Stochastic approach based on Monte Carlo (MC) simulation used for Life Cycle Inventory (LCI) uncertainty analysis in Rare Earth Elements (REEs) recovery

Dariusz Sala[1*] and *Bogusław* Bieda[1]

[1]AGH University of Science and Technology in Kraków, Faculty of Management, 30-059 Kraków, Al. Mickiewicza 30, Poland

Abstract. According to the European Commission's Report on Critical Raw Materials and the Circular Economy, the raw materials, such as rare earths, have a high economic importance for the EU, and are essential for the production of a broad range of goods and applications used in everyday life, as well as they are crucial for a strong European industrial base. Uncertainty plays an important role in the real world used Life Cycle Assessment (LCA) approach. The validity of LCA depends strongly on the significance of the input data. Data uncertainty is often mentioned as a crucial limitation for a clear interpretation of LCA results. The stochastic modelling used for Monte Carlo (MC) analysis simulation was reported in order to assess uncertainty in life cycle inventory (LCI) of rare earth elements (REEs) recovery. The purpose of this study was REEs recovery from secondary sources analysed in the ENVIREE ERA-NET ERA-MIN-funded research project. The software Crystal Ball® (CB) program, associated with Microsoft® Excel, was used for the uncertainties analysis. Uncertainty of data can be expressed through a definition of probability distribution of those data. The output report provided by CB, after 10000 runs is reflected in the frequency charts and summary statistics. The analysed parameters were assigned with lognormal distribution. The uncertainty analysis offers a well-defined procedure for LCI studies, and provides the basis for defining the data needs for full LCA of the REEs beneficiation process. Results can improve current procedures in the REEs beneficiation process management and bring closer to industrial application through the involvement of end users.

1 Introduction

All of REEs, were finally identified in the 20th century [1]. According to European Commission's Report on Critical Raw Materials and the Circular Economy, the raw materials, such as rare earths, have a high economic importance for the EU, and are essential

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* Corresponding author: dsala@zarz.agh.edu.pl

for the production of a broad range of goods and applications used in everyday life as well as they are crucial for a strong European industrial base.

Uncertainty plays important role in the real world used Life Cycle Assessment (LCA) approach. The validity of LCA depends strongly on the significance of the input data. Data uncertainty is often mentioned as a crucial limitation for a clear interpretation of LCA results. The stochastic modelling used Monte Carlo (MC) analysis simulation was reported in order to assess uncertainty in life cycle inventory (LCI) of rare earth elements (REEs) recovery.

All of rare-earth elements (REEs), were finally identified in the 20th century. For more than two decades, at least 95% of annual global supply of the REEs has been provided by Chinese rare-earth producers [2]. Mining companies are now actively seeking for new exploitable rare-earth deposits, white old mines are being reopened e.g. The Mountain Pass mine in California [3]. The purpose of this study was REEs recovery from secondary sources analyzed in the ENVIREE ERA-NET ERA-MIN-funded research project [4]. The software Crystal Ball® from Oracle (CB), associated with Microsoft® Excel, was used for the uncertainties analysis with the MC method.

2 LCI Data Quality

Inventory data used in the study have been obtained from the following sources:
- Primary data used in this study are based on the elements determined from the chemical analyses done by instrumental neutron activation analyses site-specific measured or calculated data, and values based on literature.
- Secondary data were mainly derived from the ecoinvent process rare earth concentrate, 70% REO, from bastnäsite, at beneficiation. In the present study, we discuss and model our LCI based on the proposed process for the beneficiation of REE in the flotation tailings from new Kankberg mine in Sweden [5], and Covas old tungsten mine, Portugal. Information about study area location was taken from [7].

Presented in this study the potential source of REEs is New Kankberg tailings from Sweden [7], Rosa et al. (2016). New Kankberg ore is currently mined at an annual capacity of around 400 k tonnes with underground mining. The mining started in 2012 and the gold grade is around 4 g/t with some additional value in tellurium. Around 31 k tonnes of tellurium was produced in 2014. The current expected life length is up to 2023. Kankberg is located in the Boliden Area. Information about study area location was taken from [7], Rosa et al. (2016). The Covas tailings represents 30 years (1954-1984) of mining focused in tungsten mineralization (mainly scheelite and minor wolframite) exploited by underground mining works [6]. After the regrinding stage, tailings were reprocessed using Mozley Gravity separator. This stage resulted in the production of a gravimetric concentrate and gravimetric tailing, and allows recovering 75% of REE (50% of tungsten). Final recovery for REE is 55% (feed basis) and 35% for tungsten (feed basis) [6].

After the flotation stage, the concentrate that contains a mix of phosphates (apatite and monazite) can be further enriched through magnetic separation thanks to the paramagnetic property of monazite (apatite is non-magnetic).
- Monazite which is a phosphate mineral commonly containing REEs, typically Lanthanum]oo(La), Cerium (Ce) and Neodymium (Nd).
- Magnetic separation leads to the production of a concentrate containing mainly, Ce, La and Nd [5].

In this study, other REEs were not considered interested for further investigation, because they were not separated.

3 Results and discussion

In this study, the process models for the beneficiation of REE is based on the one ton of New Kankberg flotation tailings input material so as to produce 10 kg of concentrate (after flotation, and magnetic separation) and 990 kg of residues. It is a phosphate containing rare earths mainly of the cerium group as well as that of thorium. Currently we investigate possibilities of extraction and mainly separation of Ce, La and Ne using magnetic separation regarding New Kankberg and Ce, Ne and W regarding Covas. Analysed REEs were fitted by lognormal distributions with the geometric standard deviation (GSD) σ_g equal to 1.13 obtained using the Oracle CB spreadsheet-based software, and powerful tool for performing MC simulation. The number of replications of a simulation affects the quality of results. The random number sets of 10 000 runs (replications) were chosen. It is worth noting that if the number of runs increases, the mean standard error decreases [8]. Moreover, the mean standard error can be used to construct confidence intervals as described in [8].

After 10 000 runs the results obtained by MC simulation for the Ce, La, Ne regarding New Kankberg have been presented in the form of frequency charts (histograms) and the statistics report presented in Table 1, while MC simulation results for the Ce, La, Ne, as well as W regarding Covas case study are given. The statistics reports have been presented in Table 1. The mean values of the Ce, La, Ne (New Kankberg case study) and the mean values of the Ce, La, Ne and W (Covas case study) forecast values with a 95% confidence interval around the mean values were situated between:

  – Ce [24.53 to 36.35 ppm] (see Fig. 1),
  – Ne [9.70 to 14.36 ppm] (see Fig. 2),
  – La [10.70 to 15.96 ppm] (see Fig. 3),
and
  – Ce [29.81 to 34.28 ppm] (see Fig. 4),
  – La [13.92 to 18.41 ppm] (see Fig. 5),
  – Nd [12.88 to 17.31 ppm] (see Fig. 6),
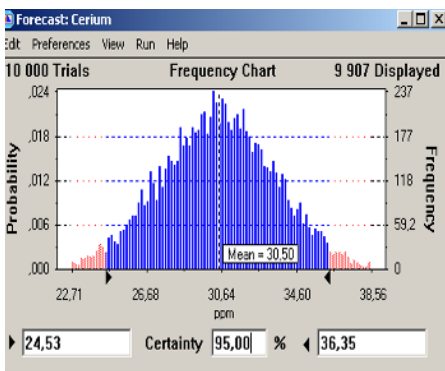 – W [1897.82 to 1901.23 ppm] (see Fig. 7).

Fig. 1. CB forecast chart: Ce after 10 000 trials (95% confidence interval). Certainty is 95.00% from 24.53 to 36.35 ppm (*source: own work*)
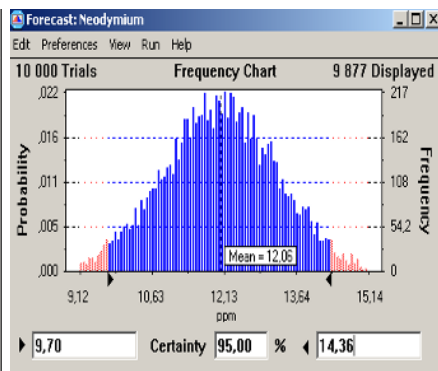
Fig. 2. CB forecast chart: Ne after 10 000 trials (95% confidence interval). Certainty is 95.00% from 9.70 to 14.36 ppm (*source: own work*)
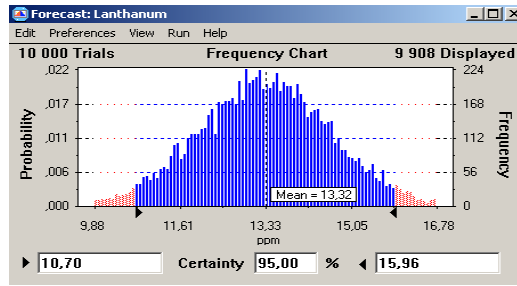
Fig. 3. CB forecast chart: La after 10 000 trials (95% confidence interval). Certainty is 95.00% from 10.70 to 15.96 ppm (*source: own work*)

Table 1. Percentiles report of outcomes from the simulation - New Kankberg case study

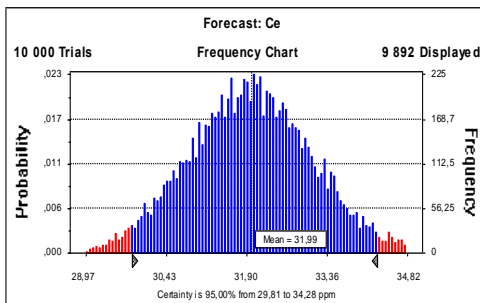| Percentile	Ce (ppm)	La (ppm)	Ne (ppm)
0%	19.58	8.10	6.94
10%	26.53	11.58	10.51
20%	27.91	12.19	11.06
30%	28.90	12.62	11.44
40%	29.75	12.99	11.76
50%	30.50	13.32	12.06
60%	31.27	13.66	12.36
70%	32.07	14.02	12.68
80%	33.07	14.44	13.07
90%	34.38	15.04	13.59
100%	42.75	18.12	16.51

(*Source: own work*)

Fig. 4. CB forecast chart: Ce after 10 000 trials (95% confidence interval). Certainty is 95.00% from 29.81 to 34.28 ppm (*source: own work*)
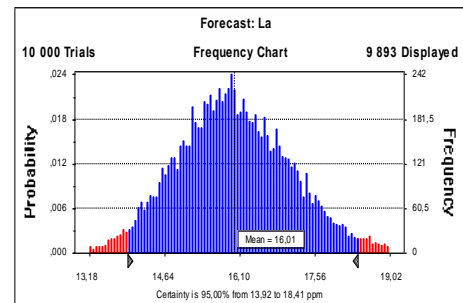

Fig. 5. CB forecast chart: La after 10 000 trials (95% confidence interval). Certainty is 95.00% from 13.92 to 18.41 ppm (*source: own work*)

The confidence interval range expresses 95% presented in the frequency chart highlighted with a darker colour marker. In other words, this means that 95% of the results are lying inside this range. Moreover, by setting the certainty values (e.g. 95%), the confidence intervals (minimum and maximum bounds) are set automatically by the

grabbers, and the corresponding numerical values are entered in the edit fields in the bottom part of the dialog boxes of the Forecast tab (see [8] and [9]).
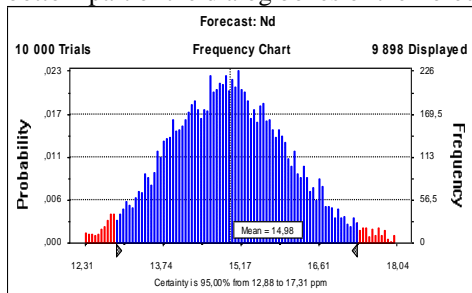

Fig. 6. CB forecast chart: Nd after 10 000 trials (95% confidence interval). Certainty is 95.00% from 12.88 to 17.31 ppm (*source: own work*)
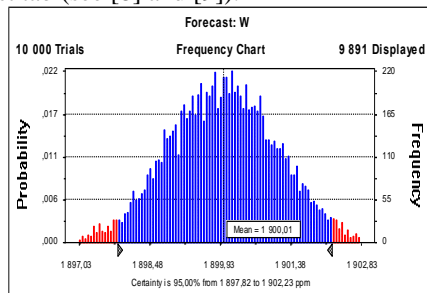

Fig. 7. CB forecast chart: W after 10 000 trials (95% confidence interval). Certainty is 95.00% from 1897.82 to 1901.23 ppm (*source: own work*)

Lognormal distribution is stable and no negative values are possible [10]. In this context, it should be pointed out that the lognormal probability distributions with the GSD equal to 1.13 were used to rare earth oxides in the ecoinvent background process. "Rare earth oxide production from bastnaesite" taken from the "Life Cycle Inventories of Chemicals Data v2.0 Ecoinvent report No. 8" [11].

4 Conclusions

–   This study aimed to express application of the stochastic approach of the LCA/LCI for the extraction process of REEs, mainly neodymium, cerium, lanthanum and tungsten, from secondary sources considered as waste produced during gold processing (Kankberg), and tungsten mine process (Covas) case studies as well as to promote the use of uncertainty approach in environmental science using MC simulation to help identify optimal solutions.

–   In real conditions, primary data parameters are usually burdened with uncertainty. A probabilistic approach to uncertainty analysis employs stochastic simulation which requires that all parameters are described by probability distributions.

–   The use of MC simulation allows for saving in time and resources. Generally, in a deterministic model, all data are known, while in a probabilistic model, data are presented and described by probabilistic distributions.

–   LCA/LCI data are full of uncertain numbers, and the MC analysis is a useful approach of quantifying parameter uncertainty in LCA studies. Lack of uncertainty analysis in LCI has influence on the LCIA results, and finally on the LCA outcomes.

–   LCA results can help in recovery of La, Ce, Ne and W through several stages and processes to be developed and proposed within the ENVIREE project. Therefore, recovery of La, Ce Ne and W can reduce the amount of the REE resources needed to be extracted.

–   Results of this study suggest that the MC method, based on the stochastic approach, is a very useful tool applied to LCI modelling under uncertainty. To our knowledge, lack publications and research presented stochastic modelling of the data used for the LCI for beneficiation of REE in the flotation tailings processes. Probabilistic techniques that used MC simulations must consider the strategy based on the specification of the optimal distribution.

–   Thanks to uncertainty analysis, a final result is obtained in the form of value range. The results obtained using MC simulation are more reliable than the deterministic approach.

- Finally, it is concluded that uncertainty analysis offer a well-defined procedure for LCI studies, early phase of LCA, and provide the basis for defining the data needs for full LCA of the beneficiation of the REEs process.
- This study represents a very important step and offers environmental information for the execution of LCIA phase, the third step of full LCA. The results of this study will encourage other researchers to consider this approach in their projects. Results can improve current procedures, can help decision makers in formulating a new recovery valuable metals production processes based on REEs beneficiation process management and bring closer to industrial application – industrially relevant focus as assured also through involvement of end users.

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