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Optimization of copper (II) removal from aqueous solution by modified oyster shells using response surface methodology

Wijdane LIMOUNI
Laboratory of Engineering, Processes and Environment
High School of Technology, University Hassan
Casablanca Morocco
wijdane.limouni@gmail.com

Mohamed CHAFI
Laboratory of Engineering, Processes and Environment
High School of Technology, University Hassan
Casablanca Morocco

***Abstract*—The ideal conditions for the adsorption of Cu (II) ions on modified oyster shells from aqueous solution were investigated. The effect of four adsorption variables (pH, initial pollutant concentration, adsorbent mass and contact time) was studied using central composite design (CCD) under the response surface methodology (RSM) in order to determine optimal copper treatment conditions. Experimental results indicated that optimal copper removal was obtained at pH (5.5), adsorbent dosage (2g), initial copper ions concentration (150 mg/l) and contact time (2.5h). The kinetic study of adsorption was tested using pseudo-first-order and pseudo-second-order models. The results showed that the adsorption of Cu (II) ions progresses according to the pseudo-second-order model. The experimental data were analyzed by Langmuir, Freundlich and Temkin isothermal models. The study concluded that oyster shells have potential application as an efficient adsorbent to remove toxic and heavy metal like copper from industrial waste water.**

Keywords—Adsorption; heavy metals; copper; oyster shells; wastewater

I. INTRODUCTION

Water is an element classified as a source of life and has always been an irreplaceable resource. The potential of natural water resources is currently estimated in Morocco at 22 billion m3 per year. This ratio will increase by 2030 to 720 m3 / inhabitant / year, which is the limit of water scarcity. At that date 14 million inhabitants, or 35% of the total population of the country, would have less than 500 m3 / inhabitant / year. As a result, its sustainable management has become indispensable today. Water quality is also affected by industrial, agricultural and domestic applications that have an immense influence on the deterioration of this resource. Indeed, the rejections of many industries (plastic, cosmetic, paper, and surface treatment industries) are heavily loaded with heavy metals and pose one of the toxicological impacts dangerous for the environment (1-2). Among these heavy metals, copper is used in a large number of industries thanks to these special properties (thermal conductivity and resistance). The main sources of copper in industrial wastewater are: pickling and plating baths, the manufacture of tin products and also in urban runoff (insecticides, tanneries, fungicide ...). This metal is highly toxic to living organisms and to human health if it exceeds a certain concentration (3). It should therefore be removed from wastewater before discharge into the environment. To purify waters polluted by heavy metals, various treatment technologies are available such as membrane filtration (4), precipitation (5), oxidation (6), electrochemical treatment (7), ion exchange (8), and adsorption (9). This adsorption technique has been chosen in the present work. Because it is considered an economical process because of its versatility, simplicity of design, economic feasibility.

Various researchers have shown that naturally occurring materials have the ability to fix significant amounts of heavy metals (10-11). Among these materials, the oyster shells were the subject of this work (12-13).

Several studies have been conducted to determine optimal adsorption conditions. Conventional methods of a process involved studying one factor at a time by maintaining other fixed factors. This approach does not describe the combined effect of all the parameters involved in the process. This method is not only tedious, but also takes a lot of time. These limitations of a conventional method can be effectively overcome by collectively optimizing all parameters using experimental statistical designs such as the Surface Response Approach (RSM) under central composite design (CCD) (14-15).

II. MATERIELS AND METHODES

Batch adsorption studies were performed for Cu2 + removal by oyster shells in 250 ml glass flasks. The experiments were conducted while stirring, by a magnetic stirrer, a mass of the shells in 180 ml of the solution of copper. After desired contact time the adsorbent was separated from solutions using filters syringes (0, 45 µm). UV / visible spectrophotometer with maximum absorbance (λ = 683 nm).

Using RSM, 26 adsorption experiments were designed and the effect of pH, contact time, initial copper concentration, and adsorbent assay for copper removal was studied. Keeping the stirring speed, granulometry and constant temperature.

The copper removal (%) and copper adsorbed (mg / g) were evaluated using the following equations:

$$Cu(II)\ removal\ (\%) = \frac{(C0-Ct)}{C0} \quad (1)$$

$$qt = \frac{(C0-Ct)V}{m} \quad (2)$$

where q is the copper ions uptake (mg metal/g of adsorbent), C0 and Cf is the initial and final of concentration Cu (II) in solution (mg/L), V is solution volume (mL), and m is the amount of the adsorbent (mg).

III. RESULTS AND DISCUSSION

The empirical relationship between Cu2+ removal efficiency and input variables expressed by the following quadratic model:

$$R=11.041+4.12X_1-0.425X_2-19.828X_3+8.653X_4+0.215\ X_1\ X_2-0.874\ X_1\ X_3-0.125\ X_1\ X_4+3.583\ X_2X_3-0.416\ X_2\ X_4-6.613\ X_3\ X_4+1.743\ X_1^2+5.237X_2^2+23.938X_3^2-3.587\ X_4^2$$

were X1, X2, X3, X4 are adsorbent mass, initial solution concentration, pH and contact time, respectively.

From the resulting RSM model equation, it can be seen that the initial concentration, pH, adsorbent dose, initial copper concentration, and contact time have an effect on percent copper adsorption. A positive value represents an effect that favors optimization, while a negative value indicates an inverse relationship between factor and response.

Pareto analysis was performed to verify the percentage effect of each factor. As can be seen in the first figure, among the variables, the pH has the main effect on the percentage of adsorption (16).

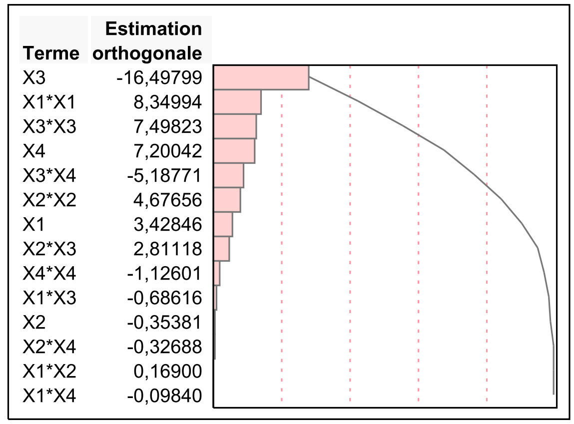

Figure 1: Pareto chart for most significant process variables

The condition of Cu (II) on oyster shells was modeled and optimized using analysis of variance (ANOVA) based on p-value and coefficient of determination (R^2) (figure 2) to evaluate the applicability and significance of the quadratic model. Furthermore, Cu (II) adsorption was also analyzed with the kinetics and isotherms models.

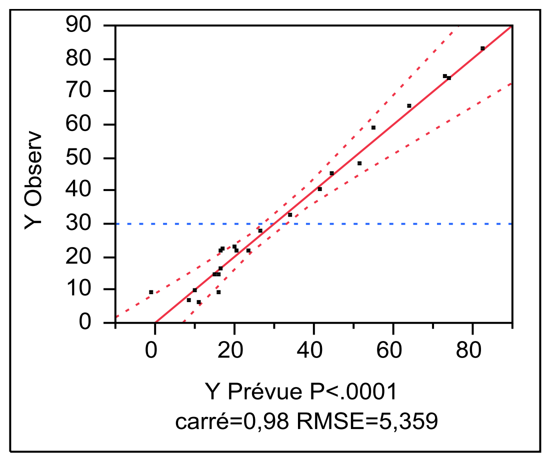

Figure 2: Regression plots of the actual data against the predicted values

The 3D response surface and is usually the graphical representation of the regression equation. It was used to find the optimal values of the process parameters.

The optimization of the Cu (II) adsorption was carried out using the JMP 10 software. The objective of the optimization process was to maximize the absorbance of Cu in the experimental range of the independent variables studied. The optimal conditions obtained for the pH (5.5), the initial concentration (150 ppm), the absorbent dosage (2g) and the contact time (2.5h). The copper removal efficiency under optimal conditions was found to be 82.54% (figure 3)

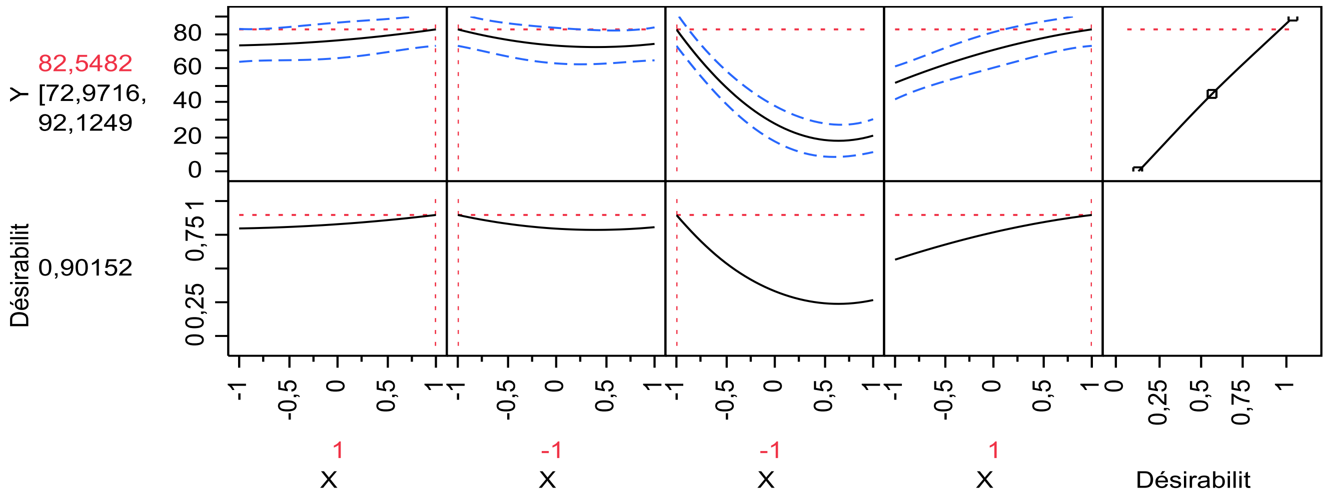

Figure 3: Response optimization plot for the maximum adsorption of Cu (II) by oyster shells

IV. Conclusion

In this study, adsorption of copper onto oyster shells was modeled and optimized using response surface methodology. The influence of operational parameters such as the adsorbent mass, initial metal concentration, pH and contact time was considered. The results revealed

that the RSM could be successfully applied for the modeling and optimizing the process variables and interactions in the response. The optimal removal obtained at adsorbent dose 2g, initial concentration 150 mg/l, pH 5.5 and contact time 1.5 h. The corresponding Cu (II) removal efficiency in these optimum conditions were found to be 82.54%. This work suggests that the oyster shells can be useful for the removal of copper from wastewater.

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