop in the size of reactor by ~10%[18]. This is beneficial for its commercial roll-out. For the hydrometallurgy related process, the PD is observed to be higher than ~10% for the spent batteries[9]. For bioleaching method to become significant in market, it should have PD higher than ~10%. The previous optimization methodology employed are basic method for identifying the optimum settings [15-19]. Here, only one parameter is dealt at a time. This is not helpful in modelling complex and varied relationship between different parameters of the process[20]. Considering the complex and varied nature of these process some previous study have undertaken multi-variable modelling such as the response surface methodology (RSM) [19]. Again, in this method the modelling of relationship is basic for the complex bioleaching process. For, the RSM to be employed accurately the underlying process interaction should be known in-depth for modelling. Considering the complex process behaviour, modelling methods based on artificial intelligence (AI) looks very promising. One of such method called as statistical automated neural networks (SANN) can be used for modelling complex bioleaching process. Modelling using SANN can then be further used for the optimization problem, which then can give the best optimum setting for high recovery yield.

These work proposes a SANN approach for modelling bioleaching process. This methodology is used for optimizing the process parameters for maximum metal recovery from spent ZMBs. Here, PD is taken in range of ~8-12% for its commercial viability. A multi-variable analysis for all the process parameters is undertaken for the given range of PD.

2. Research problem

For the bioleaching process to become a major stakeholder in recycling industry, it has to become a commercially viable option. The operational input parameters of bioleaching process has to be optimized such that it gives high yield. Before, going into optimization a accurate model of the complex bioleaching process has to be formulated. The interaction between different operational parameters and yield of metal has to be understood. The PD is one of the most influencing parameter, a lower PD can severely affect the metal recovery yield. Also a lower PD significantly increases the size of reactor, this makes it commercially non-viable. The pH of solution mix is also a important factor, if the mix is high on toxicity then it will severely affect the culture of the mix which will reduce the yield. The value of T should be in a optimum range where microorganism have high productivity. The main objective is to maximize the metal recovery from the spent batteries. The Table 1 gives the set of parameters. The details of methodology employed is given in next part.
Table 1. Parameters under study for modelling of bioleaching process.

| Input parameter | Output parameter |
|-----------------|------------------|
| SC (g/L)        | Zn metal yield   |
| T (°C)          |                  |
| PD (%)          | Mn metal yield   |
| pH              |                  |

3. Methodology: SANN based modelling

Bioleaching process is modeled using SANN. Here, the modelling architecture is same as that of artificial neural network. The STATISTICA software is used for implementing these algorithm. Here input layers consist of all the input parameters given in table 1. The objective variable consist of the amount of metal recovered. 29 set of data values are used for the input and output parameters[20]. The data is divided in three sets as training (70%), testing (15%) and validation (15%). The best networks are found using automated process, where set of values are tried and tested for a range, and the best fit network is selected.

In the case of the Zn recovery, Multilayer Perceptron (MLP) model is used where hidden units are in range of 4-6, total of 1000 different networks are trained and of that 5 best networks are finally evaluated. The activation functions are set tanh and logistic for hidden and output layer respectively, while weight decay is in range of 0.0001 to 0.001.

Radial Basis Function (RBF) model is used for modelling the case of Mn recovery. Eqn. 1 is a RBF. Here, hidden units are in range of 4-16, and all the other constraints are same as that of Zn recovery model. The best regression and good training fit model are selected for the analysis.

\[ \varphi(x) = e^{-\beta||x-\mu||^2} \]  

4. Results and discussion

The results and related discussion is undertaken in the following part. The predicted values from SANN models are inline with the observed values. The range of values predicted are well under experimental bounds [20], abolishing the chances of over-fitting problem. Out of the 1000 constructed network the best selected network are MLP 4-5-1 and RBF 4-16-1 for Zn and Mn respectively. Here, the Mn is in range of 9.2-12.50 g/L and Zn is in range of 4.7-10g/L. The validation error is 0.052 and 0.057 for Zn and Mn recovery model respectively. Which is a stable result for such complex behaviour process. Also the training performance is near to 1. Which may arise the issue of over-fitting, but that possibility is ruled-out as the predicted values are in experimental range. table 2 gives the model evaluation report for the both models.

Table 2. Selected Models for Zn and Mn.

| Metal (Response) | Net. name | Training perf. | Test perf. | Validation perf | Training error | Test error | Validation error | Training algorithm | Hidden activation | Output activation |
|------------------|-----------|----------------|------------|----------------|----------------|-----------|------------------|-------------------|------------------|------------------|
| Zn               | MLP 4-5-1 | 0.984017       | 0.931815   | 0.998092       | 0.059620       | 0.315376  | 0.052295         | BFGS 51           | Tanh             | Logistic         |
| Mn               | RBF 4-16-1| 0.997045       | 0.988236   | 0.99909        | 0.001713       | 0.011013  | 0.057928         | RBFT              | Gaussian          | Identity         |
The $R^2$ values for both the models are 0.941 and 0.964 respectively. These confirms the good fit of the model. Figure 1 gives the experimental and modeled values curves, and it can be seen that the deviation of modeled curve is very less. These confirms that SANN modeling is able to model the complex behaviour of bioleaching process. This confirms the stability of model, which is important to confirm before solving for optimization problems. If the deviations are too large than model will give wrong optimum value.

The equation for linear regression fitting curve for these models are given below as Eqn. 2 and 3,

$$\text{predicted Zn} = 0.935(\text{Observed Zn}) + 0.4119$$

$$\text{predicted Mn} = 0.945(\text{Observed Mn}) + 0.5299$$

To evaluate the SANN model the parametric and interaction analysis is undertaken. These set of analysis gives insight into the relationship among input parameters and how they affect the output yield. Firstly, coming to the results of parametric analysis. Here, all the four parameters are individually varied and the corresponding changes in metal recovery are evaluated. The parameters not under study are fixed at their mean values. For the case of Zn metal recovery, the highest metal recovery of 9.7 g/L is obtained for the PD of 9.8 %, if the PD value is more or less than this value than yield strength decreases. The SC parameter is having inverse effect on yield strength. The yield of Zn is highest at SC of 24 g/L. From these analysis it is found that T value of 30°C is best for microorganism productivity for this yield value is 10 g/L. For high productivity pH should be around ~1.9. Now, for the case of Mn metal recovery. PD is having inverse relation with yield, at PD of 8 % highest yield value of 12.5 % is observed. SC should be 24 g/L to get good yield. For this case microorganism give best productivity at T value of ~36°C. While for the stable mix pH value is around ~1.8.

Before proceeding with interaction analysis its important to have the global sensitivity analysis done. The global sensitivity analysis specifies the most affecting input parameter of the process. These analysis is performed on the models so as to determine the most influencing parameter to get higher yield.
Table 3. SANN model global sensitivity test for input parameters.

| Metal (Response) | PD %  | T °C  | pH   | SC g/L |
|------------------|-------|-------|------|--------|
| Zn               | 11    | 5.7   | 10.5 | 2.7    |
| Mn               | 21    | 6.8   | 4.5  | 2      |

The global sensitivity analysis is given in table 3 for both Zn and Mn metal recovery. It can be observed that PD is the most influencing parameter. This results are in line with experimental result as their also PD is the most influencing parameter. This also proves the consistence of model with actual behaviour of bioleaching process. It confirms the accuracy of model in predicting the yield strength for given input parameters values. After PD, the T and pH are the most influencing parameter for Mn and Zn yield respectively. The interaction analysis is performed considering the most influencing parameters obtained from table 3. The results are obtained for optimized set of values to give higher metal recovery yield. These optimum parameter values are given in table 4. For these optimum value the yield of Zn metal is 50 % and yield of Mn metal is 50.5 %.

Table 4. Optimum parameter value for high metal yield.

| Input parameter for Zn recovery | Optimized value | Input parameter for Mn recovery | Optimized value |
|--------------------------------|----------------|--------------------------------|----------------|
| SC                             | 32 g/L         | SC                             | 32 g/L         |
| T                              | 30 °C          | T                              | 35 °C          |
| pH                             | 1.9-2          | pH                             | 2              |
| PD                             | 10 %           | PD                             | 8 %            |

5. Conclusion
The bioleaching process was considered for the optimization problem. The main objective was to increase the metal recovery yield of this environment friendly recycling process. The methods used for that was SANN intelligent optimization. The input parameter under study were SC, T, PD, and pH while the objective parameter were Zn an Mn yield. The AI methods such as SANN proved to be a efficient method for modelling of such complex reaction process. The results of global sensitivity analysis confirmed the stability of the model. Finally, a optimum value of parameters were obtained for which Zn yield is 50 % and Mn yield is 50.5 %. For the scope of future work the focus of authors would be to undertake the same problem with other sophisticated algorithm of AI like genetic algorithm and deep learning neural network [6-7, 10-11].

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