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Zinc oxide nanoparticles synthesis by electrochemical method: Optimization of parameters for maximization of productivity and characterization

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ABSTRACT

In this study, zinc oxide nanoparticles were synthesized using electrochemical method. Zinc was used as electrode whereas oxalic acid in aqueous solution was used as an electrolyte. A L_9 (3^4) Taguchi optimization methodology was used to find out the individual and interactive effect of all four independent experimental parameters namely pH (pH_o): 5–8, oxalic acid concentration (m): 0.05–0.15 M, conductivity (k): 20–30 (mS/cm) and operating voltage (V_o): 5–8 V. These experimental parameters were optimized so as to maximize the productivity (g) and correspondingly find out specific energy consumption (kW h/kg) and specific electrode consumption (kg/kg). At the optimum condition of pH_o = 5, m = 0.05 M, k = 30 (mS/cm) and V_o = 8 V, values of productivity, SENC and SELC were found to be 1.03 g, 3.79 kW h/kg and 1.76 kg/kg, respectively. Nanoparticles synthesized at optimum conditions have been further characterized by scanning electron microscopy, X-ray diffraction and UV–Visible diffuse reflectance spectroscopy techniques so as to confirm its ZnO nature.

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1. Introduction

In the last few years, there has been tremendous surge in the research related to preparation, characterization and application of metal oxide nanomaterials having enhanced properties [1]. Synthesis and characterization of nanoscale materials with special morphology and desired specific structure are of vital importance [2–6]. Zinc oxide is one such promising oxide semiconductor material which shows good electrical, optical and piezo-electrical properties. It can be used in many areas such as field-emission displays, solar cells, and gas sensors. Zinc oxide nanomaterials are also used in electronic, thermal and quantum devices, in catalysis and in wastewater treatment as an adsorbent and photocatalysts [6–10].

Various type of synthesis techniques such as sol–gel [11,12], solvo-thermal [13], hydrothermal [14,15], solution-combustion [16], and sonochemical [17] methods have been reported for the synthesis of ZnO nanoparticles (Table S1 in supporting information). It may be seen in Table S1 that only scarce papers are reported on preparation of zinc based nanoparticles by electrochemical (EC) method. Only Grobelsek et al. [18] synthesized nanoscaled ZnO using zinc or Al-alloyed zinc electrodes in acetic acid solution. Shamsipur et al. [19] used electrodeposition method for optimizing the average particle size of zinc oxalate nanoparticles by using Taguchi experimental design and controlling the concentration, stirring speed, voltage and temperature. In this study, effect of pH and conductivity was not studied and the particles were not characterized. Thus in the literature, reports on synthesis of zinc oxide nanoparticles by EC method are highly scarce.

Two type of optimization procedures namely sequential and simultaneous optimization can be used for optimizing the operating conditions. Sequential optimization requires large number of experiments, is time consuming and has slow convergence. However, simultaneous optimization gives better result and is less time consuming. In Taguchi's orthogonal array (OA) design, a number of factors can be studied by a combination of experimental conditions with very less number of experimental conditions [20]. In recent years, Taguchi's experimental methodology has been applied by various research groups, industrial, chemical and environmental engineers. Srivastava et al. [21] was used this methodology for optimizing dye removal efficiency by electrocoagulation method. Kim et al. [22] applied Taguchi robust design to optimize the experimental condition for the formation of nanoparticles by chemical reduction method. Yiamsawas et al. [13] use it for optimizing the nanocrystal growth by solvo-thermal method. Kim et al. [12] also used Taguchi design for optimizing the

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particle size by controlling the parameter molar concentration ratio and amount of dispersant feed rate by sol–gel process.

The motivation for the increasing interest toward the EC synthesis method for the preparation of nanosized compound is because of its simplicity, low temperature operation, low energy consumption and greater purity of the product obtained. Synthesis of zinc nanoparticles by EC method is affected by various parameters such as pH (pH_o), concentration (m), conductivity (k) and voltage (V_o). These parameter need to optimized for study the effect of parameter on amount of productivity (g), specific energy consumption, (SENC, kW h/kg) and specific electrode consumption (SELC, kg/kg). Taguchi's OA optimization methodology can be a very effective method to study the effect of multiple factor in efficient, time and cost effective manner [23]. Thus, the aim of the present work was to synthesize and characterize zinc oxide nanoparticles by EC method. In this study, Taguchi optimization methodology has been used to study the effect of four experimental parameter pH_o, m, k and V_o on productivity, SENC and SELC. ZnO nanoparticles synthesized at optimum conditions have been further characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD) and UV diffuse reflectance spectroscopy (UV-DRS) techniques.

2. Experimental

2.1. Material and apparatus

In this study, only analytical reagent (AR) grade chemical were used. All chemicals such as oxalic acid ($C_2H_2O_4{\cdot}2H_2O$) dihydrate purified, potassium chloride (KCl), sodium hydroxide (NaOH) and nitric acid (HNO_3) was purchase from S.D. Fine Chemical Limited, India. Zn electrode sheet of thickness 6 mm was obtained from Norrust Electro Tech, Chennai. It was cut to form two T shape electrodes. Conductivity and pH was measured by multi-parameter digital meter HACH, USA and HI 2211 HANNA instruments, respectively.

2.2. Procedure

For synthesis of nanoparticles by EC process, 200 ml of aqueous electrolyte solution was taken in 250 ml EC cell. pH and conductivity of an electrolyte solution was adjusted using 1 M NaOH and KCl, respectively. Zinc electrodes were dipped into the electrolytic cell such that the distance between the electrodes was 2 cm. Dipped dimension of the electrodes was: 2.5 cm × 1 cm × 0.6 cm. Electrodes (anode/cathode) were connected with direct current power source and the voltage was maintained as per the conditions given in Table 1. Experiments were run for 1 h at 25 °C. During experiments, the agitation speed was maintained at 300 rpm by using magnetic stirrer. After completion of run, solution was centrifuged at 8000 rpm for 15 min at 6 °C. White product obtained was further washed with distilled water and centrifuged again at same condition. Collected product was dried at 85 °C for 24 h. Electrode mass before and after experiments was used to calculate the amount of dissolution of metal. Electrode was washed with aqueous nitric acid solution (10% nitric acid) and further washed with distilled water for reuse.

2.3. Characterization of ZnO nanoparticles

The morphology of the synthesized particles was determined by a SEM (Quanta 200 F). The samples were first coated with a gold layer by a sputter coater (BALTEC SCD 005) before performing SEM measurements. XRD profiles were recorded on a Bruker D8-ADVANCE diffractometer using Cu Kα radiation (λ = 1.5418 Å) in the Bragg angle range of 5–90°. Goniometer was scanned at 2° min^{-1}. UV-DRS of the ZnO photocatalysts were obtained in the UV region (200–800 nm) by a Shimadzu UV-2100 spectrometer with $BaSO_4$ as reference.

Table 1
Parameter and their values corresponding to their levels investigated in the experiments.

Parameters		Levels		
		1	2	3
A	pH, pH_o	5	6.5	8
B	Concentration, m (M)	0.05	0.1	0.15
C	Voltage, V_o (V)	2	5	8
D	Conductivity, k (mS/cm)	20	25	30

Purity of the product was tested for ZnO by titration method using Indian standard protocol [24]. First 1.5 g of synthesized product along with 2.5 g of ammonium chloride is dissolved in 50 ml of standard hydrochloric acid (1 N). This mixture is then titrated with standard sodium hydroxide solution (1 N) using methyl orange as indicator. A blank determination without using the sample is also done. Purity of zinc oxide is calculated by using equation:

$$\text{Zinc oxide, percent by mass} = 4.07\,(B - A)/(M) \tag{1}$$

where B is the volume in ml of standard sodium hydroxide solution used in the blank determination, A is the volume in ml of standard sodium hydroxide solution used in the titration with the material and M is the mass (g) of the material taken for the test.

2.4. Taguchi's methodology

L_9 (3^4) Taguchi methodology was used for the optimizing the four parameters: pH (pH_o), concentration (m), voltage (V_o) and conductivity (k) parameter and their corresponding values are shown in Table 1. Table 2 shows the experimental plan selected along with their operating conditions. In order to reduce error, every set of experiments were conduct twice and average value of results was used for further study.

For productivity, the experimental results were used with 'higher-the-better' to estimate the condition for optimum and calculate corresponding value of productivity, SENC and SELC at optimum condition. Srivastava et al. [25,26] have reported detailed methodology for analysis.

Average response curve, analysis (ANOVA) shown in Fig. 1 have been used for analysis of optimum condition for productivity. The mean value of response at optimum conditions was calculated as:

$$Y_{\text{opt}} = \overline{T} + (\overline{A_1} - \overline{T}) + (\overline{B_1} - \overline{T}) + (\overline{C_3} - \overline{T}) + (\overline{D_3} - \overline{T}) \tag{2}$$

where ($\overline{T} = T/N$) represent the overall mean value of response and T is the grand total of all results, respectively and $\overline{A_1}$ and $\overline{B_1}$ are represent average response value at the 1st level of parameter A and B, $\overline{C_3}$ and $\overline{D_3}$ represent average response value at 3rd level of parameter C and D. The confidence intervals are given by Taguchi represent as CI_{POP} (confidence interval for the population), and CI_{EC} (confidence interval for experiments at optimum conditions) using equations given below [25,27]:

$$CI_{POP} = \sqrt{\frac{F_\alpha(1, f_e)V_e}{n_{\text{eff}}}} \tag{3}$$

$$CI_{CE} = \sqrt{F_\alpha(1, f_e)V_e\left[\frac{1}{n_{\text{eff}}} + \frac{1}{R}\right]} \tag{4}$$

where R is the sample size for confirmation experiment, $F_\alpha(1, f_e)$ is the F-ratio at a confidence level of $(1 - \alpha)$ against degree of freedom (DOF) 1 and error DOF f_e, V_e is error variance.

$$n_{\text{eff}} = \frac{N}{1 + [\text{Total DOF associated in the estimate of the mean}]} \tag{5}$$

where N is the number of results. For verifying the results obtained from experiments, confirmation experiments were done in three times at fixed optimum condition. Optimum response value Y_{opt} is compared with average value of result of confirmation experiments.

Table 2
Experimental parameter, their levels and results for conducted experiments corresponding to L_9 (3^4) Taguchi's experimental plan.

Expt. no.	Run	A	B	C	D	Productivity (g)	Specific energy consumed (kW h/kg)	Specific electrode consumed (kg/kg)
1	Zn1	1	1	1	1	0.3624	0.5795	0.2586
2	Zn2	1	2	2	2	0.4552	4.1301	1.2463
3	Zn3	1	3	3	3	0.4968	4.5491	0.8321
4	Zn4	2	1	2	3	0.7289	5.474	1.5201
5	Zn5	2	2	3	1	0.1722	4.6458	1.41
6	Zn6	2	3	1	2	0.0614	0.6515	2.1661
7	Zn7	3	1	3	2	0.8441	5.8998	1.139
8	Zn8	3	2	1	3	0.3137	1.1476	0.9146
9	Zn9	3	3	2	1	0.023	9.5652	8.033

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