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Research Article

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Evaluating the Efficiency of Some Polymer Nanocomposites in the Protection of Natural and Artificial Ultramarine-Based Oil Wall Paintings Against some Environmental Agents

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Abstract This study was carried out to monitor the effect of a polymer nanocomposite composed of Matt Picture Varnish and TiO_2 nanoparticles for the protection of both natural ultramarine blue (NUB) and artificial ultramarine blue (AUB) against one of the most influential environmental factors (UV light/thermal) ageing. It required some laboratory techniques to follow the effect of the polymer nanocomposite such as Optical microscope, Colorimetry, Fourier transform infrared spectroscopy (FTIR) and Scanning electron microscope (SEM) to achieve the aim of study through studying the chemical and morphological observations before and after (UV light/thermal) ageing. Significantly, the results of this research will help in the heritage conservation by protecting rare oil wall paintings against some of environmental agents.

Keywords Oil wall paintings, Environmental ageing, Ultramarine blue, Protective coatings, Polymer nanocomposite, Nano TiO_2

1. Introduction

Recently, selection of the most appropriate restoration materials for the antiquities to be preserved has become a necessity for preserving the cultural heritage of the countries of the world. On the other hand, this process must be carried out accurately and according to the nature of the archaeological material and the purpose of its restoration, as well as the homogeneity between its components and the components of the heritage material to be restored. Hence, the current research represents one of those study cases that focus on the use of nano-polymer composites as a material to protect the pigment Ultramarine, both natural and artificial, from some environmental ageing.

Previous researches and studies have used other materials for the purpose of protecting ultramarine, Two products (Paraloid B-72 and SILRES® BS OH100) were selected to evaluate their efficiency for consolidation treatments of painted surfaces of Hawsh al-Basha courtyard dating back to Mohammed Ali's family period (1805-1952) in Egypt Pigments including, blue synthetic ultramarine [1].

The development of nanomaterials and nanocomposites has widened their applications in various sectors. The heritage conservation has also tapped its potential as it provides a range of possibilities for the development of materials at the nanoscale and methods for conservation and restoration of cultural artefacts [2].

Polymer nanocomposites which represented the main subject of the recent research can be simply described as the new class of composite materials having inorganic nanoparticles dispersed in different polymeric matrix to improve its performance properties [3].

Practically, it provides a new and alternative way to replace the conventional filled polymers. Owing to the presence of fillers of nano scale range, the nanocomposites exhibit an extraordinary properties and characteristics when compared to the neat polymer matrix [4, 5]. These extraordinary properties of polymer nanocomposites are heat resistance, flame retardancy, mechanical strength, low gas permeability and optical/catalytic properties. The advantages of polymer nano composite are reflected on its application by the way it's not only attract the researchers but also industrials as it can be used in various applications such as energy storage, information industry, medical applications, novel catalyst and diverse environmental applications.

One of the most important environmental applications of polymer nanocomposites is to be applied for preserving the archaeological and cultural heritage objects against some weathering and environmental ageing factors such as accelerated UV and thermal ageing, depending on the a aforementioned properties of these polymer nanocomposites.

Historically, ultramarine blue (recent applicable object) is considered the most expensive blue pigment which has been used in Italy by the beginning of the fourteenth century as "Fine blue derived from lapis lazuli semi-precious stone and comes from across the seas and so is called natural ultramarine and it consists chemically of: sodium silicate, aluminum, and sulfur, and the chemical formula is ($3Na_2O.3Al_2O_3.6SiO_2.2NaS$) [6, 7]. Mineralogically, Lapis lazuli is defined by the presence of the mineral lazurite. Lazurite itself is a member of the sodalite which is responsible for its overall blue hue some other minerals were also identified with various amount with lazurite such as pyrite, calcite, diopside, forsterite, and wollastonite, Nepheline, Phlogopite [8, 9].

In Europe, the pigment probably found its most extensive use in the fourteenth to mid-fifteenth centuries [10]. The use of natural lapis lazuli as a pigment remained widespread until the beginning of the 19th century, when it appears in Shubra Palace, and El-Gawhara Palace [11].

The artificial ultramarine is called the French ultramarine as its manufacture and large production began in France, and it is also called Guimet blue derived from the name of the first person who manufactured it and spread commercially in 1830 [10].

Moreover the ultramarine blue suffers a degradation phenomenon known as ‘ultramarine disease that affected negatively on its appearance as described by Reffat et al. [11]. This phenomenon occurs in ultramarine based-oil paintings as it causes a color change which converted its brilliant blue hue to pale gray without any change in the chemical composition as a result of a reaction between ultramarine with the oil binder.

The current research attempted to apply some polymer nanocomposites to enhance the efficiency of the Matt Picture Varnish which used in the surface coating protection of the ultramarine color for both natural and artificial ultramarine pigment, on the other hand, to achieve the most suitable compositions that allow to preserve the two types of ultramarine against some environmental ageing factors that affect it negatively, such as UV light and thermal ageing, consequently to preserve it for a longer period of time.

2. Materials and method

2.1. Materials

2.1.1. Pigments

Ultramarine blue of artificial dark ultramarine ((AUB) sodium alumina sulfo silicate) [12], while the natural ultramarine ((NUB) sodium alumina silicate contains sulfur with the chemical formula ($3NaAlSiO_4.NaS_3$) [13] were used. The information of all data of the natural and artificial ultramarine pigment was obtained from Kermer pigment German Company.

2.1.2. The protective coating (polymer nanocomposite)

The used polymer nanocomposite composed of two main components, the first one is a product called Matt Picture Varnish which is composed of transparent ready-to-use varnish based on acrylic resin and polyethylene wax (Pébéo Company, France) [14,15] and the second one is TiO_2 represents the nanometric filler. One of the

most popular nanomaterials that has the ability to be applied in a various fields of applications as medical, photo-catalysis and surface coating for culture heritage application. In the recent research, nano TiO_2 represents the nanometric component of the polymer nano composite with a size less than 25 nm which is white powder, spherical shape, anatase phase, well assembled, purity~ 99.9% and it was obtained from Nano Gate company, Cairo, Egypt. The produced polymer nano composite (Matt Picture Varnish+nano TiO_2) is used as innovative protective coating material for ultramarine blue against some degradation agents [16].

2.1.3. Laboratory models

a) Preparation of the experimental study samples:

Cubes of gypsum 2.5*2.5 cm were prepared for applying the colored materials of Natural Ultramarine Blue (N.U.B) and Artificial Ultramarine Blue (A.U.B) after mixing them with a binder of linseed oil, and the samples were left until drying completely and the varnish coat was applied on them.

b) Preparation of final protection hybridized nanocomposites materials:

Three concentrations of the studied polymer nanocomposites (varnish+ nano titanium dioxide 1%, 2% and 3%) were prepared and used as final protection material for both (N.U.B) and (A.U.B). The preparation of this final protection material was prepared by dissolving in toluene 3%, the process of mixing nanomaterials with the required concentrations was done through a hot plate and stirrer device and the stirring time lasted about an hour at a speed of 750 rpm, and applied over varnish coat used in the study and the codes of the used materials were for both (N.U.B) and (A.U.B) as follows:

1. Matt Picture Varnish without nanomaterial with code (V).
2. Matt Picture Varnish hybridized with 1%nano TiO_2 nanoparticle with code (VT1).
3. Matt Picture Varnish hybridized with 2% nano TiO_2 nanoparticle with code (VT2).
4. Matt Picture Varnish hybridized with 3% nano TiO_2 nanoparticle with code (VT3).

2.2. Methods and instruments

Some laboratory techniques were used as helpful methods to achieve the aim of the recent research as follows:

2.2.1 Artificial ageing protocol

Accelerated thermal and photochemical degradation is usually used to determine the stability of polymeric materials and polymer nanocomposites. UV light ageing process was occurred by exposing the tested samples to ultraviolet (UV) rays, where the exposure measurements were made in the UVA, UVB range for a period of (120) hours, and four UVA lamps and two UVB lamps were used (Fig.1), and the study was conducted through an exposure room at the institute National Measurement and Calibration of the Ministry of Scientific Research.

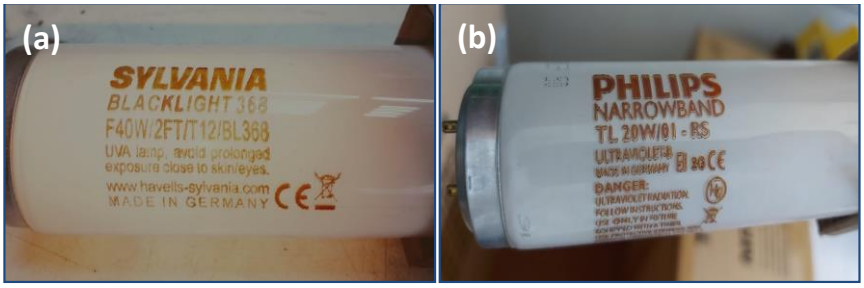

Fig.(1) (a) UVA Lamp, Sylvania, Black Light 368, F 40w/2FT/T12/BL368.
(b) UVB Lamp, Philips Narrow band, TL 20w/01-RS.

Thermal aging processes were carried out by placing the tested samples in a convection oven dried at a temperature of 70 ℃ for 3 days, and then the temperature was raised to 100 ℃ for a period of 10 days. Blackshaw and Ward (1983) stated that in the case of exposure to a temperature of 60 ℃ for a day It is equivalent to 14 days in nature, and in the case of exposure to a temperature of 70 ℃ for a day, it is equivalent to 27 days in nature, and in the case of exposure to a temperature of 100 ℃ for a day, it is equivalent to 157 days in nature. The study was conducted at the National Institute for Measurement and Calibration of the Ministry of Scientific Research.

2.2.2. Optical microscopy

Microscopic observations were used as a helpful tool to investigate and detect any changes in the appearance of the painted models. The images were obtained using a Veho VMS-004-DELUXE USB digital microscope, with a maximum magnification of 100x.

2.2.3. Colorimetry

Colorimetry is a common method usually used to evaluate the chromatic changes in the field of works of art, cultural heritage, and their preservation, such as: oil paintings, murals, and stonework, especially for comparisons made after the accelerated ageing (photo and thermal). The CIE L*a*b*color values were measured using a Macbeth Color Eye 3100 laboratory Spectrophotometer unit Module CE 3100using D65 illuminant and 10° observer. The overall color difference (ΔE*) (CIELAB, 1976) between the initial measurements and those taken after exposure was calculated using the equation (according to ASTMD2244-16 [17]):

$$\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$$

Where:

ΔL* - the lightness difference: ΔL*=L*1-L*2

Δa* and Δb* - chromaticity differences: Δa*=a*1-a*2, Δb*=b*1-b*2

L*= refers to Lightness (L= 0 indicates black and L=100 white),

a*= expresses the hue between red and green (negative a*values indicate green, and the positive ones for red),

b*= refers to the hue position between yellow and blue (negative b* values are associated with blue, and the positive values for yellow).

2.2.4. Fourier-transform infrared spectroscopy (FTIR)

The molecular structure of the studied samples was determined using a Jasco 4100 FTIR Spectrometer. Few milligrams of each model sample were removed and mixed with KBr powder to prepare the needed discs for analysis. The transmittance FTIR spectra were collected in the mid-IR region (400–4000 cm^{-1}) with accumulation of 32 scans at a resolution of 4 cm^{-1}. Essential® FTIR Spectroscopy Software (version 3.1) was applied to measure and handle the recorded Spectra.

2.2.5. Scanning electron microscope (SEM)

SEM investigation considered as a guide in measuring any alterations in the morphological features of the samples due to ageing. For this purpose a Quanta scanning electron microscope FEG 250 (Field Emission Gun, FEI, Netherlands) was used. The instrument was operated using an accelerating voltage of 20 kV and magnification of 2000x.

3. Results and Discussion

The obtained results and their interpretations were carried out to follow the effect of treatment for both of standard (NUB and AUB) samples and by a final protective material (before and after) subjecting them to light and thermal ageing processes.

3.1. Microscopic observations

Some microscopic observations were gained from the optical microscope images (Fig. 2). Firstly, there is a difference between standard sample particle sizes. Secondly, for both studied samples (NUB and AUB) no evidence was detected for change in color and morphological changes by light ageing. On the other hand, the thermal ageing effect leads to color change for both (NUB and AUB) samples which observed after thermal ageing.

It should be noticed that the optical microscope in the current research used only to differentiate between standard NUB and AUB before and after light/thermal ageing. Moreover, detailed morphological observations

to differentiate between the studied treated concentrations before and after light/thermal ageing required colorimetric and SEM analysis.

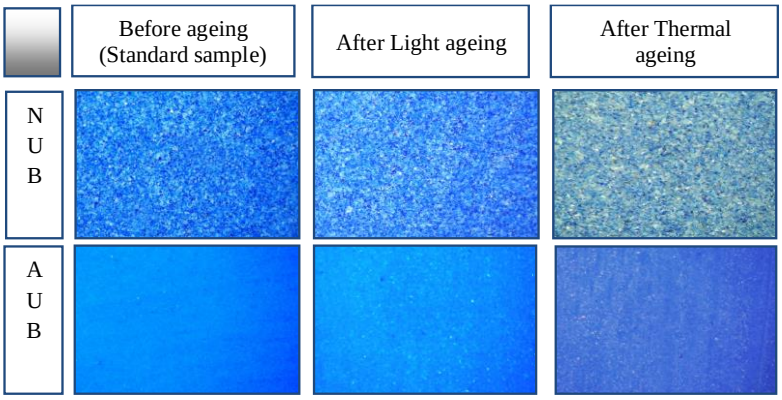

Figure 2: Optical microscope photomicrographs of the studied samples for both standards (NUB and AUB) samples and after (light and thermal) ageing

3.2. Colorimetric evaluation

This sensitive and most important analysis was carried out to all natural and artificial ultramarine blue samples before and after adding varnish and nano TiO_2 (1%, 2%, 3%). The obtained results from colorimetric analysis after light and thermal ageing exhibited the following:

1. The results of NUB samples after light ageing processes are shown in Table (1) and represented graphically by (Fig 3). NUBV samples achieved a decrease in the color change value (ΔE) to reach 2.42 compared to the standard sample NUB 3.56. NUBVT1 sample achieved the best obtained result that decreased to be 1.77.
2. By comparing the results of NUB samples after light ageing processes Table (1) with the results of AUB samples that represented at Table (2), the color change value of the standard AUB sample increased to be 4.82. AUBV samples also recorded a decrease in the color change value to reach 3.86, and the treated samples (AUBVT) 1%, 2% achieved the best obtained results, that decreased to be 2.95, 3.22 respectively.
3. The results of NUB samples after thermal ageing processes are shown in Table (3) and represented graphically by (Fig. 3). NUBV achieved a decrease in (ΔE) to reach 8.59 compared to the standard sample NUB 15.91. The obtained results of the treated samples with protective nanocomposites material didn't achieve acceptable results.
4. Comparing the results after thermal ageing processes of NUB samples with the results of AUB samples after thermal aging processes Table (3) and represented graphically by (Fig. 4). The color change value (ΔE) of the standard NUB and AUB samples were increased to be 15.91 and 18.15 respectively, and for the treated samples with varnish materials NUBV and AUBV samples achieved the best results, that decreased to be 8.59 and 9.18 respectively and the treated samples AUBVT 1%, 2%, 3% didn't achieve acceptable result sand increased to be 17.57, 17.75, 17.88 respectively.

According to the colorimetric measurements the optimum concentration was 1% of nano TiO_2 for both treated natural ultramarine blue and artificial ultramarine blue which led to the best obtained results so for detailed studies SEM based on this percentage.

3.3. FTIR analysis

It was carried out to the samples of natural and artificial ultramarine blue with protective materials (varnish + nano TiO_2 3%) before and after the light/thermal ageing process (Fig.5). The nano TiO_2 3% concentration was chosen in this analysis to measure the highest effect on the molecular structure of both natural and artificial ultramarine blue. Based on FTIR results, the laboratory specimens of natural ultramarine showed characteristic peaks as follows: O–H stretching appeared in NUB, NUBV and NUBVT3 at the region between (3610–3640 cm^{-1}), C–H stretching appeared in NUB, NUBV and NUBVT3 at the region between (2800–3000 cm^{-1}), C=O stretching appeared weak in NUB but appeared strong in NUBV and NUBVT3 due to the addition of varnish (resin) that at the region between (1750–1730 cm^{-1}). C–H bending appeared in NUB, NUBV and NUBVT3 at

the region between (1000–1152 cm^1). The infrared spectra remained almost unchanged throughout the thermal ageing. The infrared spectra of the NUBV and NUBVT3 remained almost unchanged throughout the thermal ageing, with a little decrease in the absorption intensity of carbonyl bond recorded at the final stages of ageing, indicating minor oxidative degradation of the material [18-19]. After light ageing, the chemical stability has been remained unaffected. Based on FTIR results, the laboratory specimens of artificial ultramarine blue showed characteristic peaks as follows: O–H stretching appeared in AUB, AUBV and AUBVT3 at the region between (3437–3444 cm^{-1}), C–H stretching appeared in AUB at the region between (2928–2930 cm^{-1}), C=C stretching appeared in AUB, AUBV and AUBVT3 at the region between (1631–1640 cm^{-1}) which the main character of the artificial ultramarine blue, C=O stretching appeared weak in AUB ,but appeared strong in AUBV and AUBVT3 due to the addition of varnish (resin) that at the region between (1738–1741 cm^{-1}). C–H bending appeared in NUB, NUBV and NUBVT3 at the region between (1000–1152 cm^{-1}). By studying the treated samples before and after the light/thermal ageing processes, the results showed that there was no change in the functional groups in the natural and artificial ultramarines; thus, it was concluded that no critical alterations were reported for the polymeric materials after the UV light/thermal ageing.

Table 1: Natural ultramarine blue samples treated with final protective materials after light **ageing**

Sample	Before ageing			After Light ageing			ΔE	CIE76
	L	a	b	L	a	b		
N.U.B	42.09	3.78	-35.47	44.15	5.23	-37.98	3.56	
N.U.B.V	34.12	5.73	-38.04	36.50	6.19	-38.04	2.42	
N.U.B.V.T1	29.22	7.32	-35.02	30.49	7.90	-36.11	1.77	
N.U.B.V.T2	28.18	7.16	-34.21	30.09	7.42	-35.52	2.33	
N.U.B.V.T3	29.14	6.82	-36.59	30.24	7.60	-38.63	2.45	

Table 2: Artificial ultramarine blue samples treated with final protection materials after light ageing

Sample	Before ageing			After Light ageing			ΔE	CIE76
	L	a	b	L	a	b		
A.U.B	37.09	16.99	-57.66	38.76	20.05	-60.99	4.82	
A.U.B.V	32.23	24.44	-61.26	33.23	21.26	-59.31	3.86	
A.U.B.V.T1	30.63	24.92	-59.78	31.23	22.83	-57.79	2.95	
A.U.B.V.T2	27.16	23.80	-51.07	28.45	21.13	-49.82	3.22	
A.U.B.V.T3	26.13	18.62	-43.20	25.77	20.63	-46.90	4.23	

Table 3: Natural ultramarine blue samples treated with final protective materials after thermal ageing processes

Sample	Before ageing			After Thermal ageing			ΔE	CIE76
	L	a	b	L	a	b		
N.U.B	42.09	3.78	-35.47	40.00	-3.14	-21.30	15.91	
N.U.B.V	34.12	5.73	-38.04	30.50	1.22	-30.76	8.59	
N.U.B.V.T1	29.22	7.32	-35.02	26.09	-0.59	-21.91	15.91	
N.U.B.V.T2	28.18	7.16	-34.21	27.90	-0.55	-21.05	15.27	
N.U.B.V.T3	29.14	6.82	-36.59	28.05	-0.33	-21.47	16.76	

Table 4: Artificial ultramarine blue samples treated with final protective materials after thermal ageing.

Sample	Before ageing			After Thermal ageing			ΔE	CIE76
	L	a	b	L	a	b		
A.U.B	37.09	16.99	-57.66	34.14	6.54	-43.12	18.15	
A.U.B.V	32.40	24.74	-61.57	31.09	18.11	-46.16	9.18	
A.U.B.V.T1	30.63	24.92	-59.78	25.28	10.07	-52.06	17.57	
A.U.B.V.T2	27.16	23.80	-51.07	21.02	9.02	-43.40	17.75	
A.U.B.V.T3	26.13	18.62	-43.20	21.00	3.79	-34.63	17.88	

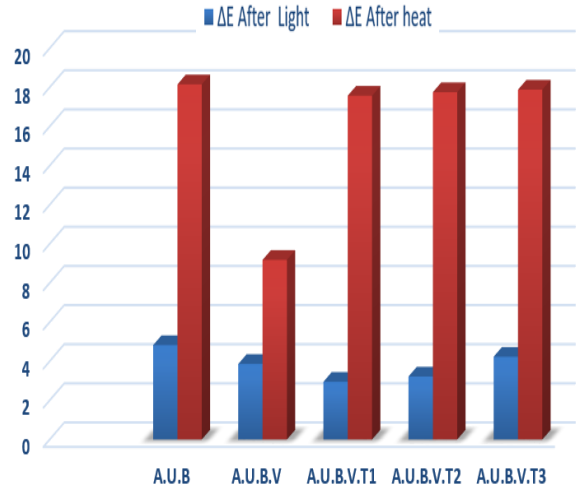

Figure 3: The relationship between the color change values of natural ultramarine blue samples treated with final protective materials after light and thermal ageing

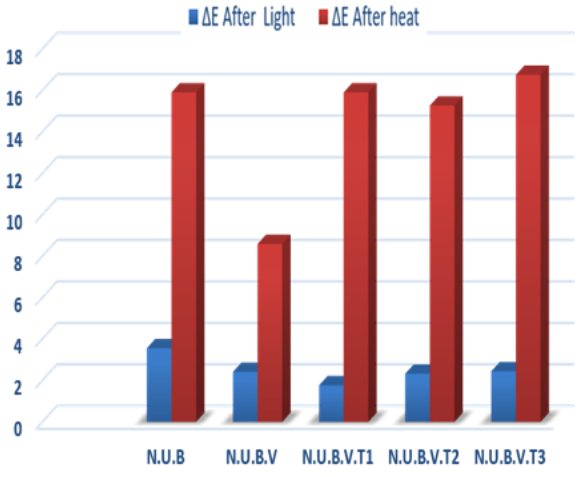

Figure 4: The relationship between the color change values of artificial ultramarine blue samples treated with final protective materials after light and thermal ageing

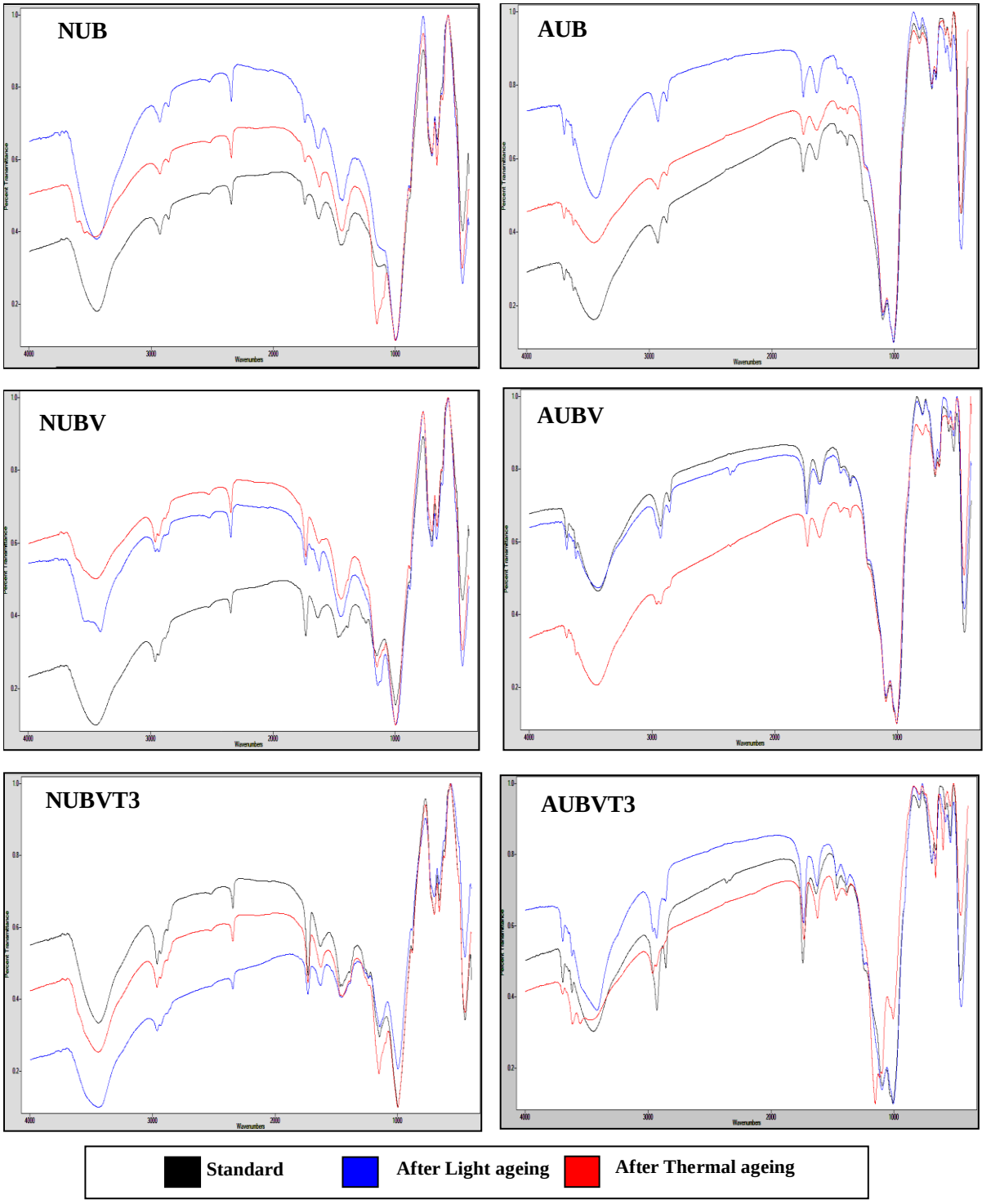

Figure 5: FTIR spectra of standard (NUB, AUB), treated with varnish and treated with nanocomposite material samples before and after light/thermal ageing

3.4. SEM morphological investigation

Both standard natural and artificial ultramarine blue samples and treated (varnish and varnish + TiO_2 1%) samples were showed respectively by SEM photomicrographs (Figs 6, 7). The SEM photomicrographs indicated the following: Positive effect for adding recent polymer nanocomposites (varnish + TiO_2) as it can be noticed

that the percent of the pores between granules decreased gradually from NUB and AUB to NUBV and AUBV and finally the most filled pores occurred in the NUBVT1 and AUBVT1 as shown in (Figs 6, 7). It can be observed also that after only varnish addition (NUBV and AUBV) some deformations occurred on the surface resulted by light/thermal ageing appeared due to the partial coating of the varnish over the ultramarine blue as illustrated in (Figs 6,7). The SEM obtained results explained by nano scale titanium dioxide as filler in the polymer nanocomposites samples NUBVT1 and AUBVT1 exhibited extraordinary properties on the surface after light/thermal ageing.

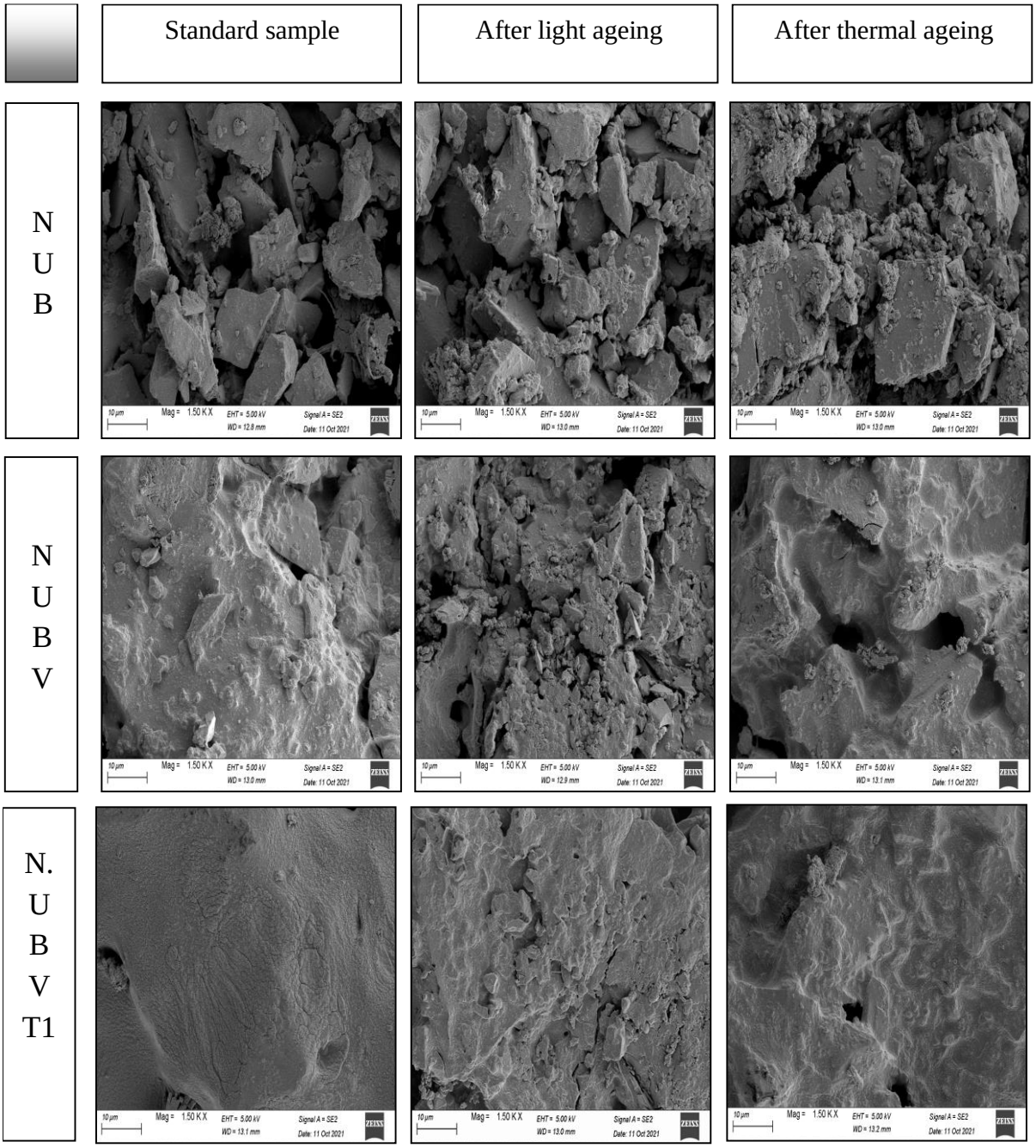

Figure 6: SEM micrographs of the standard NUB and both of the treated samples NUBV and NUBVT1 before and after light/thermal ageing processes at 1500x magnification

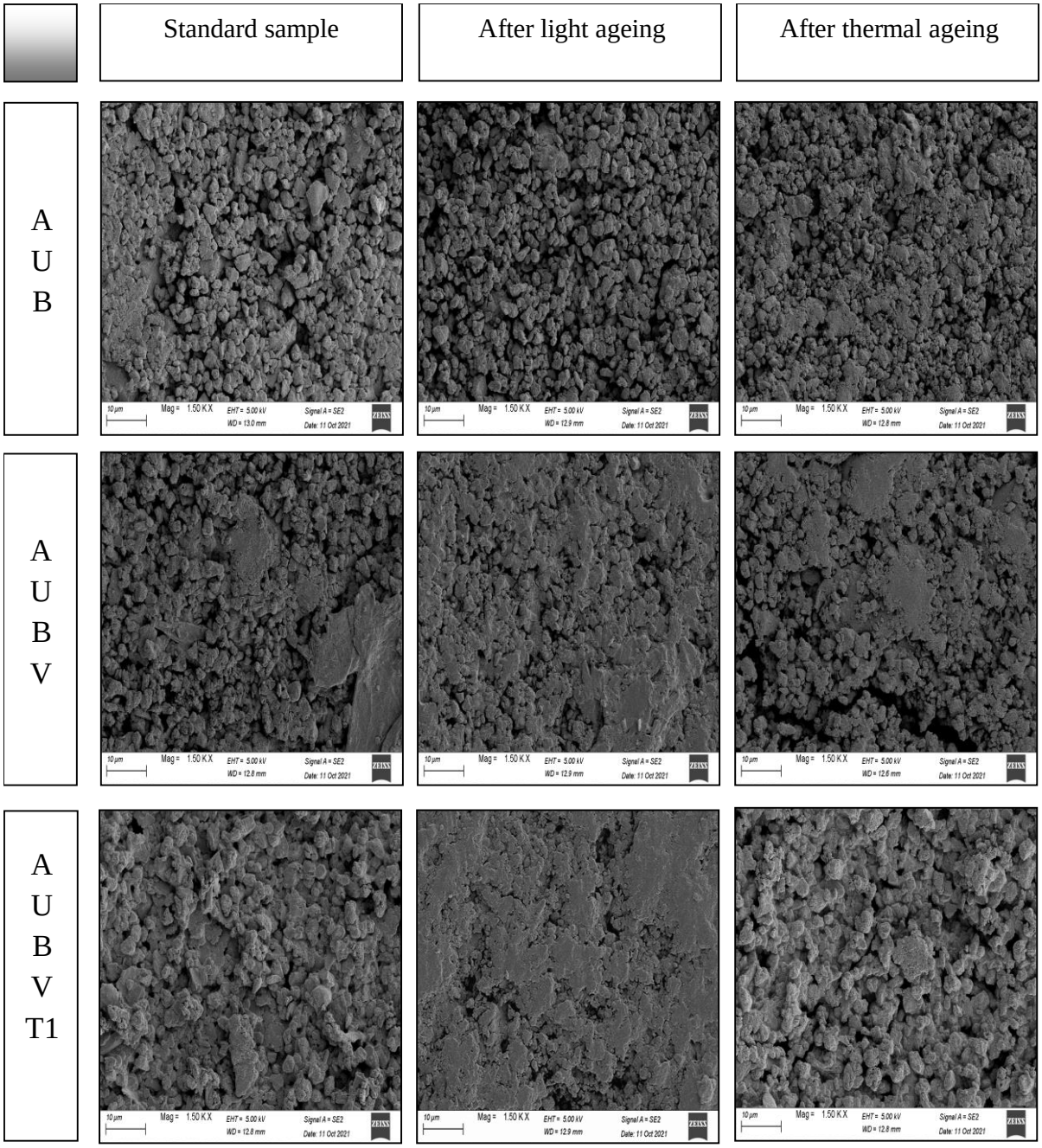

Figure 7: SEM micrographs of the standard AUB and both of the treated samples (AUBV) and (AUBVT1) before and after light/ thermal ageing processes at 1500x magnification

4. Conclusions

Studied Polymer nanocomposite (varnish+TiO_2) as a protective coating material gives extraordinary characteristics in the case of compare with polymeric matrix (varnish) as presence of TiO_2 (nano filler) leads to increase the ability of the surface penetration of the polymer nanocomposite and sequential increase its resistance against environmental ageing. The main parameters used to follow the changes occurred by the application of varnish and polymer nanocomposite for both AUB and NUB to evaluate its stability against light and thermal ageing were microscopic examinations, molecular analysis, color measurements and surface morphology. According to the colorimetric analysis the addition percentage 1% of TiO_2 for both NUBVT1 and AUBVT1 samples achieved the lowest value of ΔE results which reflected in the positive results after light

ageing, and on the contrary, they did not achieve good results after thermal ageing but NUBV and AUBV achieved the lowest value of ΔE results which reflected in the positive results after thermal ageing processes. FTIR results showed that there was no change in the functional groups of the treated samples after light and thermal ageing, thus, no chemical change occurred. SEM photo micrographs showed the positive effect of polymer nanocomposite (varnish + TiO_2) as it can be noticed that the percentage of pores between granules decreased gradually from NUB and AUB to NUBV and AUBV and finally the most filled pores occurred in the NUBVT1 and AUBVT1.

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