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Statistical optimization of effective parameters on saturation magnetization of nanomagnetite particles

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ABSTRACT

In this study, nanomagnetite particles have been successfully prepared via the coprecipitation method. The effect of the key explanatory variables on the saturation magnetization of synthetic nanomagnetite particles was investigated using the response surface methodology (RSM). The correlation of the involved parameters with the growth process was examined by employing the central composite design method through designating set up experiments that will determine the interaction of the variables. The vibrating sample magnetometer (VSM) was used to confirm the statistical analysis. Furthermore, the regression analysis monitors the priority of the variables' influence on the saturation magnetization of nanomagnetite particles by developing the statistical model of the saturation magnetization. According to the investigated model, the highest interaction of variable belongs to the pH and temperature with the optimized condition of 9–11, and 75–85 °C, respectively. The response obtained by VSM suggests that the saturation magnetization of nanomagnetite particles can be controlled by restricting the effective parameters.

1. Introduction

Nanomagnetite particles are one of the greatest importance among other phases of iron oxide nanoparticles, owing to the unique characteristics and exceptional physico-chemical properties. Nanomagnetite particles offer the most intensive magnetism of all transition metal oxides due to their inverse spinel structure [1,2]. Furthermore, large specific surface area and rich surface chemistry are regarded as other factors making them one of the most controversial research isuues for the past half century [3]. Therefore, nanomagnetite particles have been lately utilized in a wide range of applications, such as catalysis [4], high-density magnetic data storage [5], sensors [6], waste water treatment [7], and biomedical uses, such as targeted drug delivery and MRI contrast agent [8,9].

Nanomagnetite particles have been produced using a variety of synthetic methods, including thermal decomposition [10], hydrothermal technique [11], co-precipitation [12–14], sol–gel [15], solvothermal [16], microemulsion [17], and etc. However, chemical co-precipitation has been the most frequently used method, since it is straightforward, economical, and also capable of being scaled-up [18]. In the co-precipitation procedure, there are diverse parameters directly impacting the magnetic behavior of the final synthesized product. These factors are mainly considered as reaction condition and precursors, such as the medium of reaction (pH), temperature, reaction time, initial ratio, and addition rates of precursors. The key factors must be precisely identified and adjusted in favor of improving the desirable property of magnetite. The purpose of this study was to provide a proper design of effective parameters to maximize the magnetic properties of nanomagnetite particles for a particular end-use. In fact, our target was to investigate the effect of the aforementioned parameters in the synthesis of nanomagnetite particles using statistical techniques and response surface methodology (RSM), which resulted in the design of a series of experiments to determine the best optimized model.

The response surface methodology (RSM), initially introduced by Box and Wilson [19], is a technique for creating a roughly accurate prediction of engineered system input–output relationships and designing an optimized system. This method has been broadly utilized in great number of manufacturing fields for optimization [20–22]. RSM is an empirical modeling approach that identifies the association between diverse effective parameters and their respective responses with multiple favored criteria while

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determining the importance of effective parameters upon the coupled responses. The response surface methodology not only minimizes the experimental costs, but it also lessens the variability around the target when replacing the target value with the performance value. A profound privilige of RSM is the reproducibility of its optimal working conditions in actual applications, obtained by laboratory's experimental results or simulation [23].

In the current study, a systematical optimum model has been developed based on sequential experimental strategies in order to determine the quantitative estimation of different parameters influencing the saturation magnetization (M_s) of nanomagnetite particles. Furthermore, in our study, we employed the quadratic model of RSM incorporating the central composite rotatable design (CCRD) by taking into account four factors and three levels due to the setup of a functional optimal procedure that optimizes the design parameters. The associated mathematical models were improved by regressive analysis, followed by examination by analysis of variance (ANOVA) to measure its corresponding accuracy.

2. Materials and method

2.1. Materials

Iron (II) and (III) chloride, also called ferrous chloride tetrahydrate and ferric chloride hexahydrate, $FeCl_2 \cdot 4H_2O$ and $FeCl_3 \cdot 6H_2O$, respectively, ammonium hydroxide (28% NH_4OH), and dodecanoic acid ($C_{12}H_{24}O_2$, 99%) were all purchased from Sigma-Aldrich Co., (USA). All the chemicals and solvents were used without further purification. Moreover, during the experiments, Millipure water with a resistivity exceeding 18.0 MO/cm was used.

2.2. Preparation of nanomagnetite particles

In a typical method [24], 12 g $FeC1_2 \cdot 4H_2O$ and 24 g $FeCl_3 \cdot 6H_2O$ were separately dissolved in 50 ml of deionized water. The solutions were mixed into a 500-ml beaker, and then 50 ml of ammonium hydroxide (28% NH_4OH) was gradually added while the solution was being vigorously stirred. The precipitate was placed onto a permanent magnet to hasten the particles' sedimentation. After 5 min, the clear solution was decanted, and the precipitate was washed using a solution of 5% ammonium hydroxide in deionized water. This process is repeated to ensure that all impurities are removed. A certain amount of dodecanoic acid; 4.8 g (mole ratio of dodecanoic acid to magnetite=0.48:1) was then added to the precipitate, and the final solution was adjusted to a volume of 50 ml. The mixture was heated at 80 °C for 45 min while stirring. After cooling down the mixture, the precipitate was separated using a permanent magnet followed by filtration and washing three times with distilled water. Nanomagnetite particles were obtained after freeze drying.

2.3. Experimental design

The quadratic model is usually sufficient for industrial applications. For n-factors, the full quadratic model is shown in Eq. (1) [25]:

$$Y = b_0 + \sum biXi + \sum bijXiXj\,(I, j = 1, 2, 3, \ldots.., k) \tag{1}$$

where Y is the predicted response or dependent variable, Xi and Xj are the independent variables, while bi and bj are constants. In this case, the number of independent factors is four, and therefore, k=4: Eq. (1) becomes Eq. (2):

Table 1
Independent variables and their coded and actual values.

Independent variable	Symbol	Coded levels				
		$-\alpha$	-1	0.00	$+1$	$+\alpha$
pH	X_1	4	8	10	12	16
Temperature (°C)	X_2	30	60	75	90	120
Time (min)	X_3	10	20	45	70	80
Alkali rate of addition (mL s^{-1})	X_4	0.5	1	15.5	30	30.5

$$Yu = \beta_0 + \beta_1 \times_1 + \beta_2 \times_2 + \beta_3 \times_3 + \beta_4 \times_4 + \beta_{12} \times_1 X_2 + \beta_{13} X_1 \times_3 + \beta_{14} X_1 \times_4 + \beta_{24} X_2 \times_4 + \beta_{34} X_3 \times_4 + \beta_{11} \times_{12} + \beta_{22} \times_{22} + \beta_{33} \times_{32} + \beta_{44} \times_{42} \tag{2}$$

with Y being the predicted response, and X_1, X_2, X_3 and X_4 are the coded form of the input variables. The term β_0 is the intercept term; β_1, β_2, β_3 and β_4 are the linear terms; β_{11}, β_{22}, β_{33} and β_{44} are the squared terms; β_{12}, β_{13}, β_{23}, β_{14}, β_{24} and β_{34} are the interaction terms between the four variables; medium of reaction (pH), temperature, reaction time, addition rates of alkali (ammonium hydroxide) were selected as the influencing factors.

In the design expert software (v6.0.6), central composite rotatable design was used to evaluate the aforementioned four factors. There were five levels of points being analyzed, as shown in Table 1, with the range being determined based on massive screening experiments and literature review. Where: $-/+\alpha$, star point value; (-1), low value; ($+1$), high value; (0.00), center value.

In this study, α value was calculated using Eq. (3) (Rotatable).

$$\alpha = (F)^{1/4} \tag{3}$$

where F is the number of points in the cube section of the design (F=2^k), and k is the number of factors. Therefore, the total number of experimental combinations should be conducted based on the similar concept of CCRD by applying Eq. (4)

$$Total\ number\ of\ experimental = 2^k + 2k + no \tag{4}$$

Four factors in 24 full factorial CCRD with five levels resulted in 30 experimental runs, where k represented the number of independent variables or factors being selected. There were 6 runs of center point experiments that evaluated the pure error augmentation, with 8 axial and 16 factorial experimental runs. The response studied from the experiments was the saturation magnetization of the synthesized nanomagnetite particles. The result was obtained qualitatively from the VSM analysis. Table 2 indicates the complete design matrix of the experiments, performed together with the obtained results. The responses were used to develop an empirical model for the saturation magnetization of the iron oxide by the co-precipitation method.

After executing the experimental design, interpretations and analyzes of the experimental data were determined using ANOVA at a 5% level of significance using the Fisher F-test. The F-test is a simple arithmetical method that sorts the components of variation in a given set of data and provides the test for significance.

3. Characterization

The XRD patterns and crystal structure of the magnetite nanoparticles were recorded using Cu K_α radiation on a Rigaku Ru2000 rotating anode diffractometer. The structural properties were also investigated using Raman microscope, Renishaw (Gloucestershire, UK),