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## Optimization of acid mine drainage remediation with central composite rotatable design model

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### Abstract

Response surface methodology using the Central Composite Rotatable Design (CCRD) model was used to optimize parameters for Acid Mine Drainage (AMD) remediation with shrimp and aquaculture farming waste (shrimp-shell and mussel byssus). The CCRD (2<sup>3</sup>) consisting of three-factored factorial design with five levels was used in this study. The dependent variables were agitation, shrimp-shell and byssus content. The results were derived by computer simulation applying least squares method using STATISTICA 7 StatSoft software. Coefficient of determination and the standard errors results from the analysis of variance have shown the model to be adequate. Predicted values were found to be in good agreement with experimental values. This study has shown that the CCRD can efficiently be applied for the modeling of AMD remediation with biomaterials and it is economical way of obtaining the maximum amount of information in a short period of time, with the fewest number of experiments.

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### 1. Introduction

Mining practices have generated serious environmental changes since the last century and made it virtually

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impossible to maintain aquatic life in affected water bodies. Mines are associated with important environmental problems, like hindered plant growth and the siltation of rivers close to the mining activity, all around the world [1–4] and specifically in the study area in Santa Catarina State, southern Brazil [5, 6].

Coal extraction processes, processing and use generate highly toxic effluents due to the degree of acidity ( $\text{pH} < 4.0$ ), significant metal ion concentrations (Fe, Al, Mn, Cu, Zn, Pb and others) and high sulfate levels. These effluents, known as acid mine drainage (AMD), cause serious environmental problems and can damage ecosystems and human health [7].

In general, the remediation of AMD involves three different processes: i) the addition of a neutralizing agent; ii) a decrease in  $\text{SO}_4^{2-}$  concentrations normally using sulfate reducing bacteria (like *Desulfovibrio* sp); and iii) the removal of other dissolved and/or particulate contaminants (metals and/or metalloids). Traditional treatment of AMD involves chemical processes. Due to the very high cost of this type of treatment, alternative and low-tech approaches have been investigated. These include the use of natural biopolymers like chitin. Crustacean shells (such as shrimp shells) are composed of a complex solid matrix of chitin (poly-N-acetylglucosamine), protein, and  $\text{CaCO}_3$ . In an aqueous medium, crustacean shells provide the slow C, N and alkalinity release source, and they can be used to remove dissolved metals due to the sorbent characteristics of the chitin [8].

The aim of the research study reported herein was to find the best conditions for an alternative and low-cost solution to the acid mine drainage problem, by producing a pH neutral and metal-free effluent. The specific objective was to apply a statistical planning DOE factorial like central composite rotatable design model for study the best conditions.

## 2. Materials and methods

### 2.1. Acid mine drainage (AMD) and biomaterials

AMD was obtained from the coalfield of state of Santa Catarina (south of Brazil), with pH around 3, and high levels of  $\text{Fe}^{2+}$  ( $83.24 \text{ mg L}^{-1}$ ),  $\text{Mn}^{2+}$  ( $5.94 \text{ mg L}^{-1}$ ) and sulfate ( $3630.8 \text{ mg L}^{-1}$ ) among other pollutants (Nb, Mg, Co, etc.) bellow the Brazilian legal limits. Shrimp shell (SS) *in natura* flakes were used as acidity and metal removal agent. SS were meticulously washed with water to eliminate the remains of organic matter and other coarse materials; subsequently, the shells were dried in an oven for 72 hours at  $100^\circ\text{C}$  for the first 48 hours and at  $50^\circ\text{C}$  for the last 24 hours. After this process, the SS were pulverized in a blender and sieved to promote greater homogeneity and contact surface. To prevent moisture absorption, they were kept in a glass desiccator until use [9]. Dry mussel byssus (MB) was used like support material of SS, predicting the operation of the remediation system in continuous flow in a future research.

### 2.2. Preliminary tests

In order to identify the remediation process and establish minimum operating parameters, different batch microcosms to analyze the influence of the pH, the relative amount of substratum/water volume (0 and  $10 \text{ g L}^{-1}$  for MB; 10, 14 and  $18 \text{ g L}^{-1}$  for SS), and the synergy/behavior of biomaterials in the remediation of AMD were prepared [8] and maintained at constant temperature ( $25 \pm 2^\circ\text{C}$ ) and agitation (150 rpm) for 24 hours. Control experiments without biopolymers were also carried out. The metals ions were monitored during the experiment by atomic absorption spectrophotometer (AAS) analysis [10]. pH was monitored during all the process.

### 2.3. Central Composite rotatable design (CCRD).

Central composite rotatable design (CCRD) was made for three factorial variables ( $2^3$ ) and five levels, being applied for optimizing the remediation process and identifying the ideal parameters for the experiment. The experiments were consisting of 8 assays ( $-1$  and  $+1$ ), 3 center points and 6 axial points ( $-1.68$  and  $+1.68$ ), and resulted in an orthogonal distribution, for a total of 17 experiments carried out by using the rotatable central composite design as shown in Table 1 and Table 2. The mathematical models were evaluated for each response by means of multiple linear regression analysis. The significant terms in the model were found by analysis of variance

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