Design of experiments for the optimisation of a new process for the stabilisation of air pollution control residues

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Abstract

This work is part of the Cosmos-rice project (http://www.cosmos-rice.csmt.eu/), which was funded by the European Union under the Life+ program (LIFE11/ENV/IT000256). The aim of the Cosmos-rice project is to develop and optimise a new process, named Cosmos-rice process, for the stabilisation of Air Pollution Control (APC) residues coming from Municipal Solid Waste Incineration (MSWI) using rice husk ash as stabilizing agent (Bosio et al., 2014).

The objective of this work is to present and discuss the optimisation of the Cosmos-rice process parameters by means of the statistical Design of Experiments (DoE) methodology. The DoE refers to the process of planning the experiments so that appropriate data that can be analysed by statistical methods can be collected, resulting in valid and objective conclusions (Montgomery, 2001).

In the first part of this work, after a presentation of the Cosmos-rice process, the selection of the appropriate input and output factors (i.e. the variables that can influence the process performance and the variables selected to represent the process performance) is discussed. In particular, the input factors include the percentage of rice husk ash, the mixing temperature and time and the environmental conditions (air temperature and humidity) during the seasoning phase, while the output factors include the concentrations of Pb and Zn in the leachate of the treated APC residues.

In the second part, the plan of experiments set up for the optimisation of the Cosmos-rice process is presented. In particular, after showing the levels adopted for each of the input parameters, the resulting plan of 16 experiments is shown.
Finally, the results obtained from the experiments, carried out by the Chem4Tech laboratory of the University of Brescia (Italy), are analysed by means of the Multivariate Analysis of Variance (MANOVA). The MANOVA represents an extension of the univariate analysis of variance to the case of multiple dependent variables and allows for a direct test of the null hypothesis with respect to all the dependent variables in an experiment. Thanks to the statistical analysis, the relationships between input and output parameters have been identified, along with the best combination of the input factors levels. In particular, the environmental conditions during the seasoning phase have been found to be the parameter most affecting the effectiveness of the stabilisation process.

Keywords: Design of experiments; MSWI APC residues; MANOVA; ANOVA.

1. Introduction

The aim of the Cosmos-rice project is to develop and optimise a new process, named Cosmos-rice process, for the stabilisation of Air Pollution Control (APC) residues coming from Municipal Solid Waste Incineration (MSWI) using rice husk ash as stabilising agent. Furthermore, another project objective is to assess the sustainability of the process by identifying and quantifying its environmental benefits and by evaluating its economic viability.

The effectiveness of the process in stabilizing APC residues depends on many issues, for example on the percentage of the stabilizing agent, the seasoning conditions (temperature and relative humidity), the seasoning time, etc. All of these affect the costs of the process, such as the materials cost, the energy cost and the logistic cost, as far as its environmental impact.

The specific objective of this work is to present and discuss the optimisation of the Cosmos-rice process parameters by means of the statistical Design of Experiments (DoE) methodology. DoE is a statistical technique developed to test simultaneously the effects of several variables (called input factors) on a certain response (characterised by output factors) using only a limited number of experiments (Montgomery, 2001).

In the following, after a presentation of the stabilisation process (section 2), the DoE methodology is introduced and its application to the experimental data is discussed (section 3). Finally, conclusions are reported (section 4).

2. Cosmos-rice process for the stabilisation of APC residues from MSWI

The Cosmos-rice process consists in mixing APC residues from MSWI with rice husk ash, that works as stabilizing agent, plus other ingredients, in particular coal fly ashes, flue gas desulphurisation residues and water. The mixing can be performed at ambient temperature or at higher temperatures (during the experiments performed within the Cosmos-rice project a temperature of 60 °C was tested). The mud obtained from mixing need to be seasoned for a certain period of time, allowing the stabilisation to take place. Seasoning can take place at ambient temperature, requiring longer time, or at higher temperature, increasing the stabilisation speed.
Finally, the results obtained from the experiments, carried out by the Chem4Tech laboratory of the University of Brescia (Italy), are analysed by means of the Multivariate Analysis of Variance (MANOVA). The MANOVA represents an extension of the univariate analysis of variance to the case of multiple dependent variables and allows for a direct test of the null hypothesis with respect to all the dependent variables in an experiment. Thanks to the statistical analysis, the relationships between input and output parameters have been identified, along with the best combination of the input factors levels. In particular, the environmental conditions during the seasoning phase have been found to be the parameter most affecting the effectiveness of the stabilisation process.

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1. Introduction

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After the seasoning phase, the material obtained, named Cosmos-rice, can be disposed as a non-hazardous waste or reused as filler within other materials such as plastics, concrete or rubber. The reuse can take place with the Cosmos-rice as is or after washing it in order to eliminate salts, depending on the specific application and performance required. The following Fig. 1 gives an overview of the process.

![Fig. 1. Cosmos-rice process.](image)

3. Design of experiments methodology

The DoE methodology refers to the process of planning the experiments so that appropriate data that can be analysed by statistical methods will be collected, resulting in valid and objective conclusions (Montgomery, 2001). DoE is considered to be one of the most common and important methodologies for researchers who perform experiments to improve the performance of industrial production processes. In this paragraph, the plan of experiments set up for the optimisation of the Cosmos-rice process is presented, as far as the statistical analysis of the experimental data.

3.1 Identification of the problem

As stated above, the objective of this DoE application is to understand the relationship between Cosmos-rice process parameters (including the recipe of the ingredients) and the effectiveness of stabilisation, in order to optimise the Cosmos-rice process minimizing its cost and environmental impact.

3.2 Choice of factors and of response variables

Based on preliminary experiments results, three input factors have been chosen: A. rice husk ash percentage, B. mixing temperature and C. environmental conditions (i.e. air temperature and relative humidity) during the seasoning phase. For each of the three factors, two different levels have been identified. As usual and for convenience when representing the plan of experiments, the higher and lower levels of each factor are represented with figures +1 and -1 respectively. The factors and their levels are listed in the table below (Table 1).
With regard to the response variables, two factors have been chosen: the concentrations of Pb and Zn in the leachate of the treated APC residues, measured according to EN 12457-2:2002 and using TXRF technique (Borgese et al., 2009), after 1, 2, 4 and 6 weeks of seasoning.

3.3 Choice of the experimental plan and experiments run

The factorial plan chosen for the analysis of the process is a full factorial plan with 3 factors at 2 levels, leading to 8 combinations. Considering a number of replicates equal to 2, the number of experiments to be performed becomes 16. Moreover, the factorial plan has been randomised.

The experiments have been carried out by Chem4Tech laboratory of the University of Brescia, in Italy. In Table 2, both the factorial plan and the experimental data are reported. As the data referring to the 2nd week had an anomalous behaviour, due to a sample contamination, it was necessary to exclude them from the analysis. The concentrations of Zn and Pb in the non-treated APC residues are 9.04 mg/l and 31 mg/l respectively.

Table 1. Experimental factors and levels, input(s).

<table>
<thead>
<tr>
<th>Factors</th>
<th>Level -1</th>
<th>Level +1</th>
</tr>
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<tbody>
<tr>
<td>A Rice husk ash percentage</td>
<td>5%</td>
<td>10%</td>
</tr>
<tr>
<td>B Mixing temperature</td>
<td>20°C</td>
<td>60°C</td>
</tr>
<tr>
<td>C Environmental conditions</td>
<td>Cold (25°C + 60% RH.)</td>
<td>Hot (60°C + 25% RH.)</td>
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Table 2. Experimental plan and experimental data (in mg/l).

<table>
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<tr>
<th>Run Order</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>A</th>
<th>B</th>
<th>C</th>
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<td>2.48</td>
<td>8.11</td>
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<td>*</td>
<td>0.85</td>
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*Under the lower limit of detection (0.005 mg/l)
3.4 Statistical analysis and results interpretation

Since the stabilisation of Zn and the stabilisation of Pb are not dependent one from each other, to simplify the results interpretation, two different analyses have been performed: one concerning Zn concentrations and one concerning Pb concentrations, both with the software Minitab® 16 (Minitab Inc., 2014).

The statistical analysis of Zn data has been made by means of the Multivariate ANalysis Of VAriance, MANOVA, which is a statistical technique used to analyse the relationship between independent and dependent variables. In particular, it measures the statistical significance of the differences between mean vectors of groups of the dependent variables by the values of the independent variables (Pérez, 2004) and it can be useful in analysing repeated measures data (Finn, 1969), as data gathered at different seasoning times are, as an alternative to repeated measures ANOVA (Bock, 1975).

MANOVA analysis was realised considering not only the three main factors (A, B and C), but also their interactions (AB, AC, BC and ABC). In the results it was observed that none of the factors, or of the combinations, have a significant effect on the response except for factor C (i.e. environmental conditions during seasoning). To obtain more precision in the statistical analysis, MANOVA technique was applied considering only the significant factor C. In this case, it was necessary to realise a Box-Cox transformation \( y^* = y^\lambda, \lambda = 0.5 \) which is an appropriate method for stabilizing the variance of the response and for making the distribution of the response variable closer to the normal distribution. The results are shown in Figs. 2-4.

The second statistical analysis, concerning Pb, was performed considering only the 1\(^{st}\) week data, as several of the 4\(^{th}\) and 6\(^{th}\) week data are under the lower limit of detection, proving an almost complete stabilisation of Pb. For this reason, instead of MANOVA, it was adopted the ANalysis of VAriance, ANOVA, which is a statistical technique used to analyse the relationship between one dependent variable and diverse independent variables. In particular, the ANOVA model measures the statistical significance of the differences between mean values of groups of the
dependent variable by the values of the independent variables (Pérez, 2004). Also for the case of Pb, the analysis considered both the three main factors and their interactions. The results, presented in Fig. 5, confirm not only that the environmental conditions during the seasoning phase have a great influence on experimental responses, but also that the rice husk ash percentage has a significant influence, even if to a lower extent and at cold environmental conditions.

4. Conclusions

Optimising the Cosmos-rice stabilisation process, requires performing experiments in order to study the relationships between the parameters of the process and its performance.

Thanks to the DoE methodology, a proper set of experiments was designed in order to gather data for a significant statistical analysis. Using the MANOVA and the ANOVA techniques, it was possible to identify and characterise the above mentioned relationships and it was found that the main parameter affecting the process consists in the environmental conditions during the seasoning phase.

These results will allow to set up the process parameters in order not only to ensure the stabilisation of APC residues, but also to minimise the costs and the environmental impact of the process.

5. Acknowledgements

This work is supported by LIFE+ financial instrument of the European Union (LIFE/11 ENV/IT/000256).

References