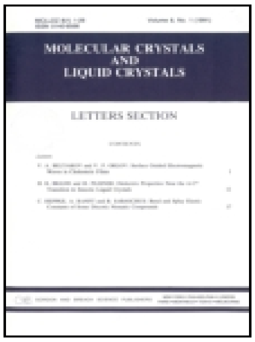

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Optimization of parameters for deposition of ZnO films by sol gel using Taguchi method

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ABSTRACT
ZnO transparent conductive films were deposited on glass substrates by sol-gel spin coating method. Taguchi method was used to find the optimal deposition parameters, three influential parameters were selected in this experiment, Concentration of zinc (II) ions. Zn^{2+}, annealing temperature T_r and pre-annealing temperature T_p. By employing the analysis of variance, we found that the annealing temperature T_r and the precursor Zn^{2+} are the most influencing parameters on the properties of ZnO films. Under the optimized deposition conditions, the ZnO films showed high crystal quality, and high transmittance of 90% in the visible region.

KEYWORDS
ZnO; Texture coefficient; Sol-gel; Spin Coating; Taguchi method

Introduction

Taguchi method is a statistical method developed by Taguchi and Konishi [1]. Initially it was developed for improving the quality of goods manufactured (manufacturing process development), later its application was expanded to many other fields in Engineering. Taguchi Method involves identification of proper control factors to obtain the optimum results of the process. Orthogonal Arrays (OA) are used to conduct a set of experiments. Results of these experiments are used to analyze the data and predict the quality of components produced.

In this work, Taguchi's design method was introduced. By applying Taguchi's design [2,3], one can significantly reduce the time required for experimental investigation, as it is effective in investigating the effects of multiple factors on performance as well as to study the influence of individual factors to determine which factor has more influence, than others and which has less.

The main objective of this work is to apply the Taguchi's design method on the optimization of parameters, as well as to study the structural and optical properties of ZnO films deposited at the optimized conditions.

ZnO thin films have been studied extensively due to their various applications, such as piezoelectric transducers, conductive gas sensors, display devices, transparent conductive electrodes, and solar cells [4–6]. There are various techniques used for preparation of ZnO thin films such as RF magnetron sputtering [7], Physical vapor deposition [8], Chemical vapour deposition (CVD) [9], spray pyrolysis [10] and sol–gel technique [11]. Among them

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Table 1. Parameters and levels used in this experiment.

		Level 1	Level 2	Level 3
A	Concentration of zinc (II) ions [Zn^{2+}]	0,70	0,75	0,8
B	Pre-annealing temperature (Tp)	200	250	300
C	Annealing temperature (Tr)	450	500	550

sol–gel method especially associated to spin coating has received high attention because of simple deposition procedure, easy control of chemical components and low-cost preparation to obtain high quality thin films.

The sol-gel method involves many process parameters such as molar concentration of precursor, rotation speeds, pre and post heat treatment, stirring time and drying time, which directly or indirectly influence the quality of thin films. Hence, it requires a greater understanding of the relation amongst the process parameters in order to optimize the quality of thin films.

Experimental details

Selection of deposition parameters and their levels

ZnO thin films have been deposited by spin coating method onto glass substrates. Zinc acetate dihydrate, 2-methoxyethanol and (MEA) Monoethanolamine were used as starting precursor, solvent, and sol stabilizer, respectively. The molar ratio of MEA to zinc acetate dihydrate was maintained at 1:1. The solution was stirred at 60°C for 2 h to yield a clear and homogeneous solution, which served as the coating solution after cooling to room temperature. The coating solution was dropped onto a glass substrate, which was rotated at 3000 rpm for 30 s. Table 1 shows the Parameters and levels used in this experiment.

Before this work, our group had done a series of experiment to study the effects of individual parameters on the properties of ZnO thin films [11–13]. Three influential deposition parameters, i.e., Concentration of zinc (II) ions, pre-annealing temperature Tp and annealing temperature Tr, were selected in this experiment. The levels of the deposition parameters were analyzed in the preliminary experiment. Table 1 shows the parameters and levels used in this experiment.

Orthogonal array and characterization

In order to optimize the deposition process, the Taguchi's experimental design, a $L_9(3^3)$ orthogonal array with three columns and nine rows, was used [9]. Each parameter was assigned to a column, nine deposition-parameter combinations being available. Therefore, only nine experiments were required to study the entire parameter space using the L_9 orthogonal array.

For characterization we have used a high-resolution X-ray diffraction for XRD patterns in the θ–2θ configuration with a copper anticathode (CuKa, 1.54A°). Scanning Electron Microscopy (SU-1500, HITACHI) was used for studying the morphology of thin films. The optical properties were monitored by transmittance using a Xe lamp in association with a 500 mm Yvon–Jobin HR460 spectro- photometer using a GaAs Photomultiplier tube detector optimized for the UV–VIS range.

Table 2. Experimental values and the corresponding S/N ratio

	Control factors			Texture Coefficient TC (hkl)		
Exp. no.	A	B	C	Sample 1	Sample 2	S/N ratio
1#	A1	B1	C1	1.49	01,34	3.463
2#	A1	B2	C2	1.53	01,7	3.693
3#	A1	B3	C3	3.35	03,15	10.500
4#	A2	B1	C3	5.09	5,03	14.134
5#	A2	B2	C2	2.41	02,04	7.6403
6#	A2	B3	C1	1.05	03,25	0.423
7#	A3	B1	C1	3.41	03,27	10.650
8#	A3	B2	C3	1.4	01,45	2.922
9#	A3	B3	C2	1.69	01,81	4.557

Results and discussion

Analysis of the S/N ratio

The S/N ratio in Taguchi method is used to measure the performance characteristic including: "lower is better" (μ_L), "higher is better" (μ_H). In this article, ρ is the μ_H performance characteristic. The corresponding S/N ratios of μ_H can be calculated according to the following equation [14]:

$$\mu_H = -10 \log \frac{1}{n} \sum_{i=1}^{n} \left(\frac{1}{Y_n}\right)^2 \tag{1}$$

Where n is the number of repeated experiments

Y_n represents the measured experimental value (i.e., the texture coefficient TC(hkl)).

The texture coefficient TC(hkl) represents the texture of a particular plane, whose deviation from unity implies the preferred growth. Quantitative information concerning the preferential crystallite orientation was obtained from another texture coefficient TC(hkl) defined as [15]:

$$\mathrm{TC(hkl)} = (\mathrm{I}_{\downarrow}\mathrm{r(hkl)}/\mathrm{I}_{\downarrow}0\ \mathrm{(hkl)})/(1/\mathrm{n}\sum[[(\mathrm{I}_{\downarrow}0(\mathrm{hkl})/[[(\mathrm{I})]]_{\downarrow}\mathrm{r(hkl)}]] \tag{2}$$

Where I(hkl) is the measured relative intensity of a plane (hkl)

I_0(hkl) is the standard intensity of the plane (hkl) taken from the JCPDS data.

The value TC(hkl) = 1 represents films with randomly oriented crystallites, while higher values indicate the abundance of grains oriented in a given (hkl) direction.

Table 2 shows the experimental data on texture coefficient and the corresponding S/N ratio calculated by the above equation (1). The mean S/N ratio for each level of the parameters is summarized and the S/N response table for texture coefficient is shown in Table 3.

Figure 1 shows the graphical representation of the means of the S/N ratios of all levels of factors allows determining the optimal conditions. The optimum value of each factor corresponds to the maximum point in the curve. The optimal condition is then constituted by the

Table 3. S/N response for texture coefficient and optimal parameters

		Mean S/N ratio (dB)			
Symbol	Parameters	Level-1	Level-2	Level-3	Optimal parameters
A	$[Zn^{2+}]$	5.886150042	**7.39949416**	6.045127462	A2
B	T_p	**9.417722865**	4.752243394	5.160805405	B1
C	T_r	4.847532976	5.297301187	**9.1859375**	C3

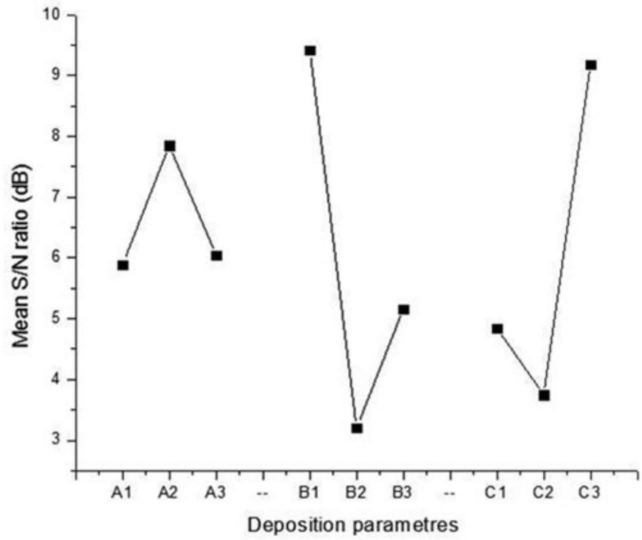

Figure 1. S/N graph for texture coefficient.

set of maximum points of each factor. It can also allow knowing the most influential factor of the experiment. If the slope of the curve is sharp, the effect of the factor is significant if the slope is low the factor has less effect on responses. According to what has been said above, the most influential factor is the annealing temperature, followed by the concentration of zinc and finally the pre-annealing temperature. The optimum condition is A2, B1 and C3.

Characterization of ZnO films

After the determination of the optimum condition a confirmation experiment was done and the characterization results are shown on figures below.

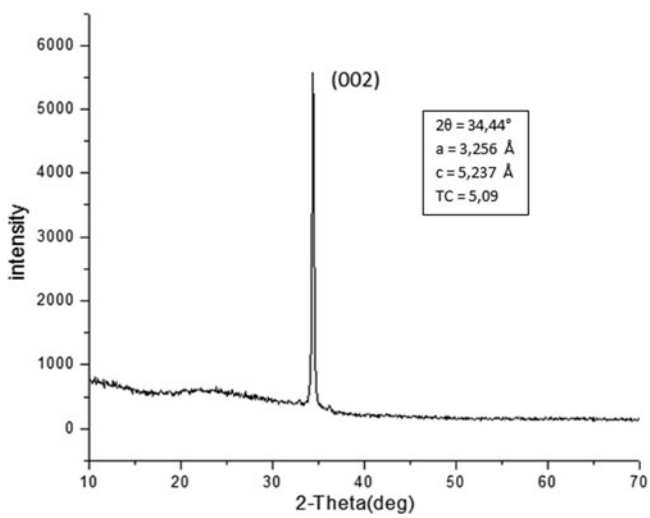

Figure 2. XRD pattern of the ZnO film deposited at optimized conditions.

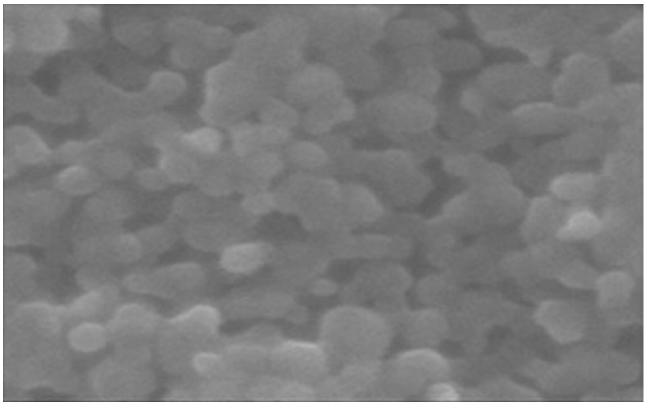

Figure 3. SEM image of the ZnO film deposited at optimized conditions.

Figure 2 shows the XRD pattern of the ZnO film deposited at optimal conditions. the film was crystallized with the hexagonal wurtzite structure and a strong preferred orientation (5574 au) along the direction (002) which is located at $2\theta = 34.44°$ along the c axis with lattice parameters a = 0.3258 nm and c = 0.5225 nm, very close to the theoretical values a = 0.3256 nm and c = 0.5237 nm.

Figure 3 shows an SEM image of ZnO thin film deposited at optimal conditions. It has a homogeneous surface morphology consisting of hexagonal crystallites in which the entire surface is covered with grains of ZnO with an average grain size of approximately 50 nm.

To identify the chemical states of elements in samples, EDX analysis was performed. Figure 4 shows the peaks associated with Zn and O elements are present; parallel peaks of Mg, Al, Si and Ca are also observed, but from the substrate on which the sample was deposited. The zinc and oxygen elements are in respective proportions of 52.7% and 47.3%.

Figure 5 conditions depending on the wavelength in the range of 350 nm to 1100 nm. The evaluated spectrum in the visible region of the optical transmission is around 90%.

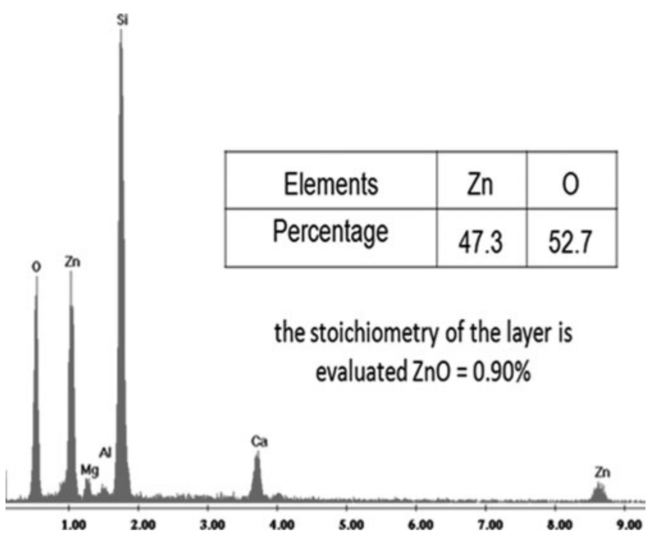

Figure 4. EDX of the ZnO film deposited at optimized conditions.

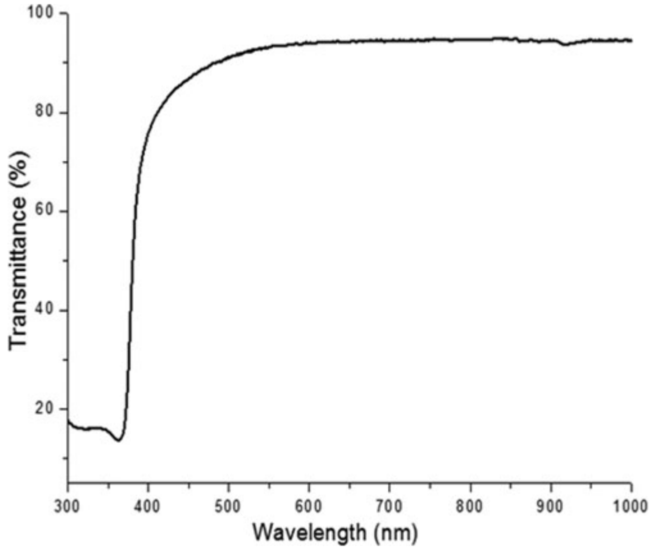

Figure 5. Transmittance spectrum of the ZnO film deposited and prepared at optimized conditions.

From the transmission curve, we can calculate the direct band gap by the following equation, which represents the variation of (α.hυ) ^{2} with (hυ) [16].

$$(\alpha.h\upsilon)^2 = B(h\upsilon - Eg) = f(h\upsilon)$$

hυ is the energy of incident photons
Eg the optical gap
B is a constant.

Figure 6 shows energy-band gap of ZnO thin film deposited at optimal condition. The value of the energy-band gap of ZnO thin films is equal to Eg = 3, 28 eV and is very close to the theoretical value Eg = 3, 28 eV.

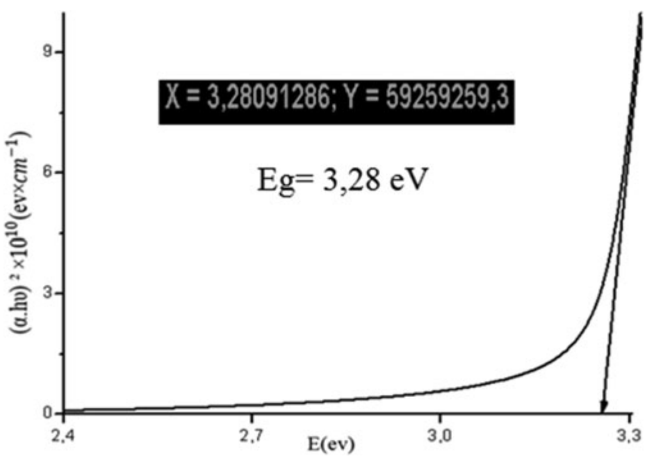

Figure 6. Determination of the energy gap by extrapolation method from the variation of (α.hυ)^2 according to (hυ) for a thin layer of ZnO.

Conclusions

In conclusion, we present the application of $L_9(3^3)$ orthogonal array to optimize the properties of ZnO thin films deposited on glass substrates by sol-gel spin coating method. The results show that this method can greatly simplify the optimization of multiple parameters. It has been showed that the annealing temperature and the precursor (Zn^{2+}) are the most influencing parameters on the properties of ZnO films. XRD, SEM/EDX and UV-Vis Transmission results confirmed the optimal conditions of deposition, which are important for the quality of ZnO thin films by sol-gel spin coating method.

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References

[1] Taguchi, G., & Konishi, S. (1987). *American Supplier Institute*, *1*, 8.
[2] Taguchi, G. (1986). *Introduction to Quality Engineering*, Asian Productivity Organization: Tokyo.
[3] Yang, W.H., & Tarng, Y.S. (1998). *Journal of Materials Processing Technology*, *84*, 122.
[4] Smirnov, M., Baban, C., & Rusu, G.I. (2010). *Applied Surface Science*, *256*, 2405.
[5] Triboulet, R., & Perriere, J. (2003). *Progress in Crystal Growth and Characterization of Materials*, *47*, 65.
[6] Shaoqiang, C., Jian, Z., Xiao, F., Xiaohua, W., Laiqiang, L., Yanling, S., & Qingsong, X. (2005). *Applied Surface Science*, *241*, 384.
[7] Dedova, T., Acik, I.O., Krunks, M., Mikli, V., Volobujeva, O., & Mere, A. (2012). *Thin Solid Films*, *520*, 4650.
[8] Zhou, Y., Kelly, P.J., Postill, A., Abu-Zeid, O., & Alnajjar, A.A. (2004). *Thin Solid Films*, *447–448*, 33.
[9] Kim, D.D., Yun, I.I., & Kim, H.H. (2010). *SUPPL*, *10*, S459.
[10] Bizarro, M., Sánchez-Arzate, A., Garduño-Wilches, I., Alonso, J.C., & Ortiz, A. (2011). *Catalysis Today*, *166*, 129.
[11] Konan, K., Hartiti, B., Aka, B., Ridah, A., Dakhsi, K., Arba, Y., & Thevenin, P. (2010). *Afrique SCIENCE*, *06*(1), 29.
[12] Ammaih, Y., Lfakir, L., Hartiti, B., Ridah, A., Thevenin, P., & Siadat, M. (2014). *Optical and Quantum Electronics*, *46*, 229.
[13] Ammaih, Y., Lfakir, L., Hartiti, B., Ridah, A., Thevenin, P., & Siadat, M. (2013). *Moroccan Journal of Condensed Matter*, 15, 10.
[14] Ghani, J.A., Choudhury, I.A., & Hassan, H.H. (2004). *Journal of Materials Processing Technology*, *145*, 84.
[15] Barret, C. S., & Massalski, T. B. (1980). *Structure of Metals*, Pergamon Press: Oxford.
[16] Daneshvar, N., et al. (2008). *International Journal of Chemical and Biomolecular Engineering*, *1*(1), 24.