OPTIMIZATION OF DEPOSITION OF THIN PHOTOANISOTROPIC FILMS FOR HOLOGRAPHIC DATA STORAGE

Lian Nedelchev, Georgi Mateev, Anna Otsetova, Dimana Nazarova, Elena Stoykova

***Abstract**: Holography offers the possibility for high density and high capacity volumetric optical data storage combined with very high data transfer rates. These parameters are required by many advanced applications – Internet of Things, 4K and 8K television, cloud data storage etc. Polarization holography allows to further increase data capacity by registering also the polarization state of light. However, the properties of the recording photoanisotropic media are crucial: precisely controlled thickness, high surface smoothness of the layer, excellent optical quality and transparency.*

In this paper, we present a study of thickness and optical absorbance of series of thin azopolymer films prepared by spin coating varying the concentration of the solutions, rotation speed and type of solvent. The optimal conditions for different applications are outlined.

***Keywords**: Optical data storage, Holographic memories, Azopolymer, Thin films deposition*

***ITHEA Keywords**: B.3 Memory Structures, E.2 Data Storage Representations*

Introduction

Optical data storage is best known to the general public by the technology line Compact Disc (CD) / Digital Versatile Disc (DVD) / blu-ray disc [O'Kelly, 2004]. By decreasing the wavelength of light from 780 nm to 405 nm, the capacity has increased from 700 MB to 25 GB i.e. by a factor of 35. In spite of the reliability and low bit error rate, this technology has a significant constraint: the information is recorded in a single layer (2D) which limits the density and capacity of this media format.

Alternative data storage approach is the volumetric (3D) optical storage. In this case, the information is stored in the entire volume of the recording media. The most common technology in this field is the holographic data storage [Coufal, 2000]. Using the approximation $1/\lambda^3$, we can determine the theoretical capacity of a CD size media to 22 TB [Nedelchev, 2011]. Combined with very high data transfer rates, allowed by the parallel reading/recording of entire data pages (with data size of the order of Megabytes), it becomes clear why this technology could be of interest for many of the latest IT applications.

Polarization holography adds yet another dimension, by registering also the polarization state of light and is sometimes referred to as 5D data storage [Zhang et al, 2016; Nikolova and Ramanujam, 2009]. Various approaches can be used to achieve high density of information – angle, shift, wavelength and polarization multiplexing [Yao et al, 2005]. However, in order to record the polarization state of light, the media should be photoanisotropic. As shown by numerous researchers, one of the most efficient materials with photoinduced birefringence are azopolymers [Todorov et al, 1984; Natansohn and Rochon, 2002; Wang, 2017]. We should also note that the optical properties of the recording photoanisotropic media are very important: precisely controlled thickness, high surface smoothness of the layer, excellent optical quality and transparency.

In this paper, we present a study of thickness and surface smoothness of series of thin azopolymer films prepared by spin coating varying the concentration of the solutions and rotation speed. Two different methods for thickness measurement are used and mutually verified – spectrophotometric and interferometric. The optimal conditions for different applications are outlined. Smoothness of the films is characterized by Atomic Force Microscopy (AFM).

Preparation of the Thin Films

The azopolymer used in our experiments is poly[1-[4-(3-carboxy-4-hydroxyphenylazo)benzene sulfon amido]-1,2-ethanediyl, sodium salt] or shortly PAZO (Sigma Aldrich). Its structure is shown in Fig. 1.

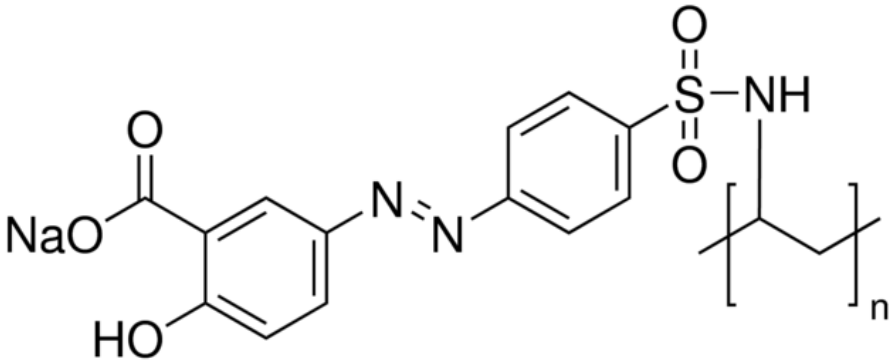


Figure 1. Chemical structure of the PAZO polymer.

The polymer is readily soluble in water, as well as in methanol. The polymer solutions are spin-coated on glass substrates (BK7). Three main parameters influence the thickness and smoothness of the

resulting thin films: 1) type of solvent – water or methanol, 2) concentration of the polymer in the solution *C*, which is varied from 25 to 100 mg/ml, and 3) the rotation speed of the spin-coater *R*, which is varied from 300 to 2000 rpm.

After the spin coating procedure, the films are left at room temperature for 24 hours to ensure complete solvent evaporation. In all the cases, homogenous films with very good optical quality were obtained.

Thickness measurement

Two methods for thickness measurement of thin films are presented in this Section. Both methods are optical and allow fast and non-destructive measurement. The measured spot on the film is approximately 5 mm in diameter, which very well corresponds with laser spot size for optical recording.

A. Spectrophotometric thickness measurement

Using a precise spectrophotometer CARY 05E (Varian) we can determine the spectra of transmittance and absorbance of each thin film sample. From our earlier studies of the azopolymer PAZO, we know precisely the spectrum of the absorption coefficient α [Berberova et al, 2016]. In particular, in the peak of absorbance, at λ = 359 nm, α = 5.95 μm^{-1}, and in this case absorbance A = 1 corresponds to film thickness of 168 nm. In the shoulder of the absorption band, at λ = 425 nm, α = 1.59 μm^{-1}, hence A = 1 results in film thickness of 628 nm.

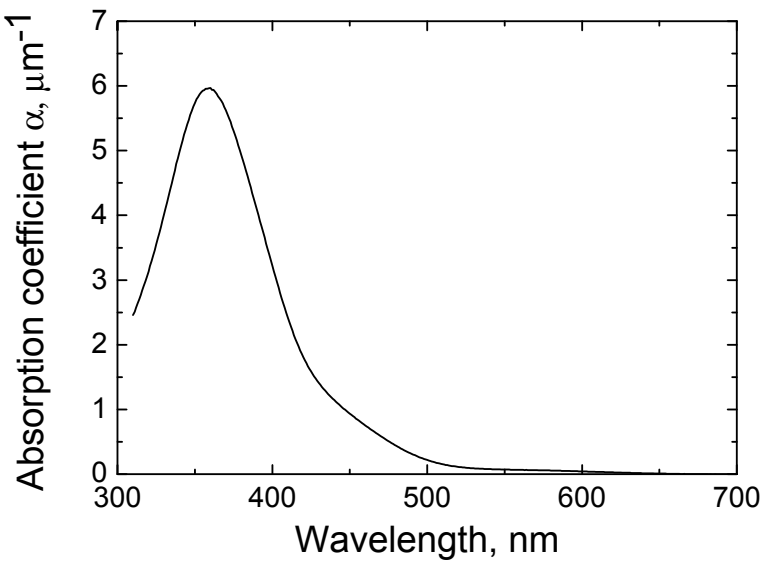
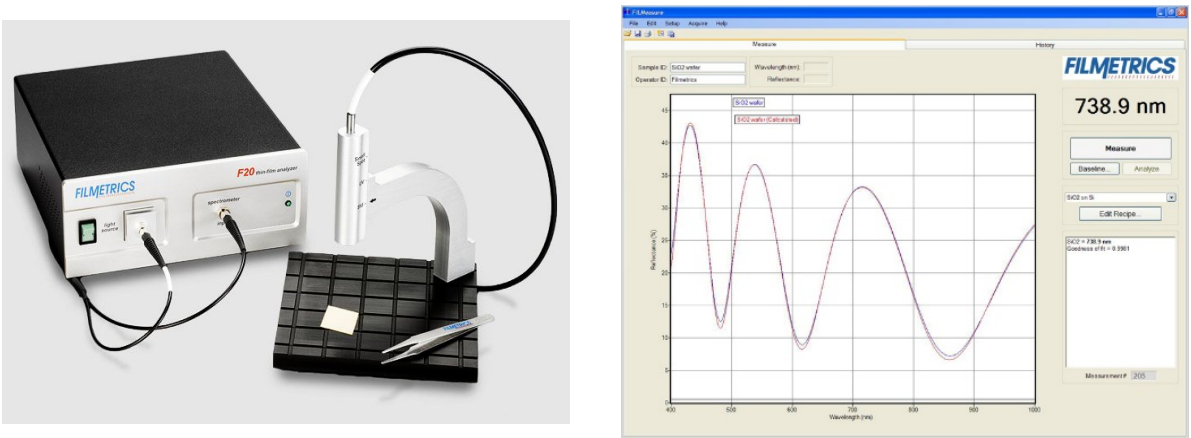


Figure 2. Absorption coefficient spectrum of PAZO (adapted from [Berberova et al, 2016]).

Therefore, measuring the absorbance of any thin film from the azopolymer PAZO at these wavelengths (λ = 359 nm or 425 nm)), we can determine its thickness, using the simple relation $d = A(\lambda) / \alpha(\lambda)$.

B. Interferometric thickness measurement

This method is based on interference of light reflected from the upper and lower boundaries of the investigated thin film. As shown in Fig. 3(b), specific oscillations in the reflectance spectrum are observed, which give information about the film thickness. We implement it, using a commercially available Thin-Film Analyzer F20 (Filmetrics). The configuration of the device is presented in Fig. 3(a).



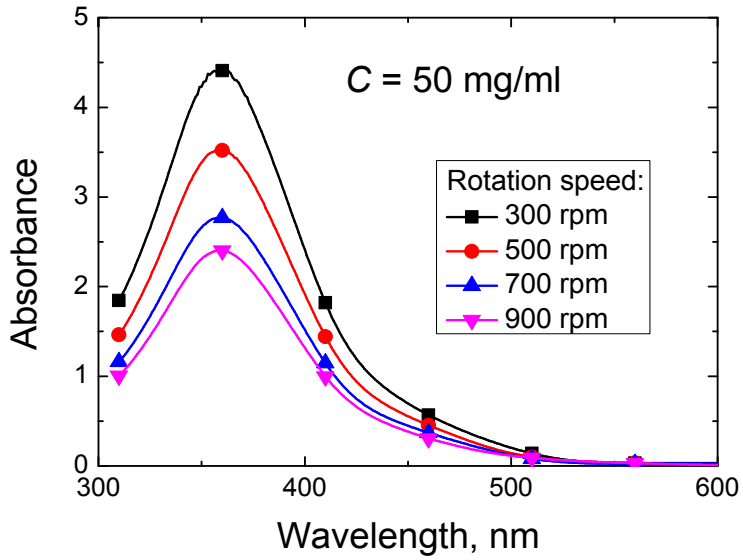
(a) (b)

Figure 3. (a) View of the F20 Thin-Film Analyzer. (b) FILMeasure software window [Filmetrics, 2007].

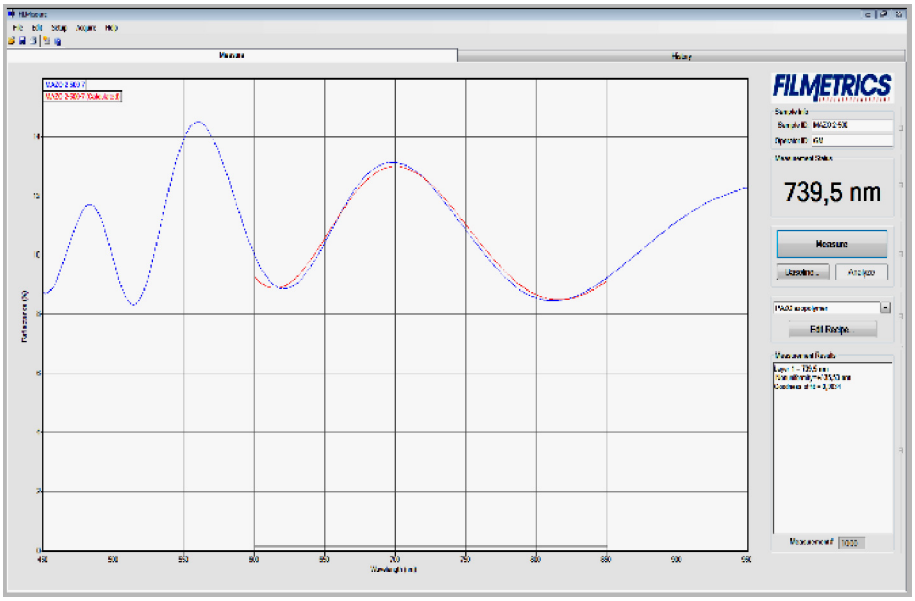
Experimental results

For this study, three series of thin films were prepared, varying the concentration of the solution of the azopolymer PAZO in methanol: C = 25, 50 and 100 mg/ml. In each series, 4 or 5 different rotation speeds were used during spin coating.

To compare the two thickness measurement methods, we have applied them for one of the series with C = 50 mg/ml. The input data are shown in Figure 4, and the results are summarized in Table 1.



(a)



(b)

Figure 4. (a) Spectral data, and (b) Interferometric data for the *C* = 50 mg/ml series.

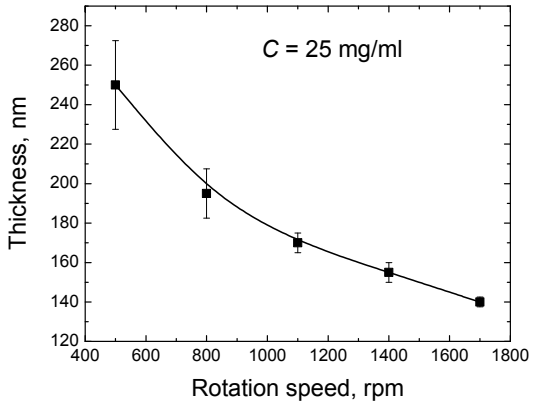
Table 1. Comparison of the spectrophotometric and interferometric method for thickness measurement

Rotation speed R, rpm	Thickness, nm (method A, λ = 359 nm)	Thickness, nm (method A, λ = 425 nm)	Thickness, nm (method B)
300	744	750	740
500	593	592	600
700	465	471	470
900	404	415	410

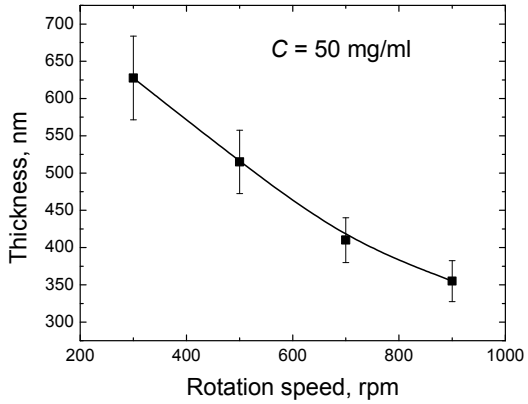
As seen, the two methods give equal values within the experimental uncertainty of 5 nm, and therefore can be used interchangeably. The data of the thickness measurement for all series of samples are presented in Fig. 5. The error bars indicate the range of thicknesses measured on a given layer.

To characterize the smoothness of the film surface we used atomic force microscopy. The scan obtained by MFP 3D Scanning Probe Microscope (Asylum Research) is shown in Fig. 6.

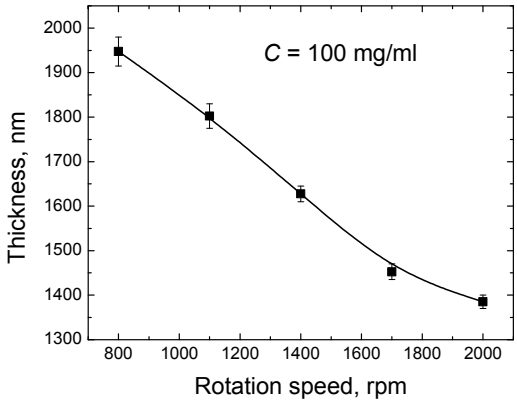
The data presented in Fig. 6 show that the surface roughness (SR) of the thin film prepared from methanol solution (SR = 8 nm) is significantly smaller than of the layer coated from water solution of the azopolymer (SR = 230 nm). In the same time, if we interpolate the curve, shown in Fig. 5(c), the thickness of film for R = 1500 rpm would be 1570 nm, as for water solution with the same concentration and rotation speed, we obtain film with thickness 450 nm [Nedelchev et al, 2015].



(a)

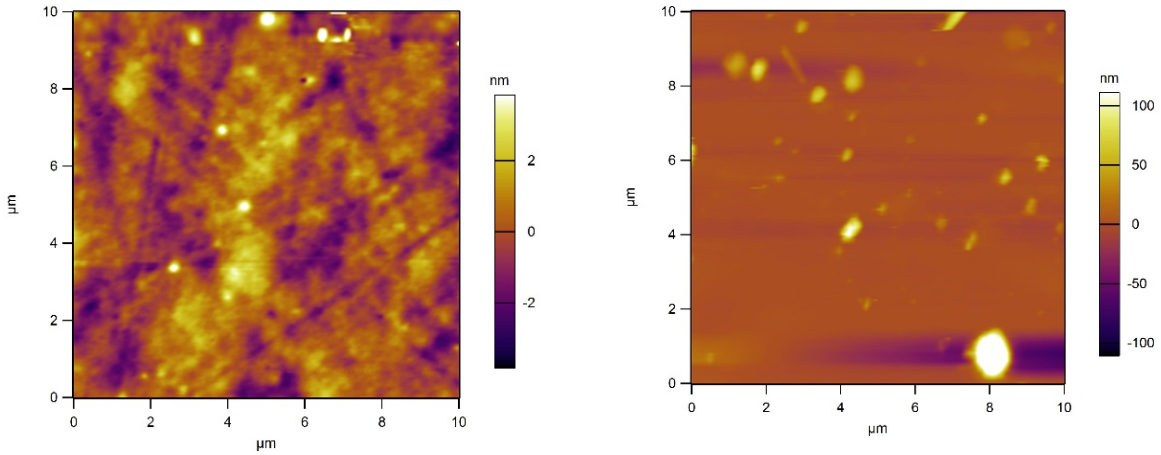


(b)



(c)

Figure 5. Dependence of the samples thickness on the concentration of the solution *C* and rotation speed: (a) *C* = 25 mg/ml, (b) *C* = 50 mg/ml, and (c) *C* = 100 mg/ml.



(a) (b)

Figure 6. AFM scan of thin film coated from (a) methanol solution with *C* = 25 mg/ml and *R* = 1700 rpm, (b) water solution with *C* = 100 mg/ml and *R* = 1500 rpm.

Conclusion

In this paper, we study the influence of two important parameters: the concentration of the solution *C* and rotation speed *R*. The results obtained for methanol solutions are compared with our previous results about thin film prepared from water solutions of the azopolymer PAZO. The following conlusions can be drawn from these experiments:

- Higher rotation speed is required to obtain films with more uniform thickness;
- Thin films prepared from a methanol solution have higher surface smoothness and in the same time higher thickness compared with the films prepared from water solution with the same concentration;
- Using this technique, films with thickness up to 2000 nm can be readily prepared. Surface smoothness is about 10 nm, or λ/50 which fulfills the requirements for optical data storage in these layers.

Acknowledgement

Authors are grateful for the financial support provided by National Science Fund of Bulgaria under the project ДН 08/10.

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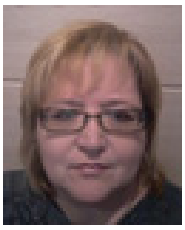

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