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Preparation of SiO_2:TiO_2 for High-Performance Double Layer Anti-Reflection Coating

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Abstract
In this work, an anti-reflection coating was prepared in the region (400-1000) nm of wavelength, with a double layer of silicon dioxide (SiO_2) as an inner layer and the second layer of the mixture (SiO_2) and titanium dioxide (TiO_2) with certain ratios, as an outer layer using the chemical spraying method with a number of 6 sprays of layer SiO_2 and 12 sprays of layer SiO_2 - TiO_2. Using the method of chemical spraying deposited on the glass as a substrate with a different number of sprays of SiO_2, and a fixed number of TiO_2-SiO_2. The optical and structural properties were determined using UV-Vis spectroscopy and atomic force microscopy (AFM). The results show that by using a mixed layer, the optimal optical performance of a broadband low-reflection ARC (anti-reflection coating) was obtained. The work also reports the comparison of experimental and theoretical results with the help of MATLAB, which relies on the characteristic matrix along the visible region – near-infrared.

Article Info.

Keywords:
Antireflection coating, interference optical filter, mix dielectric coating, multi-layer theory, optical performance.

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1. Introduction
Optical coatings are thin film layers deposited between two media in order to alter the behavior of light striking the interface of the structure. It may consist of a series of surfaces that are the boundaries between different materials. Coatings are applied to optical components intended for use at wavelength regions between UV and far-IR. The most common and important optical coatings are Antireflection Coatings (ARC), which still surpass all other filters and coatings [1]. The basic principle of such coating is reflecting specific wavelengths and largely canceling others by destructive interference and internal reflection which is related to the critical angle of the surface [2]. Antireflection coatings can range from a single layer with nearly zero reflection at just one wavelength to a multilayer system of many layers having virtually zero reflection over a broad spectral range. The performance of an optical coating depends on the number of layers, total thickness, and refractive index [3-5]. Anti-reflection coatings are often used for applications in medicine and various new applications in lenses, eyeglasses, lasers, and mirrors. Mixed materials composed of a host oxide and a small percentage of a similar oxide compound have been introduced to create a material with a desired refractive index. In addition, mixed materials promote the growth of dense film layer structures because the incorporation of the additive discourages the growth of multiple crystalline phases [6, 7]. This new mixed material can be predicted by following the Lorentz-Lorenz theory [8, 9].

This work introduces a novel wide-band anti-refraction coating (ARC) composed of a double layer that combines a thin layer of pure SiO_2 and a mixed dielectric thin layer. The mixed dielectric layer is composed of TiO_2 (n = 2.6) and SiO_2 (n = 1.46) such that ($SiO_{2(x)}$: $TiO_{2(1-x)}$) where X = 0.45. This ARC was deposited onto a glass substrate at a predetermined concentration ratio. The mixed layer was synthesized by the chemical spraying method. The SiO_2-TiO_2 mixed coatings' structure has previously been investigated [10].

Optimum results (low reflectivity) were obtained for the double-layer antireflection coating in the spectral region (400-1000) nm. A comparison was made between the experimental results and those of the theoretical work that relied on the calculations of the characteristic matrix theory. The adopted technique depends on preparing a coating consisting of a mixture of materials (SiO_2 and TiO_2) and calculating its molar refractivity according to the Lorentz - Lorenz formula. This technique helps overcome manufacturing problems and limitations in the availability of materials. This kind of ARC is used in many scientific applications and contemporary industrial settings.

2. Theoretical background

The optical performance of antireflection coatings depends on calculating the characteristics matrix to determine the spectral of reflectance (R) profile for a double layer structure on a substrate. The characteristics matrix of a composition of q thin film layers is the product of the individual matrices of the individual layers of the assembly taken in the correct order, as is given below [2]:

$$\begin{bmatrix} C \\ B \end{bmatrix} = \left(\prod_{r=1}^{q} \begin{pmatrix} \cos\delta_r & (i\sin\delta_r)/n_r \\ i n_r \sin\delta_r & \cos\delta_r \end{pmatrix} \right) \qquad (1)$$

where: $\begin{bmatrix} C \\ B \end{bmatrix}$ is the characteristic matrix of the assembly, C and B are normalized total tangential electric and magnetic fields, respectively, at the input surface, n_r is the refractive index, q is the number of layers next to the substrate, and δ_r is the phase thickness specified by the equation:

$$\delta_r = \frac{2\pi\, n_r d_r}{\lambda} \qquad (2)$$

where: d_r is the physical thickness of a layer, and λ is the design wavelength.

The reflectance (R) of the multilayer system is given as follows:

$$R = \left(\frac{n_o B - C}{n_o B + C}\right)\left(\frac{n_o B - C}{n_o B + C}\right)^* \qquad (3)$$

where: n_o is the refractive index of air (or incident medium).

3. Experimental work

In this study, dielectric materials with a large difference in their refractive index were employed for the mixed layer, specifically, ($SiO_{2(x)}$: $TiO_{2(1-x)}$) where X= 0.45.

A chemical sprayer (homemade) was used to deposit both the composite and pure layers on glass substrates at room temperature. Using the chemical approach, 10 ml of hydrochloric acid (40%) was used as the solvent to make the solution. A clear solution was obtained by combining SiO_2 solution (at a molar ratio of 0.5) with TiO_2 solution (at a molar ratio of 0.1) in a beaker and continuously swirling the mixture with a magnetic stirrer for 15-30 minutes. An ultrasonic cleaner and a mix of ethanol and deionized water were used to remove contaminants from the glass substrates' surfaces.

Different samples were prepared by spraying different numbers of SiO_2 sprays (2, 4, 6) on 12 sprays of the mixed dielectric layer, onto glass substrates. The time between sprays was 15 seconds, and the sprayer nozzle was 27 centimeters above the substrate.

To calculate the refractive index of the mixture, the Lorentz - Lorenz formula was used [8]

$$n = \left[\frac{\frac{n_1^2}{\rho_1}\left(\frac{1}{C_1}-1\right)+\frac{n_2^2}{\rho_2}\frac{a_2}{a_1}}{\frac{1}{\rho_1}\left(\frac{1}{C_1}-1\right)+\frac{1}{\rho_2}\frac{a_2}{a_1}}\right]^{\frac{1}{2}} \tag{4}$$

where: $a_1 = \frac{1}{(n_1^2+2)}$, $a_2 = \frac{1}{(n_2^2+2)}$, n_1 and n_2 are the refractive indices of the two materials, and ρ_1 and ρ_2 are their densities ($\rho_{SiO_2} = 2.65 gcm^{-3}, \rho_{TiO2} = 4.23 gcm^{-3}$) . The new refractive index was found to be equal to 1.8.

4. Optical properties

The optical performance (reflectance spectra) of a double layer was determined using UV-Vis spectroscopy, as shown in Fig. 1. This figure demonstrates that the 6:12 double layer showed the lowest reflectance. The optical performance of the ARC is less than about 2 % for the 6:12 double layer along the Visible –NIR region. This result showed the optimum performance of ARC with only a two-layer deposition. Increasing the number of sprays led to an increase in thickness, and the last led to a low reflection. This behavior is attributed to the destructive interference due to the phase difference at wavelengths (400-1100nm), as shown in Fig. 1.

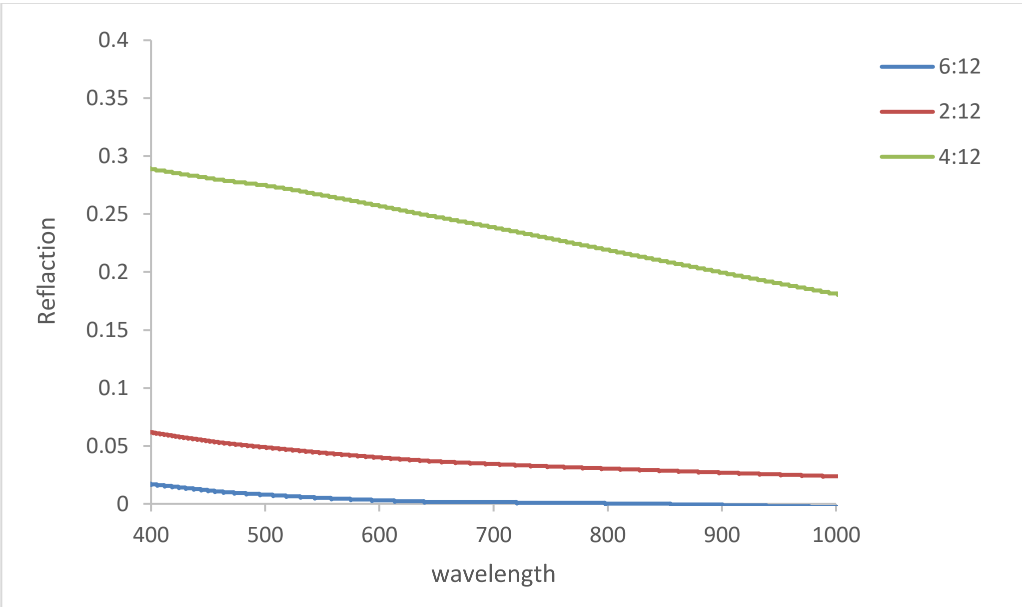

Figure 1: Reflectance as a function of wavelength for the double layer (SiO_2: Mix) samples.

The comparison between the experimental results and theoretical results is presented in Fig. 2. The theoretical results were computed using Eq. (4) for the double layer of SiO_2(n=1.46) and the mixed layer of ($SiO_{2(x)}$: $TiO_{2(1-x)}$) where (x=0.45) (n_H =1.8), using the MATLAB software. The experimental result chosen for the comparison was for the 6:12 (6 SiO_2 sprays: 12 sprays of the mix) because this sample gave the optimal optical performance. Fig.2 shows the possibility of fabricating this antireflection coating (one with two layers only) in the Visible – NIR region.

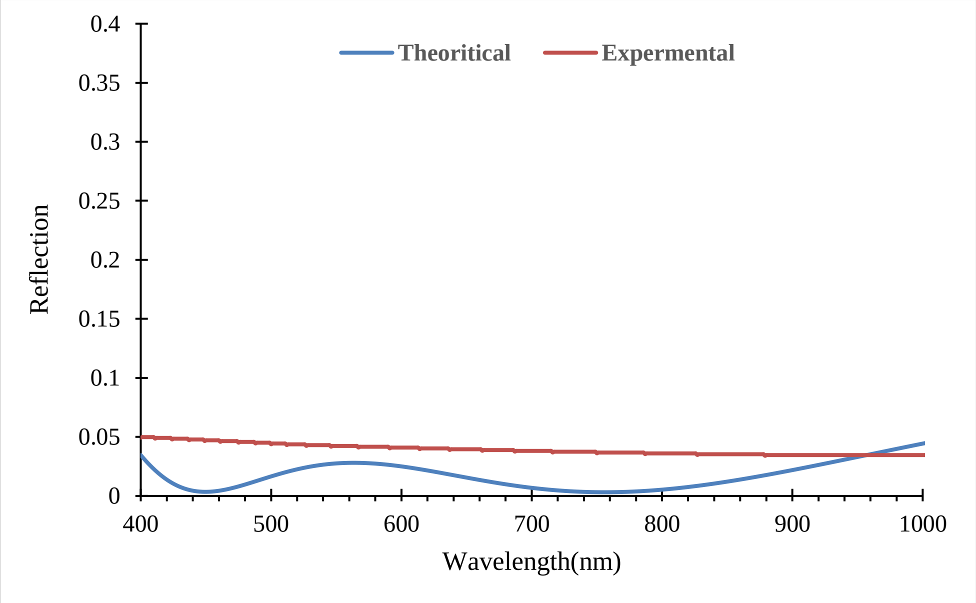

Figure 2: The comparison of the optical performance between theoretical results and experimental results for ARC.

4.1. X-Ray diffraction (XRD)

An XRD diffractometer (XRD-6000, Shimadzu) was used in a step scan mode with a measuring range of (10–80). The JCPDS card was used to measure the scan range XRD at 40 kV and 30 mA.

X-ray diffraction is an important technique for identifying crystal structure; it is used to determine the atomic arrangement, lattice characteristics, and crystalline size. The spectra of TiO_2, SiO_2, and TiO_2:SiO_2 of X=0.45 thin films prepared by the Chemical method deposition technique are shown in Fig.3(a, b, c), respectively. The diffraction peaks shown in the XRD pattern are correlated to glass (110), (004), (101), (111), (110), (200), (220), (210), (116), and (310) planes of TiO_2, the planes of SiO_2 were (003), (112), (110), (204), (012), and (013). The planes correspond to the JCPDS card no. 21-1272.

The Scherrer equation was used to calculate the crystal size. [11]:

$$D = \frac{K\,\lambda}{B\,COS\,\theta} \tag{5}$$

where: D: is particle size (cubic meter), K: shape factor of the particle, typically ranging from 0.9 to 1.2, λ: wavelength of the radiation used (meter), B: full width at half maximum (FWHM) of the diffraction peak (radians), θ: diffraction angle between the incident radiation and the sample (radians)

The peak width of a strong diffraction plane was used to calculate the values of D, which were within the nanostructure range. The average crystalline size of TiO_2, SiO_2, and TiO_2:SiO_2 are listed in Table 1.

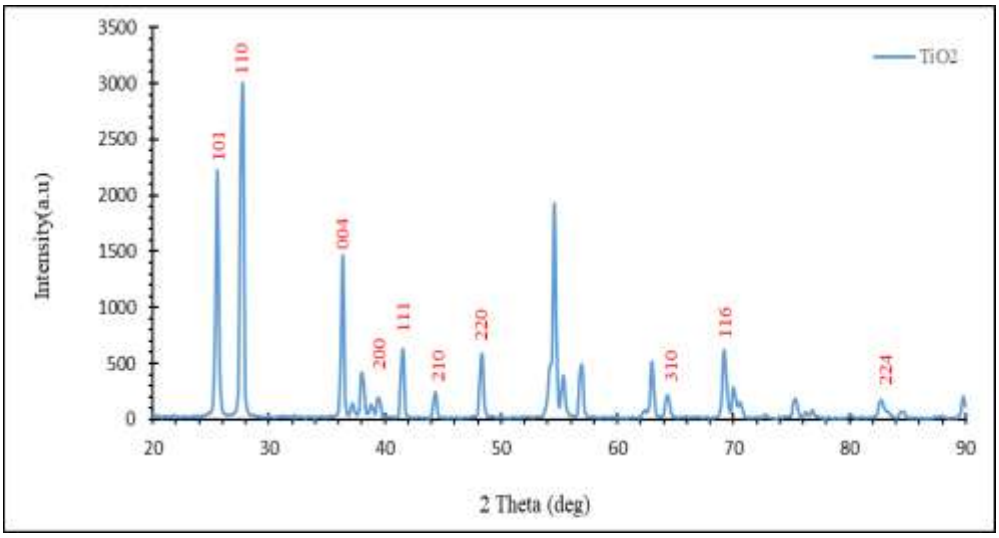

Figure 3a: XRD pattern of TiO_2.

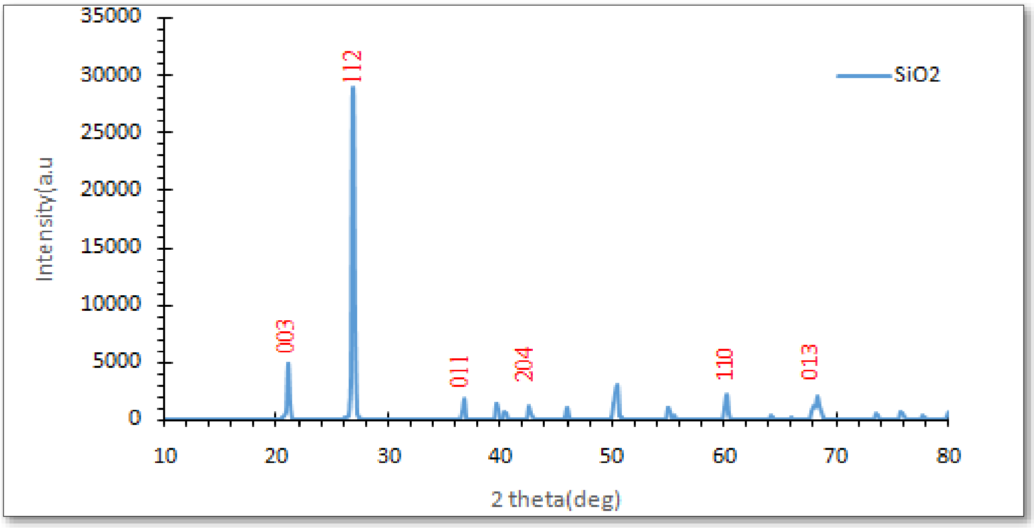

Figure 3b: XRD pattern of SiO_2.

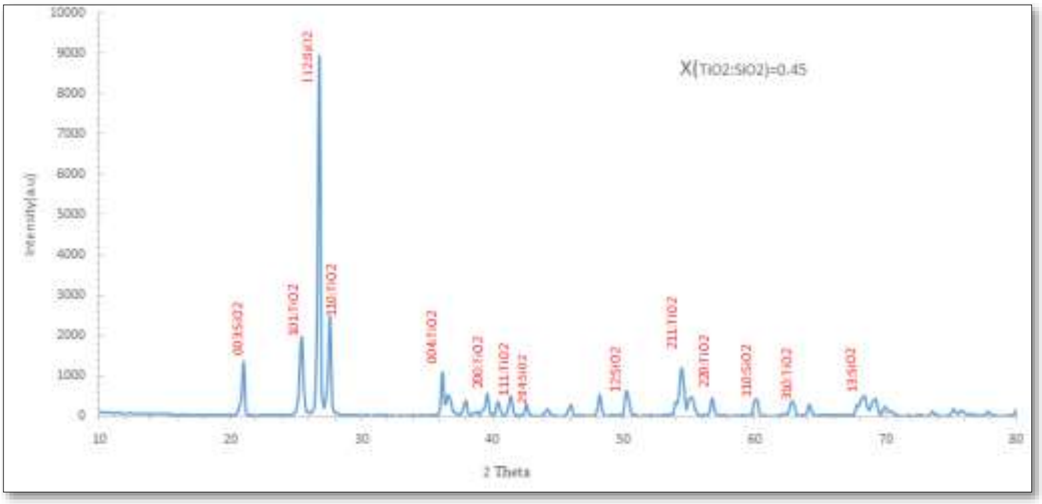

Figure 3c: XRD pattern of TiO_2:SiO_2 ($X_{(TiO2:SiO2)} = 0.45$).

Table 1: Structural properties obtained from XRD spectra of TiO_2, SiO_2, and TiO_2:SiO_2 thin films prepared by Chemical technique.

Materials	(hkl)	Crystallite size D (nm)
TiO_2	(101)	29.16874
	(110)	20.51078
	(004)	36.44492
	(111)	24.17946
	(220)	28.39092
	(211)	21.8198
	(116)	25.59208
SiO_2	(003)	20.26698
	(112)	17.79793
	(011)	27.97318
	(110)	22.99634
TiO_2:SiO_2	(003)	20.26698
	(101)	32.66898
	(112)	30.32241
	(004)	18.22246
	(200)	16.27464
	(111)	42.58639
	(012)	46.30425
	(211)	25.59208
	(110)	34.06866
	(013)	16.05057

4.2. Atomic force microscopic analysis (AFM)

The AFM three-dimensional images in Fig.4 show the surface morphology of (SiO_2: mix) thin layers deposited by the chemical deposition method on glass substrates for a different number of SiO_2 layers (2,4,6). AFM parameters (averaged diameter, root mean square, and average roughness) for these samples are listed in Table 2. From the table, it can be noted that increasing the number of sprays from 2 to 6 caused a decrease in the average diameter at the surface from 26.45 to 6.33 nm and a decrease in the average roughness from 3.70 to 1.2 nm with the increase of the SiO_2 ratio.

Table 2: AFM parameters (Avg. D, RMS, and Average Roughness) for double-layer pure and mix.

Sample	Average diameter	Roughness	RMS
2	26.45	2.58	3.70
4	19.25	1.04	1.37
6	6.33	0.93	4

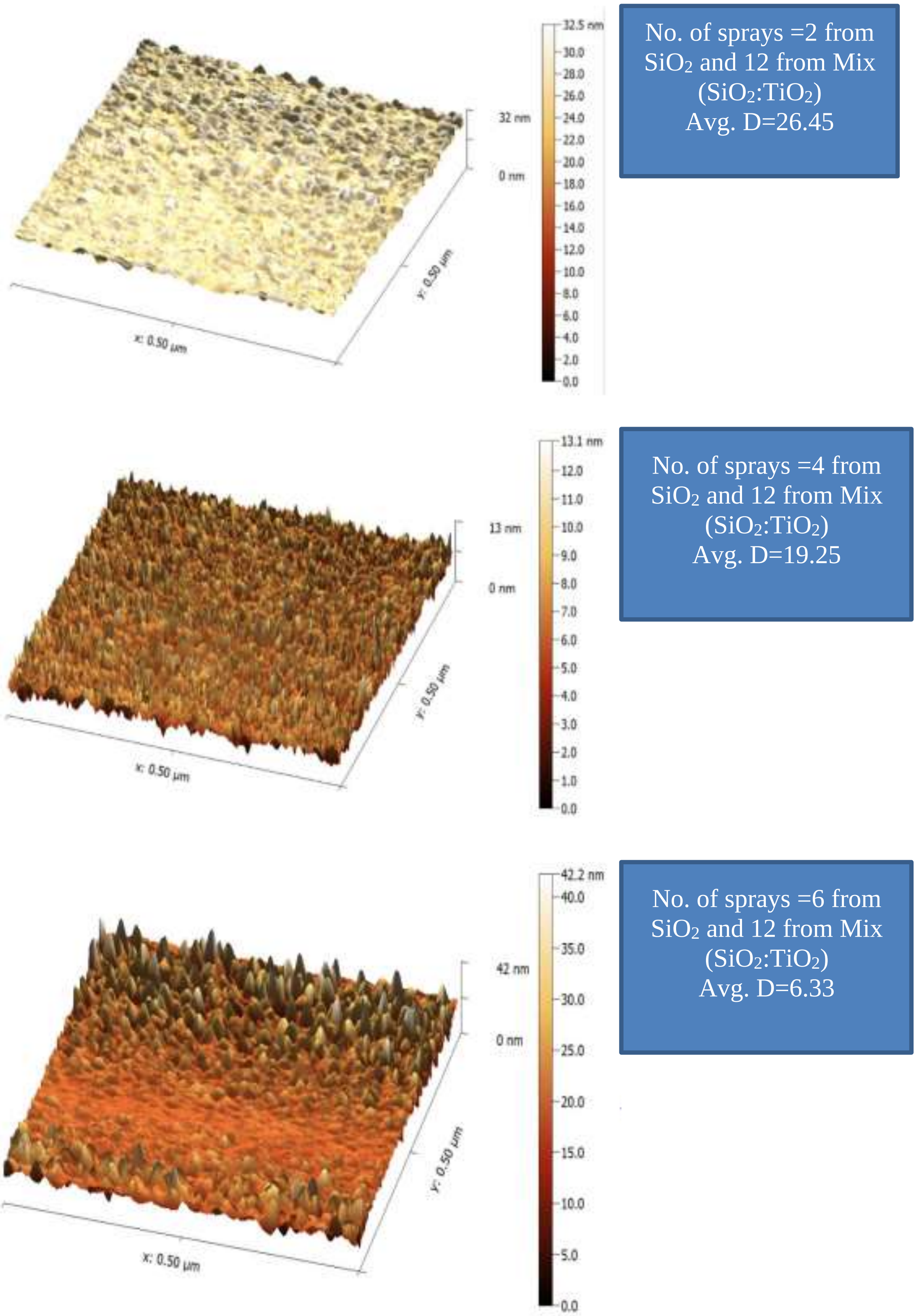

Figure 4: 3D AFM images and their granularity accumulation distribution for double layer pure and mix (TiO_2:SiO_2) composite thin films at different number of sprays.

5. Conclusions

A wide-band antireflection coating (ARC) of optimum optical performance was fabricated with only two thin layers. The achieved structure coating is characterized by low reflectivity over a wide range of the electromagnetic spectrum, which includes (VIS-NIR) regions. These coatings are used in scientific applications and advanced industries. Results showed that the high reflectance from the substrate surface decreased by adopting the coating mixture (SiO_2 and TiO_2) calculated according to the Lorentz - Lorenz formula. This technique helps overcome manufacturing problems and limitations in the availability of materials. This gives higher flexibility in manufacturing specialty optical filters in the future.

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Conflict of Interest

The authors declare that they have no conflict of interest.

References

1. G. Bao and Y. Wang, J. Optic. Soci. America A **30**, 656 (2013).
2. K. Seshan and D. Schepis, *Handbook of Thin Film Deposition*. 4th Ed. (UK, Elsevier-William Andrew, 2018).
3. J.-M. Yang and C.-Y. Kao, Proceedings of the 2000 Congress on Evolutionary Computation. CEC00 (Cat. No. 00TH8512) (IEEE, 2000). p. 978.
4. L. Li, Q.-H. Wang, D.-H. Li, and H.-R. Peng, Optic. Expre. **17**, 16920 (2009).
5. İ. G. Kavakli and K. Kantarli, Turk. J. Physi. **26**, 349 (2002).
6. B. Zhang, J. Hendrickson, N. Nader, H.-T. Chen, and J. Guo, Appl. Phys. Lett. **105**, 241113 (2014).
7. R. H. Ali, K. A. Aadim, and A. N. Abd Algaffar, J. Phys.: Conference Series (IOP Publishing, 2021). p. 012081.
8. H. G. Rashid, K. A. Mishjil, H. T. Hassim, A. N. Abd Algaffar, and N. F. Habubi, J. Phys.: Conference Series (IOP Publishing, 2021). p. 012045.
9. A. N. Abd Al-Gaffar, Bagh. Sci. J. **8**, 495 (2011).
10. A. N. Abd Algaffar and A. I. A. Al-Numan, Bagh. Sci. J. **15**, 287 (2018).
11. N. K. Abdalameer, S. N. Mazhir, and K. A. Aadim, Ener. Repo. **6**, 447 (2020).

تحضير (TiO_2-SiO_2) لطلاء مضاد للانعكاس عالي الأداء ثنائي الطبقة

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الخلاصة

في هذا العمل، تم تحضير طلاء مضاد للانعكاس في المنطقة (400-1000) نانومتر من الطول الموجي، بترسيب طبقة مزدوجة من ثاني أكسيد السيليكون (SiO_2) كطبقة داخلية والطبقة الثانية من الخليط (SiO_2) وثاني أكسيد التيتانيوم (TiO_2)، كطبقة خارجية بعدد 6 رشاشات من طبقة SiO_2 و 12 رشة من طبقة SiO_2 - TiO_2 باستخدام طريقة الرش الكيميائي. المترسبة على ركيزة زجاجية بعدد مختلف من بخات SiO_2، وعدد ثابت من TiO_2-SiO_2. بالإضافة الى تحديد الخواص البصرية والهيكلية باستخدام مطياف UV-Vis ومجهر القوة الذرية (AFM). أظهرت النتائج أنه باستخدام طبقة مختلطة، تم الحصول على الأداء البصري الأمثل لطلاء ARC ذو الانعكاس المنخفض عريض النطاق كذلك أشار الى مقارنة بين النتائج التجريبية والنظرية بمساعدة MATLAB، والتي اعتمدت على المصفوفة المميزة على طول المنطقة المرئية ـ الأشعة تحت الحمراء القريبة.

الكلمات المفتاحيه: طلاء مضاد للانعكاس، مرشح ضوئي للتداخل، مزيج طلاء عازل للكهرباء، نظرية متعددة الطبقات، أداء بصري.