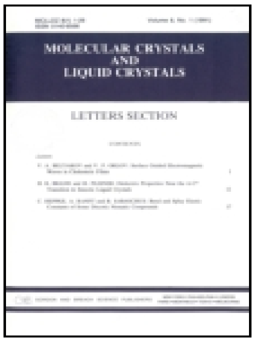

Taylor & Francis
Taylor & Francis Group

Molecular Crystals and Liquid Crystals

ISSN: 1542-1406 (Print) 1563-5287 (Online) Journal homepage: http://www.tandfonline.com/loi/gmcl20

Optimization of parameters for deposition of In_2S_3 films by spray pyrolysis using Taguchi method

S. Elfarrass, B. Hartiti, A. Ridah & P. Thevenin

To cite this article: S. Elfarrass, B. Hartiti, A. Ridah & P. Thevenin (2016) Optimization of parameters for deposition of In_2S_3 films by spray pyrolysis using Taguchi method, Molecular Crystals and Liquid Crystals, 628:1, 139-144, DOI: 10.1080/15421406.2015.1137151

To link to this article: http://dx.doi.org/10.1080/15421406.2015.1137151

Published online: 13 May 2016.

MOL. CRYST. LIQ. CRYST.
2016, VOL. 628, 139–144
http://dx.doi.org/10.1080/15421406.2015.1137151

Taylor & Francis
Taylor & Francis Group

Optimization of parameters for deposition of In_2S_3 films by spray pyrolysis using Taguchi method

S. Elfarrass[a,b], B. Hartiti[a], A. Ridah[b], and P. Thevenin[c]

[a]MAC & PM Laboratory, ANEPMAER Group, Department of Physics, University Hassan II FSTM, Mohammedia, Morocco; [b]LIMAT Laboratory, Department of Physics, University Hassan II FSB, Casablanca, Morocco; [c]LMOPS Laboratory, University of Lorraine, Metz, France

ABSTRACT
The aim of this research is to investigate the influence of the experimental conditions on the structural, morphological and optical properties of Indium Sulfide thin films deposition according to the Taguchi analysis method. The films are deposited on a glass substrate by spray pyrolysis a deposition method which is dependent on the concentrations of the precursor (indium and sulfide), the substrate temperature and the duration of the process. These four parameters are the key factors to be optimized in our study in conjunction with the morphological and optical properties of the films thanks to the Taguchi analysis, using a L_9 Orthogonal array.

KEYWORDS
Indium sulfide; Spray pyrolysis; Texture coefficient; Taguchi method

I. Introduction

An important research goal in the development of photovoltaic devices is to replace the costly materials and even hazardous heavy metals by cheaper and more benign elements. Thin film technologies aim to use less material to obtain a considerable efficiency. Attention has been paid to indium-sulfide-based thin films as a window buffer layer for solar cells replacing CdS, not only to eliminate toxic cadmium but also to improve light transmission at short wavelengths, because of its lack of toxicity, its wide energy gap and its photo-conducting nature. It can also be dry-deposited at low temperatures [1]. In_2S_3 exhibits n-type electrical conductivity, with a direct energy band gap of 2.75 eV and a high optical transmittance (70–80%) in the visible region [2]. Deposition of thin layer indium sulfide has been carried out by different methods such as MOCVD, spray pyrolysis, and chemical bath deposition (CBD) [3–4] by several groups. β-In_2S_3, the stable phase of In_2S_3 at room temperature, crystallizes in a defect spinel lattice, with a high degree of vacancies, ordering at tetrahedral cationic sites [5].

In this work, we want to optimize the key deposition parameters of our experimental process with respect of the physical and optical properties of the In_2S_3 films, thanks to the Taguchi's method analysis. This method is effective in investigating the effects of multiple factors on the performances of the final product as well as in studying the influence of individual factors in order to know which is more influential than others [6, 7]. Accordingly, we expect to reduce significantly the time required in the optimization process by working on a reduced set of samples. In this study, the structure and morphology of the In_2S_3 thin films is

CONTACT S. Elfarrass s.elfarrass@gmail.com
Color versions of one or more of the figures in the article can be found online at www.tandfonline.com/gmcl.

Table 1. Parameters and levels used in this experiment.

Symbol	Parameters	Level 1	Level 2	Level 3
A	Substrate temperature (°C)	300	350	400
B	Indium concentration (mol/l)	0.025	0.03	0.035
C	Thiourea concentration (mol/l)	0.0375	0.045	0.0525
D	Spray time (min)	10	15	20

analyzed using X-ray diffraction (XRD) and their physic-optical properties by UV–vis-NIR transmission measurements.

II. Experimental details

The indium sulfide (In_2S_3) thin films were synthesized by spray pyrolysis technique starting with an aqueous solution containing indium chloride, and thiourea. The precursor solution was sprayed, using a pneumatically controlled air-atomizing spray nozzle (14JAU, Spraying systems Co., USA), onto heated glass substrates, with a solution flow-rate of 1 mL/min. Compressed air was used as the carrier gas, and the pressure was fixed at 2.5 bar. Four influential deposition parameters are analyzed in this experiment, the substrate temperature, the concentration of indium and sulfide precursors, and the duration of the spray process, the various values (level) of these key parameters are listed in table 1, and the experimental set of combinations between them used in this work is presented in table 2.

III. Results and discussion

III.1 Taguchi method

The Taguchi method allows the determination of the effect of different parameters on the mean and variance of process performance characteristics that defines the effectiveness of the process [8, 9]. In this study, the Taguchi method with an L_9 (3^4) orthogonal array robust design is used to determine the optimal experimental conditions that produce the best deposition parameters of In_2S_3 films prepared using spray pyrolysis technique. As indicated previously, the values of the various deposition parameters are listed in table 2, and two series of morphological and optical characterization measurements on the samples are also shown in the table.

The experiments were repeated twice and the mean values for each output were subsequently used for analysis of the results. The optimal values were determined by comparing the signal-to-noise (S/N) ratio, using the Taguchi method. In this study, the structural and

Table 2. Combinations between parameters where the rows represent tests.

Experiment	A	B	C	D	Texture Coefficient TC (hkl)	S/N ratio
1	A1	B1	C1	D1	1.794	5.076
2	A1	B2	C2	D2	4.326	12.721
3	A1	B3	C3	D3	5.630	15.010
4	A2	B1	C2	D3	4.518	13.098
5	A2	B2	C3	D1	3.895	11.810
6	A2	B3	C1	D2	4.874	13.757
7	A3	B1	C3	D2	3.646	11.236
8	A3	B2	C1	D3	4.861	13.734
9	A3	B3	C2	D1	4.251	12.569

Table 3. The mean of S/N response of performance criteria.

	Level1	Level2	Level3	Optimal parameters
A	10.935	12.888	12.513	A2
B	9.803	12.755	13.778	B3
C	10.855	12.796	12.685	C2
D	9.818	12.571	13.947	D3

the optical properties of the In_2S_3 thin films were chosen as the basis for optimization of the deposition parameters, in order to allow the larger the better characteristics. The S/N ratio for the larger the better characteristic $(S/N)_L$ is expressed as follows equation (1):

$$\frac{S}{N} = -10 \log \frac{1}{n} \sum_{i=1}^{n} \frac{1}{Y_i^2} \tag{1}$$

where n is the number of the experiment and Y_i is the average measured value of the experimental data. (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 crystalline orientation was obtained from another texture coefficient TC (hkl) defined as in equation (2):

$$\mathrm{TC\ (hkl)} = [\mathrm{I_r\ (hkl)\ / I_0\ (hkl)}] \ / \left[1/\mathrm{n} \sum (\mathrm{I_r\ (hkl)\ / I_0\ (hkl)})\right] \tag{2}$$

where I (hkl) is the measured relative intensity of a plane (hkl) and 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.

III.2. Texture coefficient

Analysis of variance (ANOVA) can be used to understand the relative significance of the process effects on the experimental responses and to estimate the experimental error due to different associated factors. Table 2 shows the experimental results for texture coefficient and its S/N ratio. The mean of the S/N ratio for each level of the parameters is summarized in table 3.

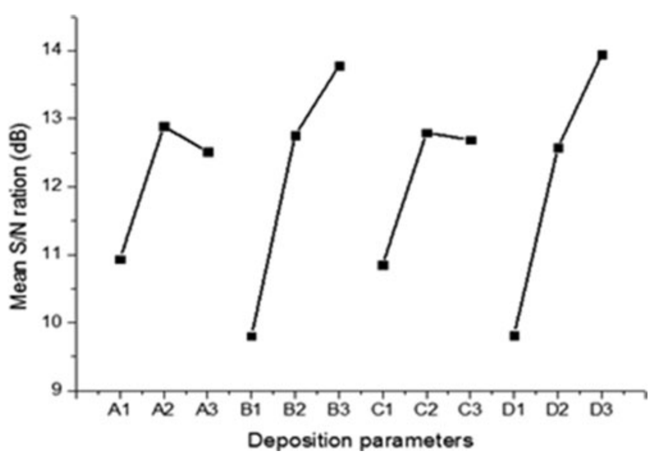

Figure 1. S/N response graph for texture coefficient.. Note: A = Substrate temperature (°C), B = Indium ion concentration (mol/l), C = Thiourea ion concentration (mol/l), D = Spray times (min).

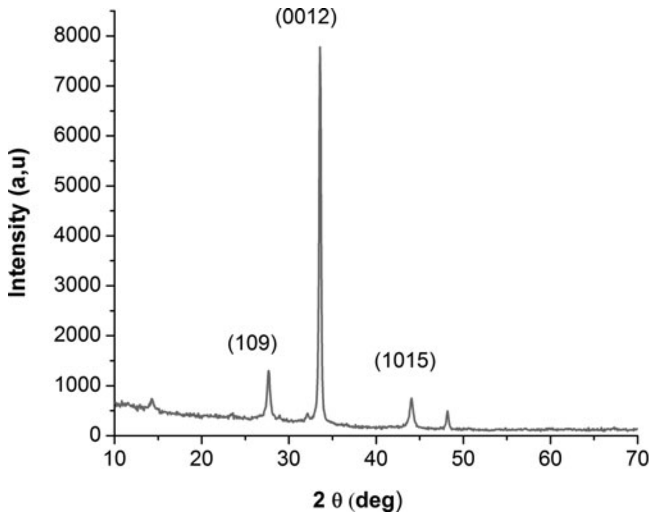

Figure 2. The XRD pattern of the In_2S_3 film deposited at optimal conditions.

Besides, the graphical representation of means of the S/N ratios of all factors at all levels leads to determine graphically the factors having more effects on film properties than others. The optimal condition is A_2, B_3, C_2 and D_3. In other words, based on the S/N ratio and ANOVA analyses showed in Table 3, the optimal parameters (conditions) for texture coefficient are the A at level 2, B at level 3, C at level 2 and D at level 3. Fig. 1 shows the S/N graph for texture coefficient.

Figure 2 shows the XRD pattern of the In_2S_3 film deposited at optimal conditions. The film was crystallized with the tetragonal structure and a strongly preferred orientation along the direction (0012) which is located at $2\theta = 33.64°$.

The optical transmittance curve as a function of the wavelength at optimal conditions is shown in Fig. 3. The films present a high transmittance in the visible wavelength range, mostly in excess of 70%.

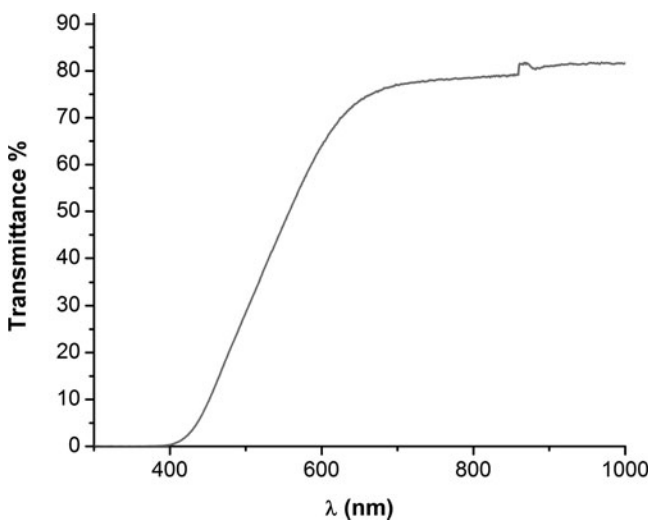

Figure 3. The transmittance spectrum of the In_2S_3 film deposited prepared at optimized conditions.

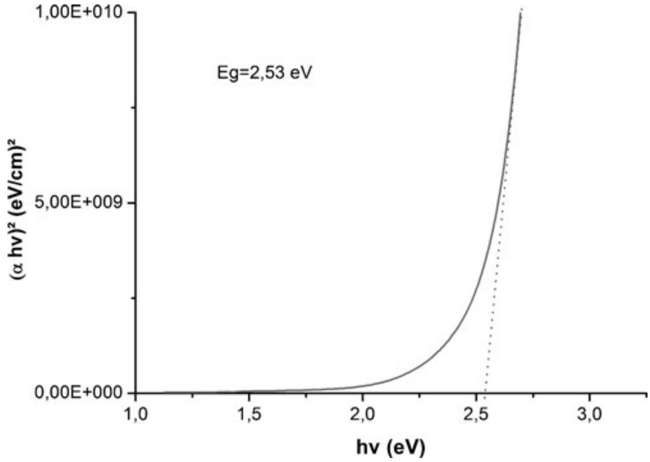

Figure 4. Variation of $(\alpha hv)^2$ with the incident photon energy hv for the sample prepared at optimal condition.

Figure 4 shows the variation of $(\alpha hv)^2$ as a function of photon energy (hv)for the deposited In_2S_3 thin film under optimal condition. Based on the direct allowed inter band transition theory, the optical band gap of In_2S_3 is determined by extrapolating the linear part of the curve to the zero absorption coefficient $\alpha = 0$ as shown in Fig. 4.The film exhibits an energy gap of 2.53eV

IV. Conclusions

In this study, an L_9 (3^4) orthogonal array with Taguchi method is used to optimize the multiple performance characteristics of In_2S_3 thin films that are deposited onto glass substrates using spray pyrolysis method with various deposition parameters. This method converts the multiple performance characteristics into a single performance characteristic and simplifies the optimization procedure. This optimal result can be used for practical deposition processes to effectively reduce production costs and to produce great improvements in film quality.

Acknowledgments

This work has been partially supported by the AAP Research 2013 Project funds) of the IFM (Institut Français au Maroc). Technical support from LMOPS & LCOMS labs (University of Lorraine) is gratefully acknowledged.

References

[1] Timoumi, A., Bouzouita, H., Brini, R., Kanzari, M., & Rezig, B. (2006). *Applied Surface Science*, 253 306–310.
[2] Lokhande, C.D., Ennoui, A., Patil, P.S., Giersig, M., Diesner, K., Muller, M., & Tributsch, H. (1999). *Thin Solid Films*, 340 18–23.
[3] Mane, R.S., & Lokhande, C.D. (2003). *Mater. Chem. Phys.*, 78, 15.
[4] Castelo-Gonzalez, O.A., Santacruz-Ortega, H.C., Quevedo-Lopez, M.A., & Sotelo-Lerma, M. (2012). *J. Electron. Mater.*, 41, 695.
[5] Kambas, K., Spyridelis, J., & Balkanski, M. (1981). *Phys. Status Solidi B*, 105, 291.
[6] Taguchi, G. (1986). *Introduction to Quality Engineering*, Asian Productivity Organization: Tokyo.

[7] Yang, W.H., Tarng, Y.S., & Mater, J. (1998). *Journal of Materials Processing Technology*, 84, 122–129.

[8] Taguchi, G., Elsayed, E.A., & Hsaing, T. (1989). *Quality Engineering in Production Systems*, McGraw-Hill: New York.

[9] Park, S.H. (1996). *Robust Design and Analysis for Quality Engineering*, Chapman & Hall: New Jersey.