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DETERMINING SPECIAL FEATURES IN THE TOPOGRAPHY OF PAPER WITH WATER MARKS AT THE MICRO- AND NANOLEVELS

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Досліджено поверхневу структуру захищеного від підробки паперу із позитивними і негативними водяними знаками при різних масштабах оцінювання – від мікрометрів до рівня нанометрів, на основі методів контактної профілометрії та атомно-силової мікроскопії. Було виявлено, що структурні параметри поверхні захищеного паперу є неоднаковими для ділянок із водяними знаками та без.

Результати вимірювання контактним профілометром дали змогу прослідкувати кореляцію між значеннями середнього арифметичного відхилення профілю та наявністю водяних знаків. Значення найбільшої висоти профілю не залежить від ділянки вимірювання, що може бути пов'язано із хаотичним розміщенням волокон та частинок наповнювача у паперовій масі, що виступають над лінією профілю.

Аналіз значень середнього арифметичного відхилення профілю, отриманого методом атомно-силової мікроскопії показав, що для ділянок із негативними водяними знаками воно є більшим, ніж для ділянок із позитивними водяними знаками. Залежність найбільшої висоти профілю від ділянки вимірювання не прослідковується чітко. Це може бути пов'язано з тим, що розмір розглянутої ділянки (3000×3000 нм) охоплює лише частину волокна целюлози.

Результати проведеного аналізу показали придатність обох методів для оцінки специфічних характеристик поверхні паперу, що визначає характер взаємодії даного виду паперу із друкарськими фарбами в процесі друку. Метод профілометрії дає змогу визначити параметри профілю поверхні паперу, сформованого певним чином, а метод атомно-силової мікроскопії дозволяє аналізувати морфологію його компонентів, що розміщені на поверхні (волокна, частинки наповнювача, тощо). Інформація про структурні властивості паперу дає змогу прогнозувати якість поліграфічного відтворення, зокрема чіткість відтворення тонких гільйошних ліній на водяних знаках, що є актуальним, оскільки попереджає відбракування поліграфічної продукції спеціального призначення

Ключові слова: захищений папір, шорсткість, контактна профілометрія, атомно-силова мікроскопія, поверхнева структура паперу

1. Introduction

One of the key factors for ensuring quality in high-quality printing of protected printed products is the manner in which paper and ink manifest their physical and technical properties.

Paper represents a complex structure consisting of interlaced fibers, particles from the filler, and supporting supplements. Structural properties of paper directly affect the quality of printing reproduction, thereby largely determining the interaction with inks.

Most types of paper for intended for printing of documents of strict accountability and securities include watermarks (WM) formed on a paper canvas in the process of its production. The presence of watermarks leads to heterogeneity of the surface and volumetric characteristics of paper, which can affect the optical density and color of the imprint, clarity of reproduction of fine guilloche lines.

Given the heterogeneity of the paper canvas, it is a relevant task to undertake a research aimed at identifying the structural differences of its adjacent areas, which would make it possible to predict the quality of printing reproduction.

2. Literature review and problem statement

Studying the structural characteristics of paper using different methods, as well as the impact of these characteristics on printing quality, has been the focus of attention by both experts in standardization [1, 2] and scientists [3] for a long time.

WM is one of the most widely used protective elements of securities, including bank notes [4–6], which is why many researchers in their studies address general trends in the development of paper with WM [4, 5], the durability of paper [7, 8], the electrical-physical properties of paper [9, 10], as well as the quality of printing reproduction on it [11, 12].

Works [13, 14] explored the depth of penetration of an offset ink into the depth of paper with WM and its surface structure, however, the study of the surface involved the macro level only as it was based on results from a mechanical profilometer.

Research [14] has been further advanced in [15] by applying the actual results and by having established the fractal geometry of the structure of paper with WM. Since the work employed the results from the microlevel only, the description of the surface requires a more detailed study at higher resolution.

Work [16] reports results of research into paper surface using different methods (optical microscope, laser profilometer, scanning electron microscope, AFM). It was concluded that all the applied methods are appropriate and make it possible to analyze in more detail the surface of paper; the study, however, did not address the paper with WM that has certain special features.

Results from studying the surface of coated, uncoated and laminated paper by the method of atomic force microscopy (AFM) are reported in [17]. It was established that this method is suitable to control quality of paper surface that is homogeneous in its nature. However, the paper with WM, used to print securities, while having the same composition of areas with WM and outside them, differs in that WM form due to the different density of fibrous composition thereby creating significant heterogeneities. Thus, the issue related to the possibility of studying the surface structure of tamper-proof paper with WM by AFM method remains unresolved.

In study [18], parameters for the roughness of paper surface were determined by the Parker method and by using AFM. Results from the reported methods were intercorrelated, which allowed the authors to argue about the possibility of using AFM for operative control over the processes of surface treatment of paper. However, it should be noted that the Parker method implies measuring the air flow rate between the surface of the paper and an analyzer [2], which is why, when used for the areas of paper with WM and outside of them, the results would be averaged, thus questioning the appropriateness of using AFM method for papers with macro-irregularities of systematic order.

Work [19, 20] analyzed the influence of morphology of the surface of several types of paper on the uniformity of print by a digital technique. The authors used coated and uncoated paper whose surface characteristics were investigated by AFM method. The study has made it possible to correlate data acquired by AFM and the uniformity of print, but the paper with WM was not investigated, for which the uniformity of print at adjacent areas is a relevant issue.

Study [21] applied AFM method to examine the topography of paper's surface (without surface coating, with kaolinitic and calcium carbonate coating), as well as the penetration and distribution of pigments in UV-ink by using a confocal laser scanning microscope. The results obtained by AFM showed that the surface of the paper covered with a layer of kaolin is the most even; the size of pores at the surface of layer of calcium carbonate is less than that for kaolinitic; the size and depth of pores in the paper without a coating are the biggest among the samples. However, this work does not apply to tamper-proof types of paper with WM for which the depth of ink penetration directly affects the quality of color reproduction.

Therefore, it is a relevant task to study the surface of paper with WM by using methods that would make it possible to explore topography at the nanolevels. A promising method for such a research is AFM.

3. The aim and objectives of the study

The aim of this study is to identify special features in the structure of paper with WM, using methods with high resolution, such as AFM, at different areas, on the front and mesh side, which would make it possible to more accurately describe the topographical structure of the surface of paper with WM, and to use it for predicting print quality.

To achieve the set aim, the following tasks have been solved:

– to define the structural parameters of the surface of paper with watermarks using the methods of contact profilometry and atomic-force microscopy;

– to perform the correlation of results obtained by applying different methods.

4. Materials and methods to study the structural parameters of the surface of paper with watermarks

4. 1. The examined materials and the equipment used

To conduct an experimental study, we used tamper-proof paper without a coating and optical bleaching with positive and negative WMs (Fig. 1). Separate specifications that we determined during earlier research [14] are given in Table 1.

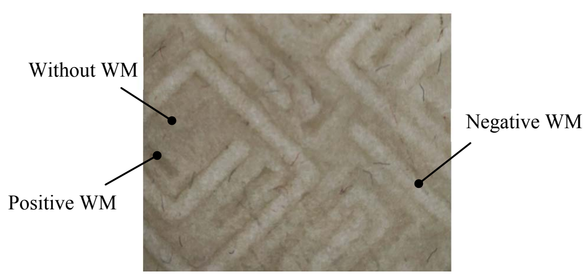


Fig. 1. Photograph of a sample of tamper-proof paper to the light

Table 1

Technical characteristics of non-printed paper [14]

No. of entry	Parameters		Values
1	Mass, g/m^2		90
2	Color coordinates (L, a, b*)		92.58; –0.31; 5.31
3	Optical density to the light	Area without WM	0.57
		Area with negative WM	0.50
		Area with positive WM	0.64
4	Thickness, μm	Area without WM	0.079
		Area with negative WM	0.074
		Area with positive WM	0.088

The structural parameters of the surface of paper with WM were determined by using the methods of contact profilometry and AFM.

Measurement of the roughness of samples was carried out using the profilometer MarSurf PS1. We applied a probe with a needle radius of 2 µm, measuring effort is 0.7 mN, trace length was 4 mm. Morphology of the samples' surfaces was studied by AFM method at the scanning probe microscope FemtoScan Online using the software FemtoScan Online.

4. 2. Procedure for determining the indicators for samples' properties

To estimate the profile of the surface irregularities, we applied a profilometry method based on contact analysis of the plot at the paper's surface with a thin needle, which makes it possible to obtain the dimensional parameters for irregularities and the magnified image of the surface's profile [22]. The measurement of roughness was carried out at the wire and felt sides of a sheet at areas with a water mark and beyond.

To study the morphology of the sample' surface using AFM method, we performed scanning under a contact mode in the air applying the cantilevers fpN10S. According to specifications, typical radii of cantilevers' curvature are less than 10 nm. The scanning was conducted at the following regions of the paper: region with a positive and a negative watermark, region without a watermark. The analysis was conducted at the felt and wire sides of the paper sheet.

The study was carried out at a temperature within 20–24 °C and a relative air humidity of around 50–56 %.

5. Results of studying the structure of paper with watermarks

The results from measuring the parameters for roughness of the paper's surface, obtained by the method of contact profilometry, are given in Table 2. We determined R_a – the arithmetic average of the profile's deviation (the arithmetic mean of absolute values for the profile's deviations within the range of a base length); R_z – the height of the profile's irregularities at 10 points (the sum of average absolute values for heights of five largest protrusions and depths of five largest profile's hollows within the range of a base length); R_{max} – the greatest height of the profile (a distance between the line of profile's protrusions and the line of profile's hollows within the range of a base length).

Fig. 2, 3 show profilograms that visualize the results of measurements by a profilometer at different regions of the paper with positive WM, with negative WM and beyond. The processing of profilometer signals has made it possible to derive the averaged parameter for roughness of the surface at each examined regions of the paper. Averaged indicator quantitatively characterizes the surface irregularities calculated for the trace length of 4 mm. The profilograms below were built using the procedure reported in [14].

Based on the acquired data, we built dependence charts of roughness parameters R_a and R_{max} on the presence of WM at different regions of paper from the felt and wire sides (Fig. 4). By analyzing values for R_a, we can conclude that for the wire side this indicator is larger (for regions with negative WM – 3.186 µm, for regions with positive WM – 3.008 µm) than that for the felt side (for regions with negative WM – 3.166 µm, for regions with positive WM – 2.949 µm). At regions of negative WM, the profile deviation's arithmetic average is the largest, while the lowest – at positive WM, due to the greater compaction of fibers at these regions and a greater pressure of cantilevers.

Table 2

Parameters for surface roughness of paper samples, obtained using the method of contact profilometry, µm

Parameters for roughness	Wire side			Felt side		
	Neg. WM	No WM	Pos. WM	Neg. WM	No WM	Pos. WM
R_z	17.450	17.380	16.760	17.890	16.820	17.010
R_a	3.186	3.163	3.008	3.166	3.0160	2.949
R_{max}	20.380	21.440	19.880	22.080	20.010	21.220
Absolute error 95 %	0.119	0.143	0.111	0.125	0.197	0.155
Relative error, %	3.741	4.512	3.674	3.952	6.528	5.243

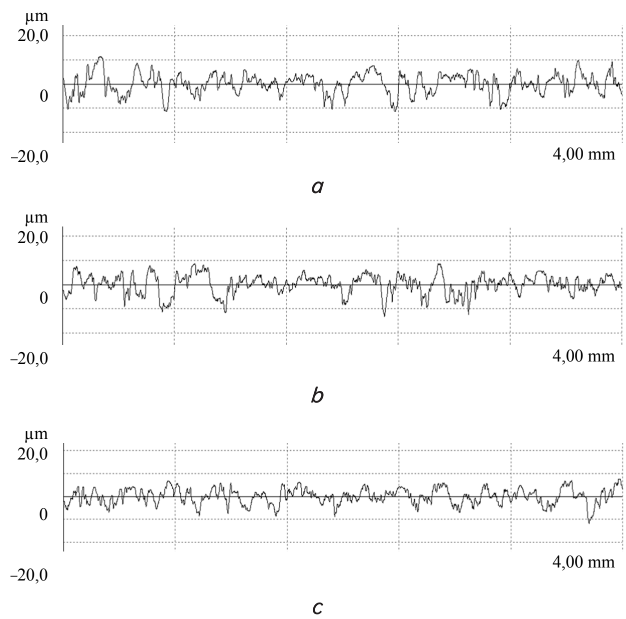


Fig. 2. Profilograms of the wire side's surface of paper samples: *a* – region with a negative watermark; *b* – region without a watermark; *c* – region with a positive watermark

Value for the profile's largest height does not depend on measurement region that can be associated with the chaotic arrangement of fibers and particles from the filler in the paper bulk that protrudes above the profile line. When comparing regions with negative and positive WM, one observes that value R_{max} is larger at negative ones (a wire side is 20.380 µm, a felt side is 20.080 µm).

Mathematical expectation regarding R_a and R_{max} is shown in Fig. 4, 5.

We have established, based on the results from AFM study, values for the arithmetic average irregularity R_a, the root-mean-square irregularity R_q, asymmetry parameters R_{sk}, and kurtosis measure R_{ku} (Table 3), we derived the topography of the sample's surface and the profiles of cross regions (Table 4).

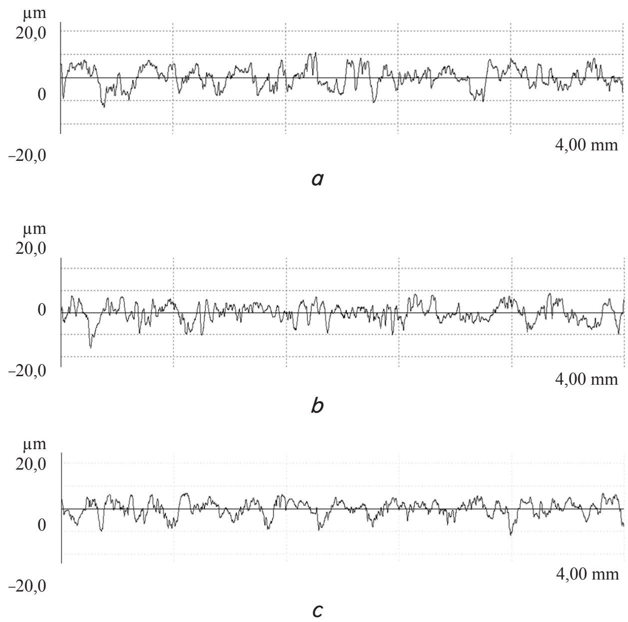


Fig. 3. Profilograms of the felt side's surface of paper samples: *a* — region with a negative watermark; *b* — region without a watermark; *c* — region with a positive watermark

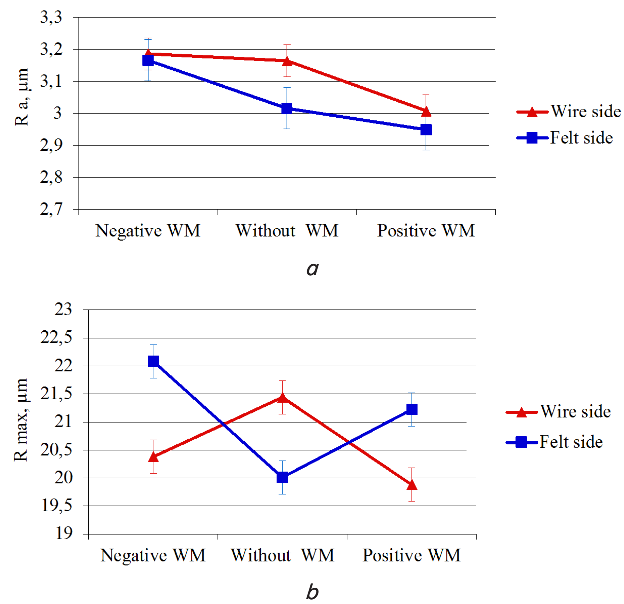


Fig. 4. Roughness parameters for different regions of paper at the felt and wire sides (method of contact profilometry): *a* — value for the arithmetic mean deviation of the profile (R_a); *b* — value for the profile's greatest height (R_{max})

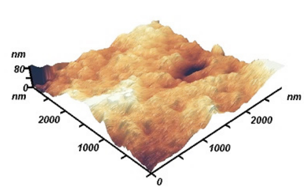
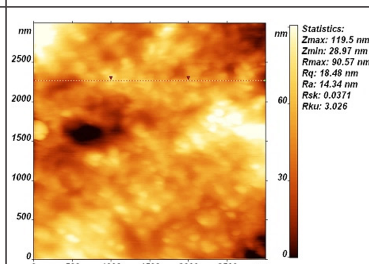
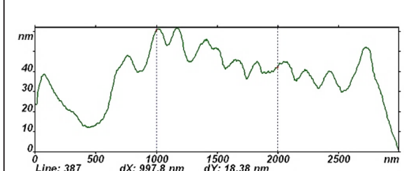
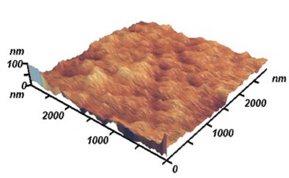
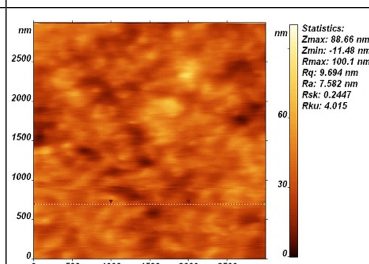
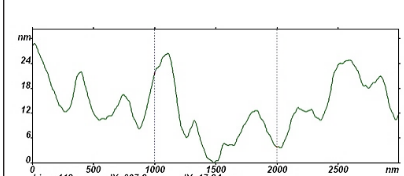
Table 3

Roughness parameters for the surface of paper samples, obtained using AFM method, nm

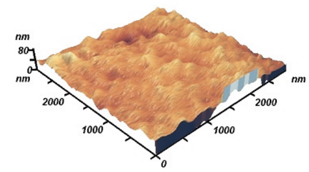
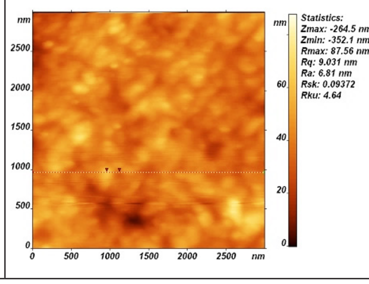
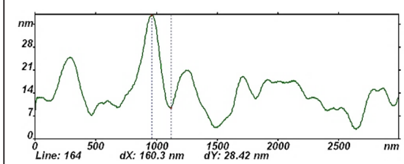
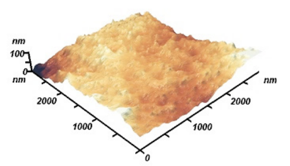
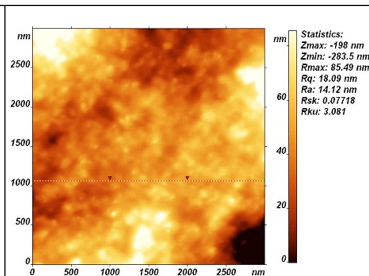
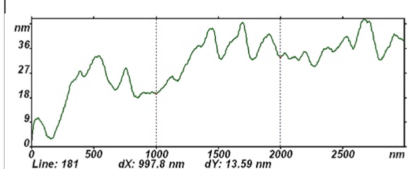
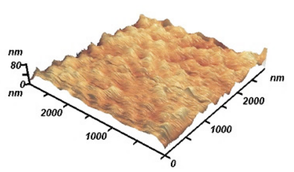
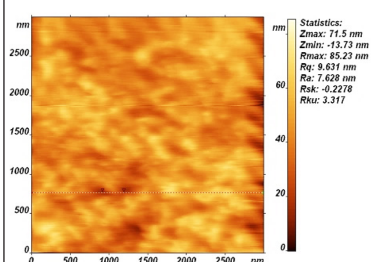
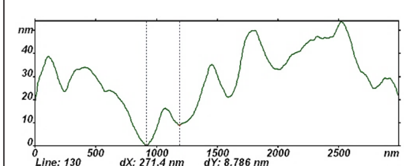
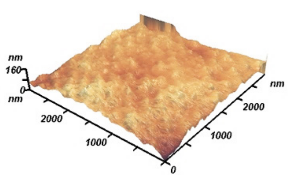
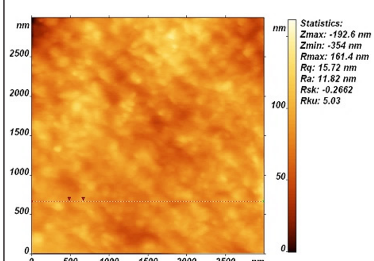
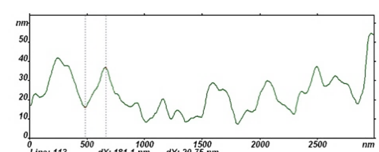
Roughness parameters	Wire side			Felt side		
	Negative WM	No WM	Positive WM	Negative WM	No WM	Positive WM
R_{max}	108.501	85.231	161.403	90.572	100.104	87.561
R_q	18.261	9.630	15.721	18.484	9.692	9.034
R_a	14.383	7.633	11.824	14.340	7.583	6.810
R_{sk}	–0.194	–0.231	–0.270	0.041	0.240	0.093
R_{ku}	3.462	3.320	5.031	3.034	4.021	4.644

Table 4

Surface topography of the examined sample of paper at different regions

3D		2D	Profile
Felt side			
1	2	3	4
region with negative WM			
region without WM			

Continuation of Table 4

<table>
<tr><td>1</td><td>2</td><td>3</td><td>4</td></tr>
<tr><td>Region with positive WM</td><td></td><td></td><td></td></tr>
<tr><td colspan="4">Wire side</td></tr>
<tr><td>region with negative WM</td><td></td><td></td><td></td></tr>
<tr><td>region without WM</td><td></td><td></td><td></td></tr>
<tr><td>region with positive WM</td><td></td><td></td><td></td></tr>
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Based on the results from AFM study (Table 3), we constructed diagrams that visualize the relationship between the parameters of roughness R_a and $R_{\max}$ and the examined regions of paper at the felt and wire sides with positive WM, negative WM, and beyond (Fig. 5).

The area of scanning for all investigated samples varied from 9×9 μm^2 to 500×500 nm^2. Surface roughness parameters changed at a change in the size of the scanning frame. Thus, for example, for the region without WM, the roughness, depending on the area of the analysis, accepts the value R_a=7.58 nm for area (3×3 μm), and the value R_a=12.6 nm for area (9×9 μm). Such a dependence of roughness on the examined paper's surface area is a predictable pattern of the relief that has different scale elements in its structure. Gradually, the surface roughness decreases in proportion to a decrease in the examined area, until its value becomes a constant magnitude. In a given case, a constant value for the surface roughness was registered for all samples at a scanning area of 3×3 μm^2; a further decrease in the scanning area did not lead to a change in the parameters for roughness.

Upon obtaining the results from a study that involved both methods, they were statistically processed. We determined the absolute error and a relative error in line with a procedure given in [23, 24]: for contact profilometry, the values obtained are given in Table 2, for AFM, the results were processed using the software FemtoScan online [25].

By analyzing the value R_a, we see that for regions with negative WM it is larger (wire side – 14.383 nm, felt side – 14.340 nm) than that for positive WM (wire side – 11.824 nm, felt side – 6.810 nm). No dependence of values $R_{\max}$ on the measurement region was established. This could relate to that the size of the examined region (3,000×3,000 nm) covers only part of the pulp fiber.

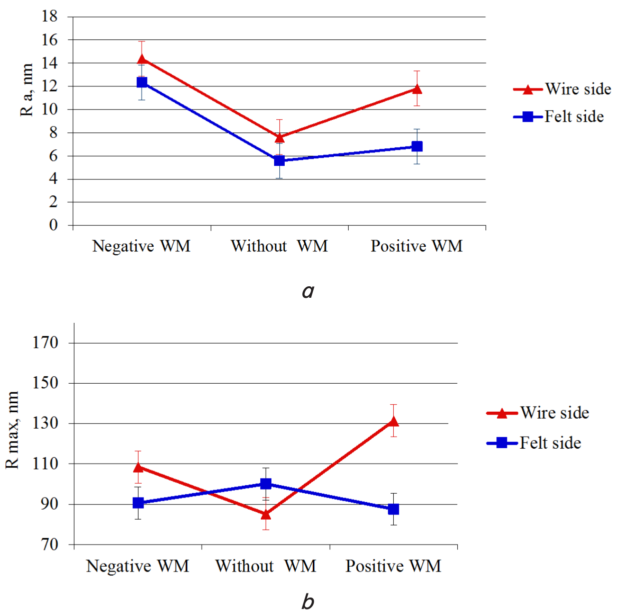

Fig. 5. Roughness parameters for different regions of paper at the felt and wire sides (AFM): *a* — value of the arithmetic mean deviation of the profile (R_a); *b* — value of the profile's greatest height (R_{max})

6. Discussion of results of studying the structure of the surface of paper with water marks

Following an analysis of the structure of tamper-proof paper one can argue that both study methods are appropriate for exploring the structural parameters of paper surface. The data obtained by different methods are combined via positioning and measurements at the defined region. The results from measurements by a contact profilometer have made it possible to trace a correlation between values of arithmetic mean deviation of the profile and the presence of negative and positive watermarks (Table 2 and Fig. 2, 3). This occurs both when using a contact profilometer and when applying AFM. A value for the profile's largest height R_{max} does not depend on a measurement region, which may relate to the chaotic arrangement of fibers and particles form the filler in paper bulk that protrude above the line of the profile. The identified patterns refine at the nano level the data reported in studies [13–15, 17, 18], aimed at a precise reproduction of the structural parameters of paper in the nanometer range by AFM method, the visualization of its structure [11], determining the roughness indicators [14, 15].

It was established by analyzing the value of the arithmetic mean deviation of the profile, obtained by AFM method (Table 3, 4), that for regions with negative water marks it is larger than that for positive. No dependence of the profile's largest height on a measurement region was established. This may relate to that the size of the examined region (3,000×3,000 nm) covers only part of the fiber in the pulp.

Based on the results obtained, we proposed a model of the paper's surface profile, which is a superposition to the profile derived using a contact profilometer, or the profile obtained using AFM (Fig. 6).

The combination of these methods will make it possible to analyze the surface of paper at the level of micro- and nanometers. A profilometry method yields realization of the profile of the surface of paper formed in a certain way, while AFM – the morphology of its components (fibers, fillers, etc.).

At present, using the results from measurements makes it possible to draw a conclusion about quality of the paper canvas's surface and its suitability for printing using various printing techniques, taking into consideration the presence of protective elements – WM. In addition, the data obtained could be used for entry control of materials at printing enterprises and to control quality of products in the pulp and paper industry.

The results reported here refine and complement earlier studies [9, 13, 14] by employing different scales of assessment. In further studies, it would be relevant to explore, by using the developed methodology, quality of offset printing, in particular, the reproduction of fine guilloche lines on paper with different structural characteristics that would make it possible to correlate the quality of reproduction and the parameters for the structure of paper's surface.

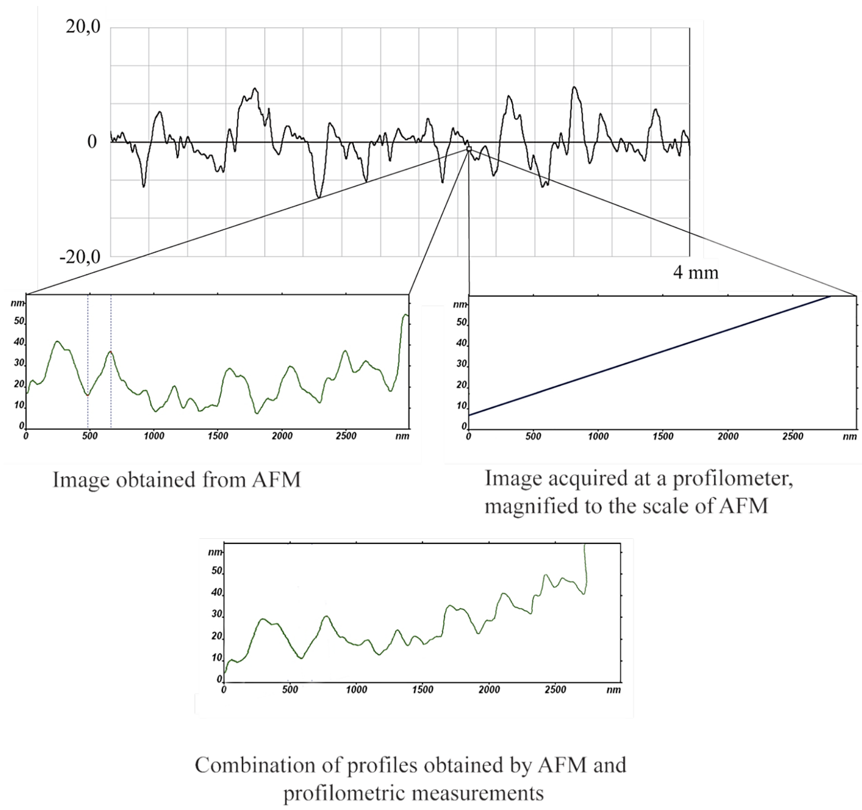

Fig. 6. Topography of sample's surface at the level of micro- and nanometers (wire side, region of positive watermark)

Since the methods applied in this study do not make it possible to obtain intermediate data between nano- and micrometers, then in the further studies, for a clear visualization of its structure, we plan to involve intermediate ranges of measurements, in particular, optical profilometry. In addition, the experimental data obtained in this study on the topography of the surface of non-printed tamper-proof paper could be supplemented in the future with data on the samples that are printed by offset technique, which would be appropriate to use for determining the fractal parameters of paper with WM.

7. Conclusions

1. The structural parameters of the surface of tamper-proof paper are different for regions with watermarks and without them. By analyzing the roughness parameters, obtained using a method of contact profilometry and AFM, one can see a clear correlation between results. At regions with negative WM the roughness parameters are the largest (wire side – 3.186 μm, 14.383 nm; felt side – 3.166 μm, 14.340 nm) and at regions with positive WM, they are the lowest (wire side – 3.008 μm; 11.824 nm, felt side – 2.949 μm, 6.810 nm), due to the greater compaction of fibers at these regions and a greater pressure of cantilevers.

2. Methods of contact profilometry and atomic-force microscopy are suitable for the assessment of specific characteristics of the surface of paper, thereby determining the character of interaction between different areas of tamper-proof paper and printing inks. A profilometry method yields realization about the profile of the surface of paper formed in a certain way, while an atomic-force microscopy method – about the morphology of its components (fiber, fillers, etc.). Information about the structural properties of paper makes it possible to predict the quality of imprints, and therefore is relevant because it prevents flaws in printed products for special purposes.

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ANALYSIS OF CHANGE IN THE DECORATIVE PROPERTIES OF GRANITES UNDER THERMAL EXPOSURE

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Експериментально досліджувалися зразки з дев'яти родовищ гранітів, які видобувається в Україні. Випробування зразків гранітів проводилося високими температурами 200, 400, 600, 900 °С.

Всі представлені граніти показують зміну кольору поверхні при температурі від 200 °С і вище. Поведінка гранітів з нагріванням залежить від їх мінерального складу, структури та текстури.

Поверхні всіх дослідних зразків стали світліше, деякі зразки гранітів втратили насиченість кольорів. При нагріванні зразків до 900 °С природного каменю найбільше зростання компоненти L (зображення зразків каменю світлішає) кольорової системи CIELab відбулося у зразках гранітів Cardinal Grey та Carpazi відповідно на 42 та 44 %. Найменше зростання компоненти L при нагріванні зразків до 900 °С відбувається у гранітах Grey Ukraine, L відповідно на 4 та 8,5 %.

Вплив температури на зразки з червоного граніту візуально менш виражений, оскільки як свіжі, так і нагріті, зразки мають подібний червоний колір. Завдяки вмісту апатиту та флюориту зразки граніту Flower of Ukraine набувають рівномірного фіолетово-рожевого кольору при температурі 900 °С. В сірих гранітах при нагріванні з'являються почервоніння, які переважно сконцентровані навколо слюди та інших мінералів, які багаті Fe. На зеленому граніті Verde Oliva з'являються руді плями при температурі 200 °С. При нагріванні до 900 °С руді плями займають 67 % площі зразка.

Найбільшу зміну кольору отримали граніти, в яких відбувся фазовий перехід темноколірних мінералів (біотиту та піроксену) в поліморфні мінерали. Це надало зразкам гранітів світлішого кольору, так як мінерали змінили колір з чорного на сірий чи білий. Відтінки білого кольору надав кварц, в якому при нагріванні з'являлися білі мікротрішини.

Помітні естетичні пошкодження поверхні зразків природного каменю починаються при температурах від 200 до 400 °С. Таким чином, вогонь з температурами нижче цього порогу можна вважати «безпечним» з точки зору естетичного пошкодження, якщо врахувати тільки коефіцієнт нагріву вогню і виключити золу і гази

Ключові слова: граніт, високі температури, декоративність природного каменю, мінеральний склад, структура граніту

1. Introduction

Physical appearance of a natural facing stone is important for its use as a facing material. People built a considerable number of architectural monuments using facing rocks. Taking into consideration their considerable age, questions arise regarding their restoration [1]. There are certain possible problems: deposits ceased to develop, or deposits develop other horizons. A color of natural stone may have different shades at different terraces in this case.