This study investigates the process of cardboard surface modification with an aqueous solution of Al2(OH)nCl6–n and its effect on the indicators of contact angle and surface charge of dissolved substances in cardboard for the manufacture of packaging and disposable tableware.

The task addressed relates to the lack of a technological approach to modifying the surface of cardboard, which would simultaneously enable high adhesion between cardboard and polymer by controlled change in wettability and surface energy without loss of its barrier properties. A solution option is to improve the technology of modifying cardboard for the production of disposable tableware with an aqueous solution of $Al_2(OH)_nCl_{6-n}$.

The experimental studies involved manufacturing a series of cardboard samples with different compositions, determining the contact angle by the lying drop method and the surface charge of dissolved substances. The effect of surface treatment on the indicators of the contact angle and surface charge of dissolved substances of cardboard for the manufacture of packaging and disposable tableware has been established. It was found that the contact angle decreases to 82.8–84.7° compared to 91.8–93.4° for unmodified samples (without applied polymer) and 116.1–116.6° versus 112.1–115.9°, respectively, for samples with a polymer coating. At the same time, the surface charge of dissolved substances increases from –18.3–25.9 to –51.3–54.1 μg-equiv./l, that is, by 2–3 times.

The results indicate that the treated cardboard samples have better wettability. This property will contribute to better contact between the cardboard surface and the polymer melt during its application and subsequent gluing of the structural elements of an article.

The proposed approach to modifying the cardboard sheet is promising for further implementation in industrial processes, in particular to produce packaging materials and disposable tableware. This will make it possible to influence the barrier properties of the finished cardboard paper, as well as to improve the functional properties of packaging and disposable tableware made of modified cardboard

Keywords: laminated cardboard, raw material modification, contact angle, cationic charge

UDC 621/798-181-2

DOI: 10.15587/1729-4061.2026.357229

EFFECT OF MODIFYING THE SURFACE OF CARDBOARD WITH ALUMINUM HYDROXYL CHLORIDE SOLUTION ON THE INDICATORS OF THE CONTACT ANGLE AND SURFACE ENERGY

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Received 23.01.2026
Received in revised form 31.03.2026
Accepted date 09.04.2026
Published date 30.04.2026

How to Cite: *Roik, T., Bychkar, Y., Oliynyk, V. (2026). Effect of modifying the surface of cardboard with aluminum hydroxyl chloride solution on the indicators of the contact angle and surface energy. Eastern-European Journal of Enterprise Technologies, 2 (6 (140)), 51–61. https://doi.org/10.15587/1729-4061.2026.357229*

1. Introduction

Paper and cardboard are inexpensive materials that are widely used in the production of packaging products and disposable tableware, due to their sufficiently high mechanical strength and flexibility.

Manufacturers of disposable tableware and cardboard packaging products face numerous limitations, in particular, insufficient mechanical and barrier properties, such as the water permeability of cardboard, which is due to its hydrophilic nature and porous structure.

However, although hydrophobicity, which is achieved by gluing [1–3] paper pulp and which increases the material's resistance to moisture, may be insufficient to enable the required level of water resistance of cardboard materials and articles. To achieve the required level of properties of raw materials for the production of disposable tableware, attention is paid to the use of composite materials, such as cardboard/polyethylene, cardboard/foil/polyethylene [2, 3].

Important indicators of the quality of raw materials for disposable tableware and cardboard packaging are moisture resistance. In papers [2, 3] it is indicated that moisture-proof cardboard has a fairly wide range of applications: packaging of dry food products (bread, flour, confectionery, tea, yeast), various light industry products, pharmaceuticals, medical supplies, flowers, etc. [4].

It has been shown that for the storage of many food products, the moisture resistance of the packaging material is a determining protective property, which consists in increased resistance to moisture penetration [5, 6].

Achieving moisture resistance of the starting raw materials for the manufacture of disposable tableware and composite packaging materials from cardboard should be enabled by appropriate pre-treatment.

Since manufacturers often use cardboard with polyethylene melt applied to it using extrusion technology [3–5] for the production of disposable tableware, the question arises of achieving a reliable degree of adhesion between cardboard and sizing polymers in finished articles.

Increasing the degree of adhesion between fibrous materials, such as cardboard, and polyethylene melt can be achieved, as is known [6, 7], by improving the wettability of the cardboard surface and increasing its surface energy. However, when cardboard manufacturers try to improve the mechanical and barrier properties of cardboard [8, 9], its hydrophilic properties decrease: wettability, absorbency, etc. The consequence of this is unsatisfactory adhesion of the cardboard surface with polyethylene and the lack of continuity of the polyethylene coating on the products. However, existing methods of increasing adhesion, namely increasing the stickiness, which indicates increased hydrophilicity and insufficient barrier protection [10], are not capable of providing high barrier properties of cardboard, which leads to unsatisfactory quality of finished articles [11, 12]. Because of this, the task of modifying cardboard and paper raw materials arises in such a way as to achieve the best degree of adhesion between the polyethylene melt and the cardboard and paper base.

Therefore, in-depth research on the surface treatment of cardboard sheet for the manufacture of disposable cardboard articles is a relevant task.

2. Literature review and problem statement

As is known, chemical surface treatment of raw materials is one of the ways to improve the mechanical and protective characteristics of packaging paper or cardboard. In study [1], it was found that the application of nanocrystalline cellulose in combination with polyvinyl alcohol on packaging cardboard increases the tear resistance by 15–20%. At the same time, the decrease in vapor permeability indicates the limited use of such materials for packaging, where gas exchange control is important. In work [2], the hydrophobic properties of cardboard coated with polylactic acid and ethylene absorbers are considered. Experiments have shown that the wetting angle increases to 95–105°, which provides high water resistance but the adhesion between the layers at high temperatures remains unstable. In [4], the design of superhydrophobic paper (CA > 150°) is described by introducing halloysite nanotubes into the material structure; however, the technology requires the use of aggressive solvents, which limits the industrial implementation of such technologies.

Thus, work [2] demonstrates that the improvement of some properties of cardboard such as increased water resistance is often accompanied by a deterioration of others – instability of adhesion at high temperatures, which indicates the absence of a universal technological solution and the need for in-depth study of the problem.

It should be noted that raw material modifiers are usually introduced into the paper pulp at the stage of its manufacture to provide moisture resistance. This is the approach proposed when applied as a coating directly to the surface of the finished sheet at the technological stage of the sizing press [3, 6].

The task to control surface wettability is of considerable interest in the scientific community due to its importance for both theoretical research and practical applications [5]. These substances are added to improve the stiffness and wet strength. The use of NaOH/urea has also shown a positive effect on the tensile strength and air permeability of paper [5]. However, obtaining materials with better barrier properties has not been achieved: with increasing tensile strength, the tensile strength decreases, and air permeability increases: treatment of corrugated cardboard with NaOH–urea increases the tensile strength by 8–12% and reduces air permeability by 15–20% [5, 6].

Studies have shown that polymer coatings of polylactide provide a 25–30% reduction in water absorption but partially reduce the stiffness of the cardboard, which demonstrates a compromise between improving barrier and mechanical properties [5]. However, papers [5, 6] that investigated the effect of alkaline treatment (NaOH/urea) on cardboard from recycled materials showed that with an improvement in tensile strength by 25%, the Hurley air permeability index increases, which indicates a loosening of the fiber structure and a deterioration in barrier properties, which requires further research and resolution of these limitations.

In [7], the association of cellulose microfibrils with silicates was studied. It was shown that the rigidity of the cardboard increases by 12%, but the surface roughness becomes uneven, which complicates uniform wetting by the polymer melt. In [9], the results of studies on the effect of modifying the cardboard surface with calcium silicate (Ca_2O_4Si) and magnesium silicate (MgO_3Si) and evaluating their characteristics as a coating on cardboard are reported; their characteristics are evaluated. The authors of [9, 11] studied the effect of the coating, which was prepared in different concentrations, on packaging materials, which were evaluated by the indicators of thickness, stiffness, water absorption, wetting angle, and surface tension. It was shown that chemical treatment (modification) of the starting raw materials for the production of packaging materials significantly changes the properties of cardboard or paper. Study [9] establishes a direct correlation ($R^2 = 0.94$) between the surface free energy and adhesion between polyethylene and cardboard. The authors argue that for reliable adhesion, the polar component of the surface energy should be maximized. In turn, the high correlation between surface energy and adhesion ($R^2 = 0.94$) established in [9] indicates the important role of surface energy characteristics in shaping the quality of composite materials.

The authors [10] demonstrate methods that make it possible to obtain, on one substrate, zones with a wetting angle of $\theta \approx 10°$ (superhydrophilicity) and $\theta > 150°$ (superhydrophobicity).

Chemical treatment of cardboard has both positive and negative consequences, such as a decrease in strength and stiffness by 12–15%; water absorption is reduced by 18% compared to uncoated cardboard or paper on the one hand and an increase in plasticity on the other hand [11]. In an ideal event, the chemically treated (modified) surface layer of cardboard should form a rougher surface, while the deeper (middle) layers should remain unchanged [11]. However, issues related to the scaling of this method of surface modification, which are limited by the conditions of research, remain unresolved.

In the packaging sector of the printing industry, various techniques of coating are being tested and investigated to improve the surface appearance and performance characteristics of paper and cardboard (strength, tensile strength, shrinkage during drying, warping, surface energy) [1, 11].

One of the key indicators in the analysis of the impact of chemical treatment of cardboard is the value of the contact angle [12, 13]. It clearly demonstrates the degree of wettability of the surface of the cardboard raw material and depends on the contact time of the liquid with the cardboard surface and its physical and mechanical properties (roughness, absorbency, porosity, etc.). Thus, for cardboard coatings with nanocoating, the contact angle value ranges from 55° to 78° for water, depending on the concentration of the modifier and the application technique [13]. This indicates the possibility of controlling wettability, but such approaches are mostly implemented for printing technologies and are not adapted to the production of materials for disposable tableware and packaging.

An option to overcome the problem is to study and apply controlled surface wettability for printing technologies, with an emphasis on designing and manufacturing precise surface wettability by enhancing the contrast between hydrophilicity and hydrophobicity, for example, superhydrophilicity and superhydrophobicity, which are reported in [14, 15].

This is the approach used in [15, 16], which mainly cover surface wettability in lithographic printing with various methods of making molds, wettability for printing with a patterned stamp, as well as special microtransfer printing with indirect wetting.

In addition, there is information on the wettability of suitable materials for inkjet printing with control over the wettability of the substrate surface and the print head with ink, as well as patterned wettability, which is achieved by combining different printing methods [14, 17].

Studies [17, 18] confirmed the existence of a correlation between the energy characteristics of the material surface, which depend on its structure, and the magnitude of the wetting angle. The authors of those papers established that the wettability directly depends on the atomic structure of the plane: on polar faces, the wetting angle with water is $\theta \approx 40°–45°$, and on non-polar or stable faces, the angle increases to $\theta \approx 65°–80°$. This confirms that changing the energy characteristics of the surface is an effective tool for controlling its properties.

In particular, in studies that report analyses of the wettability of single crystals, it was established that the higher the density of broken bonds per unit area of the solid surface, the higher its reactivity and, accordingly, the better the wettability [14, 15]. The researchers considered the wettability of both inorganic and organic (such as thymol, salol, benzophenone, etc.) [16, 18].

Studies on the influence of preliminary modification of cardboard raw materials for the manufacture of packaging and disposable tableware on a change in the indicators of the contact angle of wetting and surface energy are extremely limited [18, 19]. Namely, these indicators directly affect the degree of wetting of the cardboard surface during the subsequent application of the polymer melt.

Among the known modifiers that are usually introduced into the composition of the starting raw materials at the stage of manufacturing cardboard and paper pulp are acrylic dispersion, acrylic emulsions, polyvinyl alcohol or lignosulfonate solutions, moisture-resistant adhesives, etc. [20, 21].

However, studying the relationship between the indicators of the wetting angle, surface energy, and the quality of raw materials for the manufacture of disposable tableware and composite packaging materials has not been widely implemented. This does not make it possible to obtain consistently high quality characteristics of the specified cardboard articles.

Studies [21, 22] on the use of modification of cardboard and paper raw materials for the manufacture of packaging and disposable tableware show the prospects of using surface treatment of paper and cardboard. Papers [23, 24] have shown that surface modification of cardboard sheet could have an effect on the indicators of the contact angle, which would make it possible to influence its adhesive properties. Among the modifiers, the authors of [25, 26] considered aqueous solutions based on aluminum sulfates and oxysulfates, in particular, based on $Al_2(SO_4)_3 \times 18H_2O$ coagulants and hydroxychlorides – $Al_2(OH)_nCl_{6-n}$-coagulants.

The properties of hybrid flocculants and coagulants based on aluminum hydroxychloride are described in detail in [27, 28]. It has been proven that these substances are capable of effectively changing the surface potential of particles, which is key to their enlargement and removal from the aqueous phase.

These substances are usually used for the treatment of drinking water, wastewater treatment, as well as in technological processes of various industries, including the pulp and paper industry.

Such modifiers-coagulants contribute to the increase in the surface energy of harmful substances dissolved in water and thereby enable their aggregation (coagulation) and subsequent removal from water [27, 28]. This creates the prerequisites for their use as cardboard surface modifiers.

Determining the advantages and disadvantages of preliminary modification of raw materials for packaging and disposable tableware opens up opportunities for improving the absorption capacity, roughness, surface energy index, and contact angle. This, in turn, at the next technological stage of applying a polyethylene coating could contribute to better penetration of the polyethylene melt into the depth of the cardboard and, accordingly, to achieving a greater degree of adhesion.

Therefore, given the modern requirements for disposable cardboard articles and packaging, there is a need to devise technological approaches that will allow for a controlled influence on the wettability of the cardboard surface and its energy characteristics without deteriorating mechanical and barrier properties.

The practical significance of such research implies the possibility of ensuring better interaction between cardboard and polymer coating, which will contribute to increasing adhesion between the cardboard sheet and the polymer and improving the quality of finished articles.

All this gives grounds to argue that it is advisable to conduct a study to determine the influence of technological factors of cardboard surface modification using an aqueous solution of $Al_2(OH)nCl_6$-n on the indicators of the contact angle of the surface and surface energy. These indicators are decisive for ensuring such a level of wettability of the cardboard surface, which, when further coated with polyethylene melt, will contribute to achieving a high degree of adhesion of the polyethylene film to the cardboard. The specified technological approach is capable of enabling the production of cardboard packaging and disposable tableware of high quality.

The above motivated our implementation of research aimed at improving the technology of modifying raw materials for the production of disposable tableware. This will significantly improve the mechanical barrier properties, as well as adhesion between cardboard and polyethylene, which will contribute to the high quality of finished articles. The coagulant aluminum hydroxychloride was chosen to modify the cardboard surface.

3. The aim and objectives of the study

The purpose of our study is to determine the effect of surface treatment of cardboard with an aqueous solution of aluminum hydroxide chloride on the indicators of the contact angle and surface energy, and to identify the relationship between the use of the specified modification of the cardboard surface. This will make it possible to influence the quality parameters, namely the barrier properties of the finished cardboard article, and will significantly improve the functional properties of packaging and disposable tableware made of modified cardboard.

To achieve this goal, the following tasks were set:

– to measure the contact angle, compare the results with untreated cardboard samples, and establish the influence of the obtained surface parameters on the barrier properties of the studied samples;

– to conduct a study on the surface charge indicators of dissolved substances, compare the results with untreated cardboard samples, and establish the influence of the obtained surface parameters (hydrophilicity, energy indicators) on the barrier properties of the studied samples.

4. The study materials and methods

The object of our study is the process of surface modification of cardboard with an aqueous solution of aluminum hydroxychloride and its effect on the indicators of the contact angle and surface charge of dissolved substances of cardboard for the manufacture of packaging and disposable cardboard tableware.

The hypothesis of the study assumes that surface treatment of cardboard sheet with coagulants containing aluminum hydroxychloride could make it possible to form such a sheet structure that would provide an increase in the cationic charge of the cardboard surface. In turn, an increase in the cationic charge and surface energy of the cardboard sheet would make it possible to improve the wettability of the surface and expect improved adhesion between the cardboard and the polymer melt during its subsequent application in the process of manufacturing an article.

It was hypothesized that changing the surface properties of the cardboard sheet by surface modification would affect the degree of adhesion between the cardboard and the polyethylene melt at the stage of its application. It was expected that surface modification with aluminum hydroxychloride could change such indicators as absorption capacity and moisture resistance and make it possible to improve the physical, mechanical, and barrier properties of the finished article.

Before the study began, it was assumed that cardboard samples manufactured using standard technology are homogeneous and comparable to each other, the differences in their properties are due to surface modification. It is believed that the conditions for manufacturing and processing the samples are reproducible, the methods used to determine the contact angle, surface energy, and surface charge of dissolved substances make it possible to characterize changes in the physicochemical properties of the samples under study.

In the process of preparing for the study, existing coagulants were analyzed, a comparative analysis was conducted, and on its basis, the aluminum hydroxychloride substance $Al_2(OH)nCl_{6-n}$ was selected, which has the greatest impact on the surface energy of the materials processed by it [28].

It is known [29] that the intermolecular interaction between a solid and a liquid can include several types of components, in particular polar, dispersive, hydrogen, etc.

The surface energy of solid materials is closely related to the contact angles of surface wetting by various liquids [29, 30].

In the process of research, an aqueous solution of aluminum hydroxychlorides with the general formula $Al_2(OH)nCl_{6-n}$ was selected from among existing coagulants, which correspond to the PAC (Polyaluminium Chloride) type coagulants in the European classification.

It was assumed that surface treatment with an aqueous solution of aluminum hydroxide chloride would increase the cationic charge of the cardboard surface at the stage of its manufacture. And this, in turn, would affect the wettability and barrier properties of the cardboard.

At the beginning of the research, several types of cardboard sheets of different compositions were manufactured using standard industrial technology [26].

At the stage of drying the cardboard sheet, surface treatment of part of the cardboard was carried out by wetting with an aqueous solution of aluminum hydroxide chloride, and the rest of the cardboard sheet was left without surface treatment.

For our research, 20 samples measuring 210 × 291 mm were manufactured from each type of cardboard sheet, the composition of which is given below.

Given the assumption that, according to the test results, the cardboard used will be applied for the production of disposable tableware, part of the manufactured samples was covered with a layer of polyethylene by extrusion.

Since brown cardboard is often used in the manufacture of disposable beverage containers, some of the samples for research were made with the addition of brown dye.

Thus, samples with the following compositions were used for our experiments:

No. 1 – Cardboard samples made of bleached cellulose on the front side and wastepaper on the back;

No. 2 – Cardboard samples made of bleached cellulose on the front side and wastepaper covered with a layer of polyethylene on the back;

No. 3 – Cardboard samples made of unbleached cellulose on the front side and wastepaper, with the addition of brown dye on the back;

No. 4 – Cardboard samples made of unbleached cellulose on the front side and wastepaper, with the addition of brown dye and covered with a layer of polyethylene on the back.

The front and back surfaces of samples No. 1 and No. 2 were not modified with an aqueous solution of aluminum hydroxide chloride.

Samples No. 3 and No. 4 were treated with an aqueous solution of aluminum hydroxychloride on both sides by immersion in a bath with a solution of aluminum hydroxychloride in the following dosage: 5.5 μl per 1 l of water for treatment of 11 g of the tested samples, immersion time – 0.065 s.

After the treatment, a layer of polyethylene was applied to samples No. 2 and No. 4.

To assess the achieved modification effect, the indicator of the contact angle and the surface charge of the dissolved substances of the obtained samples were investigated.

Wetting was carried out with distilled water as a chemically inert substance.

Measurements were carried out at a relative humidity of the environment of 50 (+/–2) % and a temperature of 23 (+/–1) °C.

The contact angle was measured using a goniometer (Fig. 1) using the lying drop method [3, 26]. The contact angle indicators were determined according to the scheme in Fig. 2 [27].

Fig. 1. General view of the contact angle measuring device DSA25, Krüss (Germany)

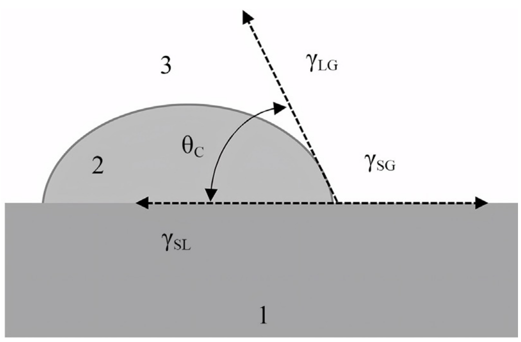

Fig. 2. Schematic of measuring the contact angle [27]: 1 — cardboard surface, 2 — liquid drop, 3 — gas; θ_C — contact angle, defined as the angle between the tangent to the drop surface at the point of three-phase contact and the surface of the solid; γ_{SL} — surface tension at the solid-liquid interface; γ_{SG} — surface tension at the solid-gas interface; γ_{LG} — surface tension at the liquid-gas interface

A drop of water was applied to the surface of cardboard samples placed horizontally in the device, after which the contact angle was measured after 0.5; 1; 2; 5; 10; and 60 seconds. Almost all colloids and solid particles carry electrical charges when dissolved in water [28, 29]. This leads to a concentration of oppositely charged ions, the so-called counterions, on the surface of the particles. If these counterions are separated from the particle or torn off from it, a flow potential arises, which can be measured in mV and the surface charge of the solutes of the materials under study can be estimated [28, 29].

Since, as noted above, most materials dispersed in water carry an electrical charge [28, 29], this makes it possible to predict and estimate the relationship between the surface charge of the solutes and the surface energy of the cardboard sheet. This is fundamental for understanding the behavior of colloidal systems and adsorption processes [30].

To measure the surface charge of wastepaper and cellulose fibers, samples were ground in a disintegrator (Fig. 3) for 15 min and mixed with distilled water to obtain a suspension with a solid phase concentration of 1% (Fig. 4).

Fig. 3. General view of the Labtech disintegrator (Canada)

Next, the obtained suspension was placed in the measuring cell of the device. The measurement of the surface charge of the dissolved substances of the studied samples was carried out using the Mütek™ SZP-10 System Zeta Potential device (Fig. 5) according to the scheme in Fig. 6.

The state-of-the-art equipment that was used in the research allows us to obtain correct and reliable results.

Measurement of the contact angle and surface charge of dissolved substances allows us to assess the effectiveness of the surface treatment of the studied cardboard samples with aluminum hydroxychloride.

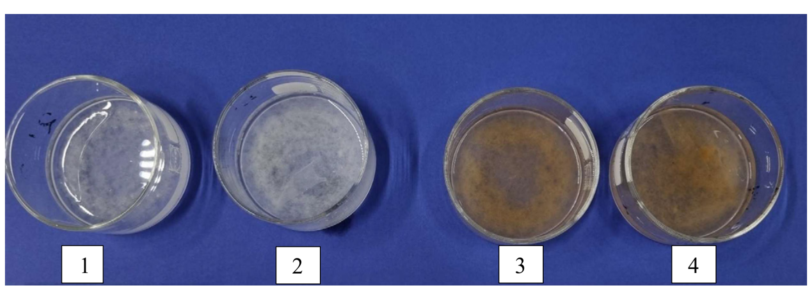

Fig. 4. General view of the samples crushed in the disintegrator: 1 — a sample of cardboard made of bleached cellulose on the front side and waste paper on the back; 2 — a sample of cardboard made of bleached cellulose on the front side and waste paper covered with a layer of polyethylene on the back; 3 — a sample of cardboard made of unbleached cellulose on the front side and waste paper, with the addition of brown dye on the back; 4 — a sample of cardboard made of unbleached cellulose on the front side and waste paper, with the addition of brown dye and covered with a layer of polyethylene on the back

Fig. 5. General view of MütekTM SZP-10 (Germany)

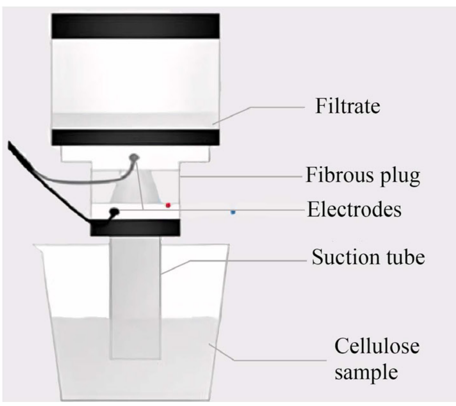

Fig. 6. Schematic representation of the Mtek TM SZP-10 measuring cell with a sample

5. Results of investigating the influence of modification of the cardboard surface with $Al_2(OH)_nCl_{6-n}$ solution on wettability and energy indicators

5. 1. Results of measuring the contact angle

The measurement data, contact angle indicators, were entered in Table 1.

The measurement results given in Table 1 show a comparison of the values of the contact angle of the surface of pre-treated with aluminum hydroxide chloride solution and untreated samples when wetting with water.

As can be seen from Table 1, samples No. 3 and No. 4, pre-treated with an aqueous solution of aluminum hydroxide chloride, are characterized by lower values of the contact angle compared to untreated samples No. 1 and No. 2.

In particular, for samples No. 3 this indicator is 116.1–116.6°, and for samples No. 4 – 82.8–84.7° (front side) throughout the entire observation period – 0.5, 1, 2, 5, 10, and 60 seconds. For the reverse side, the contact angle is 120.0–121.1° for samples No. 3 and 119.4–120.5° for samples No. 4.

For comparison, in untreated samples No. 1, the contact angle reaches 112.1–115.9° and sample No. 2 – 91.8–93.4°, respectively, on the front side and 122.3–122.6° for sample No. 1 and sample No. 2 – 114.9–115.6°, respectively, on the back side under similar measurement conditions.

A visual illustration of the compared values of the contact angle for the front and back sides of samples No. 1 and 3 is shown in Fig. 7, 8.

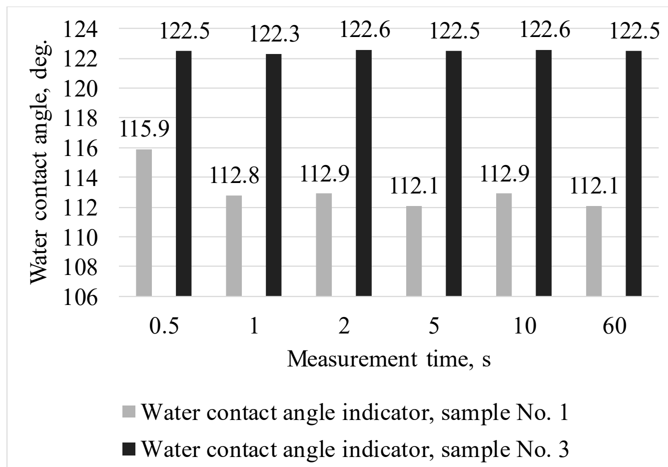

Fig. 7. Comparison of water contact angle indicators, front side (for samples without polyethylene coating)

As can be seen from Table 1 and Fig. 7, 8, samples No. 3, which were treated with an aqueous solution of aluminum hydroxide chloride, have lower contact angle values, which, as is known [3], indicates better surface wettability of the studied samples. The decrease in contact angle occurs both from the front and back sides of the studied samples.

Table 1

Results of measuring the contact angle, degrees

No.	Sample composition	Treatment with aluminum hydroxychloride	Measurement side	Contact angle, degree					
				Measurement time, s					
				0.5	1	2	5	10	60
1	Bleached cellulose	–	front	115.9	112.8	112.9	112.1	112.9	112.1
	Waste paper		back	122.5	122.3	122.6	122.5	122.6	122.5
2	Bleached cellulose	–	front	93.4	93.2	92.8	91.8	92.8	91.8
	PE coated wastepaper		back	114.9	115.2	115.6	115.3	115.6	115.3
3	Unbleached cellulose	+	front	116.1	116.9	116.8	116.6	116.8	116.6
	Waste paper with added brown dye		back	120	120.6	121.1	120.2	121.1	120.2
4	Unbleached cellulose	+	front	84.7	84.6	84.2	82.8	84.2	82.8
	Waste paper with added brown dye with PE coating		back	119.4	120	120.5	119.5	120.5	119.5

Notes: PE – polyethylene.

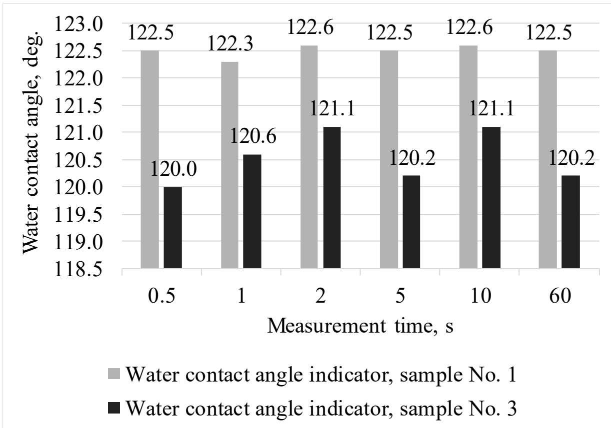

Fig. 8. Comparison of water contact angle indicators, reverse side (for samples without polyethylene coating)

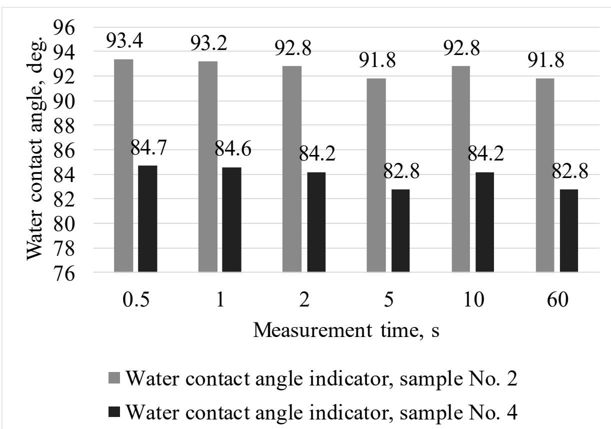

Fig. 9. Comparison of water contact angle indicators, front side (for samples coated with polyethylene)

A similar pattern was observed when studying the contact angle values for the front side of samples No. 2 and No. 4 with polyethylene coating (PE), which is shown in Fig. 9, 10.

As can be seen from Fig. 9, 10, the front side of samples No. 3, 4, which were modified with an aqueous solution of aluminum hydroxychloride, demonstrate lower contact angle values, compared to samples No. 1, 2, which were not modified.

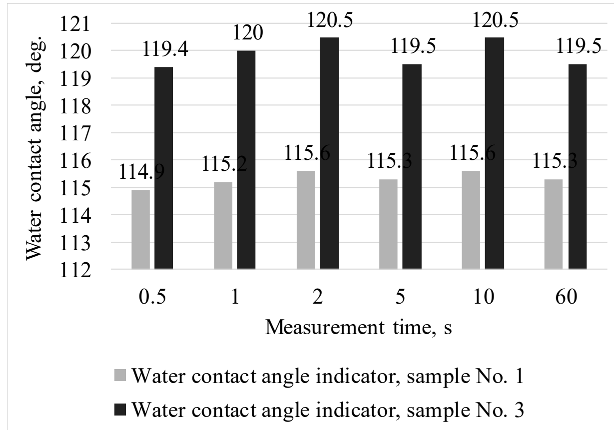

Fig. 10. Comparison of water contact angle indicators, reverse side (for samples coated with polyethylene)

5. 2. Results of measuring the surface charge of dissolved substances

The next stage in the research process was to determine the value of the surface charge of dissolved substances of samples that were pre-treated with a solution of aluminum hydroxychlorides $Al_2(OH)_nCl_{6-n}$, and samples without treatment.

The results of measurements of the surface charge of dissolved substances of the studied samples are given in Table 2.

Table 2 shows that the surface treatment of cardboard significantly affects the surface charge of solutes, namely, it is –51.3 µg-equiv./l for samples No. 3 (with surface treatment) and –25.9 µg-equiv./l for samples No. 1 (without treatment). A similar trend is observed for samples with a layer of polyethylene applied No. 2 and 4: –54.1 µg-equiv./l for samples No. 4 (with surface treatment) and –18.3 for samples No. 2 (without treatment).

Fig. 11 illustrates the relationship between cardboard modification and the surface charge of solutes of the studied samples.

As can be seen from Fig. 11, there is a clear relationship between the modification of the cardboard sheet, a decrease in the average contact angle, and an increase in the surface charge of dissolved substances.

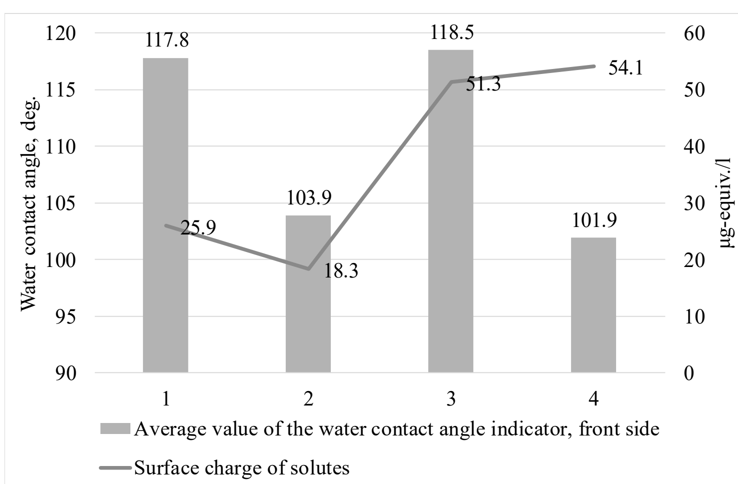

Fig. 11. Relationship between the contact angle indicators and the surface charge indicator of solutes: 1, 2, 3, 4 – numbers of the studied samples

Table 2

Results of measuring the surface charge of dissolved substances of the studied samples

No. of entry	Composition of the test sample	Mass 1 m^2, g	Treatment with $Al_2(OH)_nCl_{6-n}$	Surface charge of dissolved substances, μg-equiv./l	Mean value of the contact angle indicator
1	Waste paper, bleached cellulose	234	–	–25.9	117.8
2	Waste paper with PE coating, bleached cellulose	246	–	–18.3	103.9
3	Waste paper with added brown dye, unbleached cellulose	179	+	–51.3	118.5
4	Waste paper with added brown dye with PE coating, unbleached cellulose	195	+	–54.1	101.9

The relationship between the increase in the value of the surface charge of dissolved substances with a slight decrease in the contact angle is explained by the increase in the number of polar functional groups on the surface of the fibers, which increases their surface energy and enhances the interaction with water. Such indicators testify to an increase in the surface energy of the studied samples that were modified, which could make it possible to obtain better adhesion between the cardboard and the polyethylene melt.

6. Discussion of results based on investigating the influence of the modification of cardboard sheet with an aqueous solution of aluminum hydroxychloride $Al_2(OH)_nCl_{6-n}$

During our experimental studies, the influence of the surface treatment of cardboard with an aqueous solution of aluminum hydroxychloride on the key physicochemical characteristics of the raw material for the production of disposable tableware was determined. The basic parameters of our assessment were the contact angle and the free surface energy of the cardboard. Special attention was paid to these indicators as determining the degree of adhesion and hydrophilic properties of the material.

The results of the observations are given in Table 1, Fig. 8. As can be seen from the results of our measurements, lower contact angle indicators of the modified samples are observed compared to the untreated samples. Namely: for samples No. 3 – 116.1–116.6° and for samples No. 4 – 82.8–84.7° from the front side throughout the entire observation period, compared with sample No. 1, for which the contact angle reaches 112.1–115.9° and No. 2 – 91.8–93.4°, respectively. A larger contact angle is also observed from the back side: 120.0–121.1° for samples No. 3 and 119.4–120.5° for samples No. 4, respectively, throughout the entire observation period.

Our results indicate an increase in the wettability of the surface of the modified cardboard, which gives reason to expect an improvement in its adhesive properties. The results are consistent with works [18, 19]. In our study, the effect was on the surface of the cardboard, unlike studies [20, 21], in which modifiers were introduced mainly into the mass during its formation to change the surface properties of the cardboard. This approach changes the structure of the material but is not aimed at influencing the contact angle of the cardboard.

Lower values of the contact angle indicators testify to better adhesion between the polyethylene-coated and the side of the article that does not have a polymer coating at the stage of bonding the structural elements of the finished disposable tableware. At the same time, better surface wettability helps enable better adhesion of cardboard and polyethylene melt when applied to a cardboard sheet.

Our study of the surface charge of dissolved substances showed a significant (≈ 2–3 times) increase in this indicator for samples that were modified with a solution of aluminum hydroxychloride $Al_2(OH)_nCl_{6-n}$. This indicates the formation of a surface that is more energetically stable. This is consistent with works [10, 17], which show that the treatment of materials with secondary cellulose significantly affects the indicator of free surface energy and its components. At the same time, the increase in the surface charge of dissolved substances does not depend on the presence or absence of a polymer coating. Our research result makes it possible to obtain better indicators of adhesion of cardboard sheet and polymer in further technological operations.

As can be seen from Fig. 9, at approximately the same average indicators of the contact angle, the samples that were modified have significantly higher indicators of the surface charge of dissolved substances. This is a necessary prerequisite for increasing the adhesion between cardboard and the polyethylene melt both when it is applied to the cardboard sheet and when soldering the structural elements of the finished article.

Analysis of the results given in Table 2 and Fig. 10 proves that there is a relationship between the preliminary modification of cardboard with an aqueous solution of aluminum hydroxychloride $Al_2(OH)_nCl_{6-n}$ and a significant increase in the charge indicators of dissolved substances. Our results indicate better surface wettability and an increase in the surface energy of the studied samples, which is consistent with papers [3, 24–26].

Thus, a feature of the proposed modification approach is the combination of increasing surface wettability and increasing the surface charge of dissolved substances of the cardboard sheet for the manufacture of disposable products from it. This favorably distinguishes our results from the known approaches reported by colleagues.

As a result, preliminary surface modification of cardboard with an aqueous solution of aluminum hydroxychloride improves the wettability of the surface during subsequent polymer application. Such treatment will enable reliable bonding of structural elements of products during the manufacture of disposable tableware from cardboard and other products of short-term use.

The limitations of this study are attributed to the fact that our results relate to the manufactured series of cardboard samples and the conditions of their experimental modification with an aqueous solution of aluminum hydroxychloride $Al_2(OH)_nCl_{6-n}$. The adequacy and reproducibility of the obtained wettability and surface charge of dissolved substances are ensured within the limits of the investigated solution concentrations, drying, acclimatization modes, and used compositions of cardboard samples.

The results are relevant for the processes of manufacturing raw materials for disposable tableware and cardboard packaging products of short-term use with polyethylene coating. However, the results do not take into account the use of alternative polymer coatings.

The disadvantage of this study is the limited range of studied physicochemical parameters of the cardboard surface, which does not make it possible to fully characterize the mechanisms of the influence of modification on the surface of the cardboard sheet. In the future, this drawback could be eliminated by involving additional methods for analyzing the cardboard surface and expanding the range of studied indicators.

Further advancement of our study may involve improving the parameters for the surface treatment of cardboard and studying the influence of modification on cardboard with other polymer coatings. In this case, possible difficulties may include methodological and experimental complications associated with the reproducibility of results and the stability of the obtained effects when changing the technological modes of both cardboard production and the production of disposable products from it.

7. Conclusions

1. We have found that the surface treatment of cardboard with an aqueous solution of aluminum hydroxychloride makes it possible to influence the contact angle and obtain its lower values compared to unmodified samples, which contributes to an increase in its hydrophilicity. Specifically: for samples No. 3 and No. 4, which were modified with a solution of $Al_2(OH)_nCl_{6-n}$, the contact angle is within 116.1–116.6° and 82.8–84.7°, respectively, on the front side and 120.0–121.1° and 119.4–120.5° on the back side, respectively, throughout the entire observation period. Smaller values of the contact angle indicators contribute to ensuring better adhesion between the polyethylene-coated and untreated sides of the article at the next stage of bonding the structural elements of the finished disposable tableware. Our results from experimental studies of the contact angle indicate that the treated cardboard samples have better wettability. Accordingly, the treated samples have better adhesion to the polymer melt during subsequent technological operations, both in the case of untreated material and when applying a polyethylene coating.

During our study, it was proposed to use the coagulant aluminum hydroxychloride as a surface modifier of cardboard sheet. The principle is that $Al_2(OH)_nCl_{6-n}$ is usually used for the preparation of drinking water, wastewater treatment, etc. due to its ability to increase the surface energy of the treated substances.

2. It has been proven that the modification of cardboard with an aqueous solution of aluminum hydroxide chloride causes a significant increase in the surface energy (an increase in the surface charge of dissolved substances by 2–3 times) of samples that were modified with a solution of $Al_2(OH)_nCl_{6-n}$ compared to untreated samples. Namely, for samples No. 3 and No. 4 that were modified with a solution of $Al_2(OH)_nCl_{6-n}$, the indicator of the surface charge of dissolved substances is –51.3 μg-equiv./l for sample No. 3 and –54.1 μg-equiv./l for sample No. 4, respectively. This property will contribute to better contact between the cardboard surface and the polymer melt during its application and subsequent gluing of the structural elements of disposable tableware. Therefore, the proposed approach to modifying cardboard sheets is promising for further implementation in industrial processes, in particular in the production of packaging materials and disposable tableware. The use of such technology makes it possible to improve the barrier properties of materials and enable reliable adhesion of structural elements of products and their integrity during operation.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study and the results reported in this paper.

Funding

The study was conducted without financial support.

Data availability

The data will be provided upon reasonable request.

Use of artificial intelligence

The authors confirm that they used artificial intelligence technologies ChatGPT (OpenAI), a large language model, version GPT-5.2 when creating the presented work in terms of checking grammar, spelling, punctuation, without changing the text, as well as searching for sources by keywords.

Authors' contributions

Tetiana Roik: Conceptualization, Supervision; **Yevhenia Bychkar**: Methodology, Writing- review & editing, Validation; **Volodymir Oliynyk:** Methodology, Resources.

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