Experimental assessment of packaging paper properties effect in combination with storage conditions on the adhesion performance of automotive glass

Kordová T., Mareška A., Havlík Míka M.
Faculty of Chemical Technology, University of Chemistry and Technology, Prague
E-mail: kordovat@vscht.cz

The aim of this work was an experimental comparison of different types of packaging paper, determining the effect of packaging material properties in combination with storage conditions on the purity of glass surface and the related quality of adhesion of bonded materials. In the automotive industry, the quality of adhesion is a key factor, as automotive glass must meet the safety requirements. During the experiments, the glass sheets were wrapped in four types of paper and stored according to the conditions that are common in a real production. The research was focused on the influence of different composition of packaging papers on the resulting adhesive properties of glass sheets. According to the results obtained in this work, it can be stated that during the storage there is an interaction between paper and glass. According to XRF analyzes, ongoing corrosion of the glass can be inferred. The resulting adhesion was assessed based on a peel test, which took place under defined conditions given by standard customer specifications. It has been observed that the negative effect of unsuitable paper increases over time. For optimal adhesion it is necessary to emphasize the packaging materials and the storage conditions of the glass. The results and knowledge achieved in this work can significantly improve the quality of production in which a material is glued to the glass surface.

INTRODUCTION

The production of automotive glass includes the production of float glass, the processing of glass by tempering or lamination, and other operations in which additional materials are applied to the glass [1-3]. When joining materials, adhesion and cohesion are key properties of substrates. To adhere the material to the glass surface, it is necessary among other factors, that the surface is clean, free from dust and grease [4,5].

Paper directly affects the process of corrosion of the surface of the material that is wrapped in it. The aggressiveness of packaging materials is most affected by lignin content, pH, and the impact of environmental conditions. The main sources of paper acidity are alum or aluminum sulfate, which are used to size paper. Other sources of paper acidity are substances that remain in the paper after bleaching and delignification. In general, the degradation processes in packaging materials take place by different mechanisms and the individual mechanisms are applied to different degrees depending on the exposure conditions [6].

Many studies deal with glass dissolution and corrosion processes [7,8]. In experiments in these studies, the dissolution rate was determined as a function of temperature and pH [7,9]. The process of dissolving the glass matrix is influenced by the composition of the glass, temperature, pH value and the composition of the acting substances [10,11]. Alkaline solutions cause the glass to dissolve [12-15]. The principle could be interactions between cations and deprotonated hydroxyls, where instead of hydrogen is silicon, aluminum, or zirconium [15]. These processes could weaken the bond of silicon to oxygen in the glass network. As the reaction progresses more, net hydrolysis no longer limits the rate, but the effect of alkalis on changes in the glass has never been studied under these conditions [16-18]. Chemical weathering glass also involves the precipitation of carbonates [19]. These events are the subject of work examining the weathering of antique glass [20]. The reactions that take place between aqueous liquids and glass involve the dissociation of water into hydroxyl ion and hydronium ion, the penetration of a hydrated proton into the glass mesh, the diffusion and exchange of sodium and calcium ions with hydronium ions. The bond between oxygen and silicon is broken, creating silanol groups at the glass-solution interface. Anions from the corroding aqueous medium react with the ions released from the glass to form a precipitate [21].

Corrosion of glass occurs not only by aqueous solutions, but also by atmospheric humidity. Leaching

of the glass components can be limited using a suitable treatment, which includes dealkalization of the surface with gaseous substances [22]. Dealkalization is temperature dependent. The extraction of sodium from glass due to the use of sulfur dioxide gas and its dependence on air humidity and temperature were also studied in several works [22,23]. The resulting white deposit on the glass surface is associated with the formation of sodium sulfate [24]. The aim of the dealkalization process is to prevent leaching of alkalis and other undesirable glass components [25].

If the process of degradation of the glass material takes place in an acidic environment, the silica framework dissolves more than cation leaching. The pH value increases due to the accumulation of cations, which results in a slowing down of dissolution [26]. When the silicate material dissolves, a layer containing a large amount of silica is formed on the glass surface. Alkaline cations are leached in this layer and hydrogen ions are implemented instead [27-29]. According to recent studies, the surface layer can form with little or no dissolution of the silicate framework [30]. Material corrosion could occur through a direct process of dissolution and precipitation, as further observations suggest, which also refutes the traditional surface layer concept [31-34].

In addition to rainwater, other components such as chlorine, sulfuric acid ions, fly ash particles, etc. increase the corrosion of the glass [35]. The classical model of water corrosion of glass is described as the process of hydrolysis, hydration, ion exchange and recondensation of the hydrolyzed glass network. However, further work shows that the process involves still further dissolution and precipitation of silica on the mobile reaction front [36,37].

The experience has shown that the cleanliness of the glass at the end of the production process, the type and properties of the packaging paper, and the storage conditions are the most important for good glass surface condition. Resources directly related to the topic of the effect of packaging paper on the glass surface and the adhesion are very limited, and therefore this issue was chosen as the work task.

EXPERIMENTAL

Based on actual experience from the manufacturing process, the problem of adhesion between the glass and the bonded material is known, due to the use of a white sulfate paper or a sizing paper for packaging stored glass. Better results are obtained with an unbleached paper, which is brown. The aim was to determine the differences between the papers, their effect on the purity of the glass surface and on the quality of adhesion in the process of gluing to the glass.

Methods of analysis

Regarding the need to determine the elemental composition in the sample area and with a focus on the elemental composition, the XRF method was chosen. This analysis was also the most suitable for the analysis of smears, because it is not possible to directly create the necessary samples from tempered glass, because the breakage creates too small cubes, which are also degraded by glass fragments. A Panalytical Axios automatic sequential XRF spectrometer with Omnian software was used to analyze the composition of papers and smears from glass samples. The root mean square value was 0.005 % and the absolute deviation was in thousandths of percent.

Materials

In total, four types of packaging paper were tested (basic information from the manufacturer see in Tab. 1, composition see in Tab. 2). However, during testing, the number of tested papers varied depending on the testing phase. In the initial testing phase, only two types of paper were used, later the remaining two types were added to include a wider portfolio of packaging materials.

Tempered glass sheets were used for the purposes of the experiments. The size of each glass was 40×40 cm. Soda-lime-silica glass has been processed by the same process used in the automotive industry to produce automotive glass. The glass surface has not been treated or cleaned in any way, which guarantees the authenticity

Tab. 1. Information about tested samples

Sample identification	Paper 1	Paper 2	Paper 3	Paper 4
Color	white	brown	brown	beige
Type	sulfate paper	sulfate paper	sulfate paper	sizing paper
Weight [g/cm^2]	40.00	50.20	40.00	40.00
pH	5.80	5.22	5.00	4.80
Ash [%]	2.39	1.20	1.26	3.12
Extractible [%]	0.17	0.20	0.21	0.26

of the results. The results of the smear composition analysis were obtained by subtracting the reference values of the glass composition without wrapping it in a paper.

Tab. 2. Average paper composition in weight percent based on XRF analysis

Component	Paper 1	Paper 2	Paper 3	Paper 4
Na	0.073	0.112	0.076	0.030
Mg	0.026	0.103	0.028	0.185
Al	1.020	0.337	0.113	1.191
Si	0.762	0.105	0.013	1.007
P	0.004	0.000	0.003	0.010
S	0.022	0.136	0.111	0.169
Cl	0.027	0.000	0.005	0.022
K	0.026	0.019	0.009	0.038
Ca	0.036	0.111	0.182	1.358
Ti	0.011	0.000	0.000	0.041
Mn	0.000	0.006	0.012	0.002
Fe	0.010	0.000	0.000	0.029
Cu	0.000	0.000	0.000	0.002
Zn	0.000	0.000	0.000	0.001
La…Lu	0.002	0.002	0.001	0.000
W	0.000	0.000	0.004	0.000
Cellulose	**97.982**	**99.069**	**99.444**	**95.915**

Betaseal™ 8000-1F polyurethane adhesive was used to test the adhesion of the bonded material to the glass surface. It is a one-component moisture-cured adhesive. More detailed specifications are given in Tab. 3.

Tab. 3. Information about polyurethane adhesive Betaseal™ 8000-1F

Name	Betaseal™ 8000-1F
Color	black
Type	polyurethane prepolymers
Density [g/cm^3] (ISO 845)	1.22 – 1.26
Solid content [%]	≥98
Cure rate, after 48h [mm]	≥3.5
Processing Temperature [°C]	10 – 40
Stress at break [MPa] (ISO 1798)	9
Strain at break [%] (ISO 1798)	500
Shear modulus [MPa]	1 – 1.5
Lap shear strength, 7 days [MPa] (DIN EN 1465)	≥5
Shore hardness A (DIN 53505)	55 – 65

Betaprime™ 5500 was used as an adhesion promoter. This substance ensures the formation of a suitable bond between the glass and the adhesive. It is a one-step promoter. Further information on this substance is given in Tab. 4.

Tab. 4. Information about adhesion promoter Betaprime™ 5500

Name	Betaprime™ 5500
Color	black
Type	polyisocyanates
Density [g/cm^3] (ISO 845)	0.901 – 1
Solid content [%]	35 – 40
Open time [h]	0.05 – 72
Processing Temperature [°C]	10 – 40

Methods of tests

The following are two experiments. The first dealt with the comparison of a bleached and an unbleached paper. The second experiment included two additional types of paper in the test to broaden the pareto results and clarify which type of paper was most suitable for glass packaging and storage.

1st test – comparison of Paper 1 and Paper 2

The first test was based on the experience that the results are better in the case of the unbleached paper. Therefore, a test was performed during which wrapped pieces of glass were left for 33 days under defined conditions. The sample preparation process was such that one sheet of glass was wrapped in one sheet of tested paper. The packaging was not sealed in any way, the edges of the paper were only folded, as is usually packaged glass in production. Ten glass sheets were wrapped in the white paper (Paper 1). The glass packed in this way was transported on a pallet to a covered outdoor warehouse, where neither humidity nor temperature was controlled. These conditions were only recorded by a thermohygrograph. The humidity ranged from 72% to 93% and the average temperature was 18 °C, the details are shown in Fig. 1 and Fig. 2. At the end of this period, the glass pallets were transported and stored for 2 days in a room where humidity and temperature were controlled. The reason is the acclimatization of the glass, which corresponds to the real conditions of production. The humidity value was from 45% to 55% and the temperature ranged from 16 °C to 20 °C.

An important factor for comparing wrapping papers is water absorption (hydroscopicity and hygroscopicity) and drying speed. Water absorption is one of the key parameters of a packaging paper for the mentioned application. In this case, it was a simulation where the

packaged glass is stored in an uncovered place and the pallet may be exposed to weather conditions such as rain, drizzle, snow, or fog. During the water absorption test, it is measured as the rate of infiltration of a water drop of a certain volume by a paper sample. Samples of paper measuring 10 × 10 cm were prepared for the paper water absorption test. A sample of the paper was placed on the surface of the glass sheet. A drop of water with a defined volume of 0.05 ml was applied to a paper sample from a defined height of 10 cm. Dropping and soaking was detected by a high-speed camera. This experiment was not performed according to a standardized method, it was used only to compare paper samples, i.e., the white paper (Paper 1) and the brown paper (Paper 2). An analytical balance with a measurement accuracy of four decimal places was used to assess the hygroscopicity and drying rate. Hygroscopicity and drying time was tested by leaving paper samples under defined conditions at 25 °C and 65% relative humidity for 21 hours and 55 minutes. Samples were weighed before and after this test. According to the measured values, the weight increase was assessed, which corresponded to the absorbed air humidity, i.e., hygroscopicity. The drying rate of a paper was determined as the weight loss over time. Water removal from paper samples was performed using a desiccator. The weight of the paper samples was measured before being placed in the desiccator and after removal. As soon as the weight value remained unchanged, this experiment was terminated. Based on the measured values, the drying rate was calculated as a weight percentage per minute.

Based on previous experience and manufacturing knowledge, it has been found that adhesion problems are associated with the presence of various visible impurities on the glass surface. It was assumed that the impurities were caused by the white packaging paper (Paper 1), as this did not occur when using the brown paper (Paper 2). An analysis was performed to evaluate the composition of the impurities. Impurities from the surface of the glass, which was wrapped in the white paper (Paper 1), were analyzed by X-Ray fluorescence (XRF).

2nd test – comparison of four packaging papers

The second experiment involved four paper samples. In addition to the standard white paper (Paper 1) and the brown paper (Paper 2), another brown unbleached paper (Paper 3) was tested, as well as the beige sized paper (Paper 4), which was expected to have the lowest moisture content. Three containers with glass sheets wrapped in four types of paper (Paper 1, Paper 2, Paper 3 and Paper 4) were prepared. Each container contained 30 pieces of glass wrapped in one type of paper, so 120 pieces of glass in one container in total.

All three containers were stored in the same place in a covered warehouse and under the same conditions, i.e., without regulated temperature and humidity. Only the storage time differed and was based on the usual average storage lengths. The three storage lengths chosen were inspired by real production. The first container was stored in the warehouse for 14 days. The second container was stored under the same conditions for 56 days and the third container for 126 days. The storage conditions

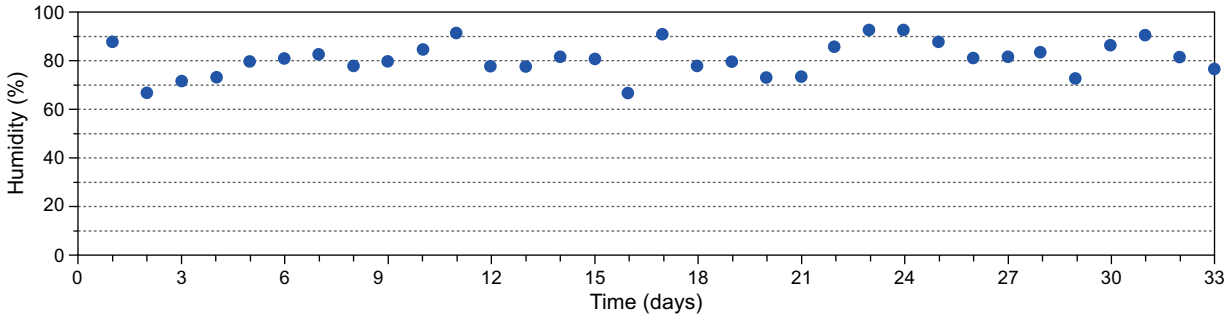

Fig. 1. Humidity during the 1st test measured by a local thermohygrograph

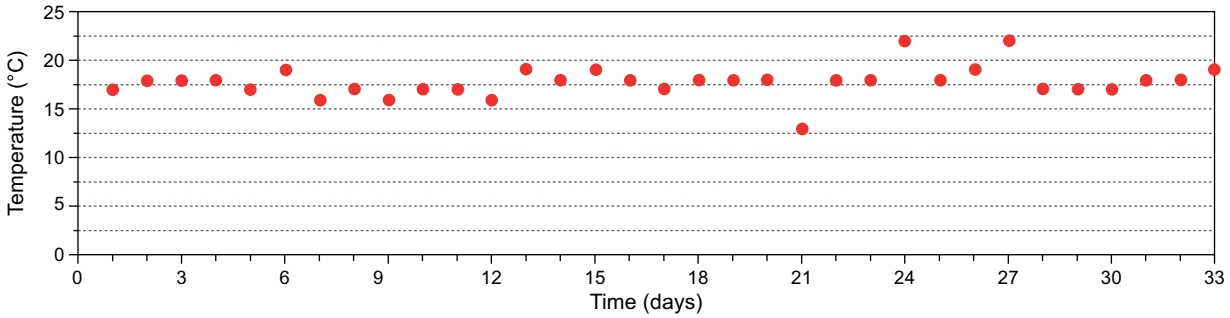

Fig. 2. Temperature during the 1st test measured by a local thermohygrograph

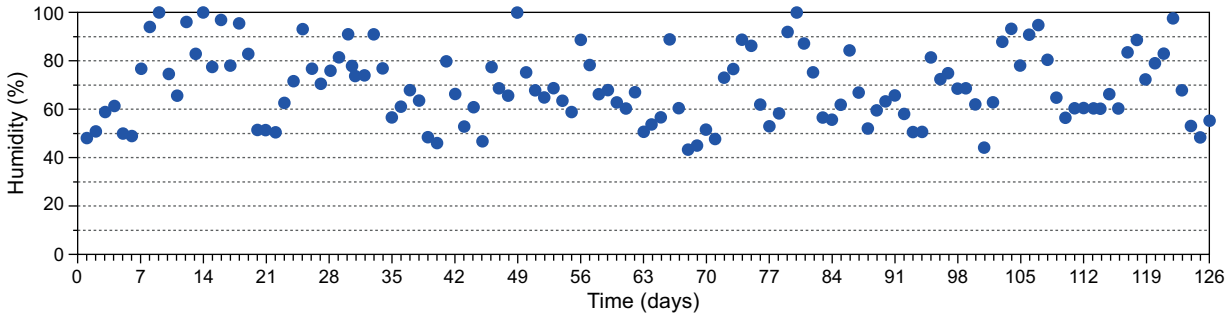

Fig. 3. Humidity during the 2nd test measured by a local thermohygrograph

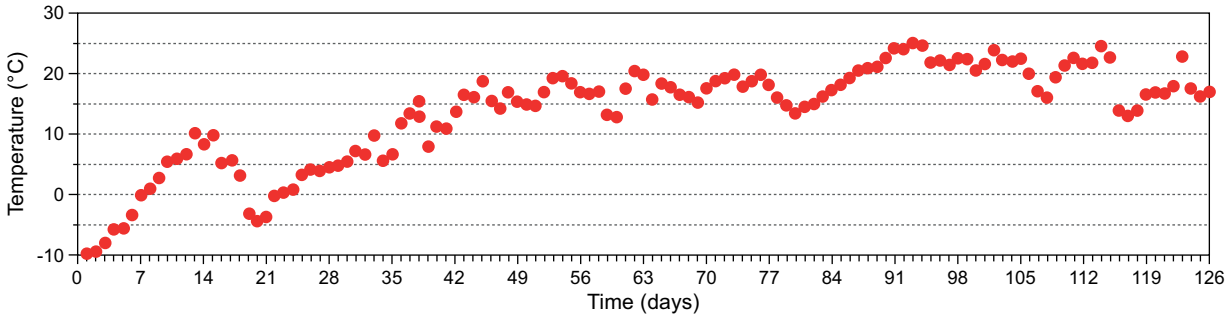

Fig. 4. Temperature during the 2nd test measured by a local thermohygrograph

were chosen based on the real conditions of storage of the glasses before further processing. Data from the thermohygrograph including temperatures and humidity in the tested time period are shown in Fig. 3 and Fig. 4.

From each series of samples wrapped in the same paper in one container, three sheets of glass were selected. A third glass, the fifteenth glass and the twenty-seventh glass were always removed from a package of glasses. Their surface was wiped with clean pulp measuring 3 × 3 cm. The strokes were performed in a total of fourteen rows so that the entire surface of the glass was wiped off. Samples of smears on pulps were used for composition analysis using the XRF method. For the evaluation, the analysis was performed again with the white paper in order to ensure the same conditions and comparability of the results of other tested papers.

The adhesion promoter Betaprime™ 5500 was applied to the remaining 27 glass sheets in five rows measuring 4 cm wide and 35 cm long. The product was allowed to ventilate according to the manufacturer's recommendations given in the technical data sheet. Then a defined amount of polyurethane adhesive Betaseal™ 8000-1F was applied to the glass. The width of the applied glue was given by the width of the application head of the cartridge, i.e., approximately 1.5 cm, the length of the glue was approximately 30 cm, and the height was approximately 0.5 cm. In total, glue was applied in five rows side by side to each glass.

The curing of the adhesive took place under the conditions recommended by the glue manufacturer. The humidity in the room was set at 50% and the temperature at 25 °C. The resulting adhesion was assessed in each case 72 hours after the application of the adhesion promoter and the adhesive, then after 7 days (normal conditions) and after a climatic test. The climatic test was performed by moistening each prepared glass and sealing it in a foil. The samples were placed in a chamber for 7 days and heated to 70 °C. They were then placed in a -20 °C chamber for 2 hours. Finally, the samples were placed in room temperature and after 24 hours a peel test was performed to assess adhesion. The peel test was performed by cutting the adhesive with a knife at first to allow it to be gripped by the hand. Subsequently, an upward force perpendicular to the glass surface was applied by hand. During the tension, it was assessed whether the joint failed adhesively, i.e., the adhesion of the adhesive to the glass, resp. adhesion promoter to the glass, or if there was a cohesive disruption of the adhesive mass. The percentage of cohesive or adhesive failure was obtained as the ratio of the appropriate failure area to the total area.

RESULTS

Results of the 1st test – comparison of Paper 1 and Paper 2

The result of the comparison of papers in terms of water absorption was such that in the case of the brown paper (Paper 2) the water was absorbed significantly more slowly than in the case of the white paper (Paper 1). Exactly it was 2.21× slower than the absorption of

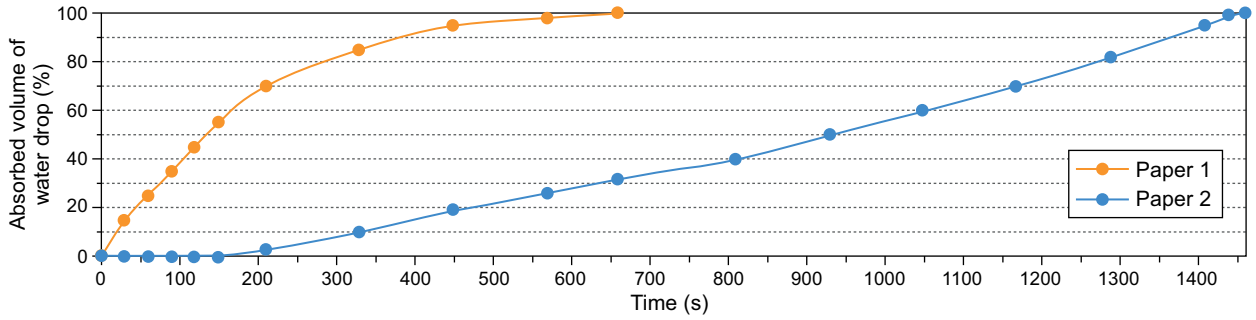

Fig. 5. Water absorption rate over time expressed as a water volume percentage for the white paper (Paper 1) and the brown paper (Paper 2)

a drop by the white paper (Paper 1). The process of water absorption was assessed according to the recording of a camera. A drop of water was recorded from the profile to determine the rate of absorption. The record was then evaluated by calculating the percentage absorption of the droplet over time. This calculation was performed as a slice content corresponding to a water drop profile. Based on this data, a graph was compiled, which is shown in Fig. 5. After absorbing the entire volume of the water drop with the paper, this time was used to calculate the absorption rate expressed as the total water drop volume over time. The character of the absorption curve of the drop with the white paper (Paper 1) was logarithmic, in the case of brown paper (Paper 2) no change in the volume of the drop was observed from the beginning during the first 150 s, then the curve was gradual.

Tab. 5. Results of the test to determine the hygroscopicity in the defined conditions: temperature 25 °C, humidity 65% and time 21 hours and 55 minutes. Measured data for the white paper (Paper 1) and the brown paper (Paper 2)

	Weight of Paper 1 [g]	**Weight of Paper 2 [g]**
Paper before testing	6.4568	7.4550
Paper after testing	8.1373	9.0516
Weight gain (+ H_2O)	1.6805	1.5966
Relative weight gain (+ H_2O)	26.03 wt. %	21.42 wt. %

Tab. 6. Results of the test to determine hygroscopicity, hydroscopicity and drying speed in the defined conditions: temperature 25 °C, humidity 65% and time 21 hours and 55 minutes. Measured and calculated data for the white paper (Paper 1) and the brown paper (Paper 2)

Paper	**Hygroscopicity [wt. %/min]**	**Hydroscopicity [cm³/s]**	**Drying speed [wt. %/min]**
Paper 1	0.019795	7.58E-05	2
Paper 2	0.016289	3.42E-05	4

The weight gain after 21 hours 55 minutes at 25 °C and 65% relative humidity is in Tab. 5 for both test papers (Paper 1 and Paper 2). The calibrated KERN ABJ 320-4NM analytical scale with 0.1 mg resolution was used to weighing samples. The resulting values for measuring hygroscopicity, hydroscopicity and drying rate are summarized in Tab. 6.

The drying speed of the brown paper (Paper 2) was higher. It was 1 wt. % per 15 seconds. In the case of the white paper (Paper 1) the drying speed was approximately 0.5 wt. % per 15 seconds.

The results of the composition of the wiped glass surface after unpacking from the white paper are summarized in Tab. 7.

The wiped surface contains sodium, aluminum, magnesium and calcium, and increased content of silica and sulfur.

Some of the identified elements may come from the paper bleaching process in which for example, chlorine and its compounds, aluminum compounds, Satin white ($CaSO_4 \cdot Ca(AlO_2)_2$) and titanium white (TiO_2) are used [6].

Tab. 7. Average X-ray fluorescence results of wiped glass surface for the white paper sample. The surface has been wiped with a clean pulp measuring 3×3 cm in a total of fourteen rows to ensure that the entire surface of the glass was wiped off. The following values were obtained by subtracting the composition of the reference pure pulp from the result of the pulp analysis after wiping the glass surface in weight percent

Component	**Quantity of elements**
	Paper 1
Na	0.457
Mg	0.015
Al	0.029
Si	0.071
S	0.050
Cl	0.009
Ca	0.021
Sum La…Lu	0.001
W	0.008

Results of the 2nd test – comparison of four packaging papers

For each storage time and for each paper, the average wiped surface composition results were evaluated by XRF analysis. A graphical representation of the development of individual elements in the given test periods is for each tested paper in Fig. 6-9. The scale was adjusted according to the achieved values.

According to the obtained data, the trend is that with increasing storage time, the content of individual elements increased for all tested papers. An exception was made for some minority elements, with no or only minimal changes.

Based on the summary of the XRF analysis in Fig. 10, the most of the residues on the glass surface are left behind by the sizing paper (Paper 4). The second worst result was determined for the white paper (Paper 1). The best results were obtained using the brown papers (Paper 2 and Paper 3), both of which had almost the same amount of elements left on the glass. The standard white paper (Paper 1) left on the glass surface sodium, aluminum, and silicon the most, followed by magnesium, calcium, sulfur, chlorine and potassium. These identified elements could come from a layer of corrosion products on the glass surface.

Adhesion of the polyurethane adhesive to the glass, resp. adhesion of the promoter to the glass was considered as a percentage of the area where the joint cohesively failed. By cohesive failure is meant rupture inside the adhesive. The adhesive failure, on the other hand, is the peeling of the glue, resp. adhesion promoter, from the

Tab. 8. Average values of cohesive joint failure for each tested paper and for three tests – peel test after 72 hours (normal conditions), after 7 days (normal conditions) and after climatic test (70 °C for 7 days, then -20 °C for 2 days and finally 24 hours at room temperature)

Peel test conditions	**Cohesive failure [%] (average values)**			
	Paper 1	**Paper 2**	**Paper 3**	**Paper 4**
after 72 hours	75%	100%	100%	10%
after 7 days (normal conditions*)	80%	100%	100%	10%
after climatic test**	50%	95%	90%	0%

**Normal conditions – room temperature and normal humidity, i.e., 25 °C and 50% RH.*
***Climatic test – 70 °C/7 days; -20 °C/2 days; 24 hours in room temperature.*

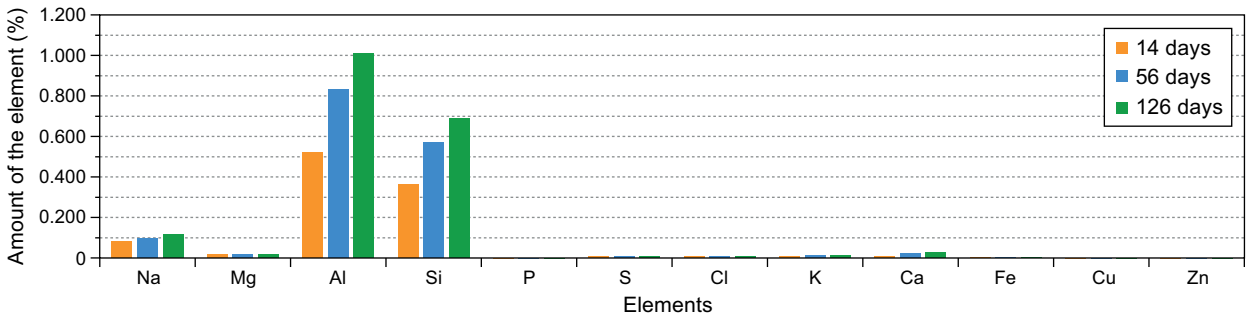

Fig. 6. Development of the content of individual elements determined by XRF analysis for Paper 1 and time periods of 14 days, 56 days, and 126 days. The y-axis scale was adjusted according to the achieved range of values

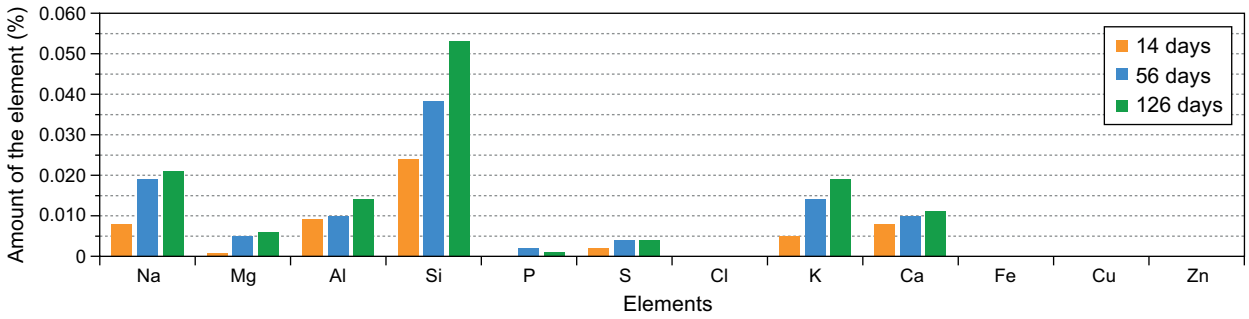

Fig. 7. Development of the content of individual elements determined by XRF analysis for Paper 2 and time periods of 14 days, 56 days, and 126 days. The y-axis scale was adjusted according to the achieved range of values

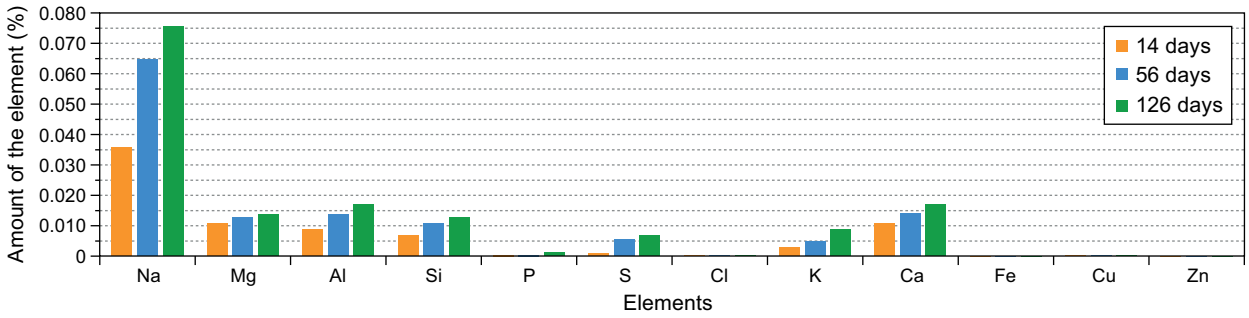

Fig. 8. Development of the content of individual elements determined by XRF analysis for Paper 3 and time periods of 14 days, 56 days, and 126 days. The y-axis scale was adjusted according to the achieved range of values

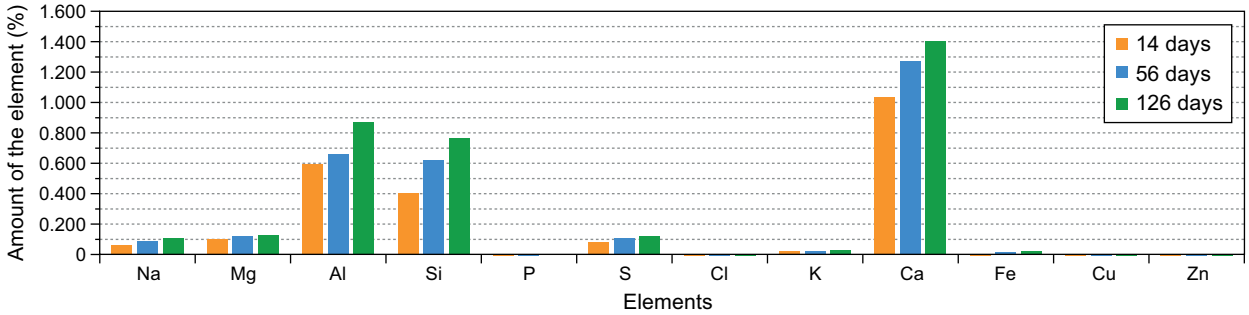

Fig. 9. Development of the content of individual elements determined by XRF analysis for Paper 4 and time periods of 14 days, 56 days, and 126 days. The y-axis scale was adjusted according to the achieved range of values

glass surface. Therefore, if the adhesion failure is 100%, the cohesive failure is 0%. The summary of results is together with the percentage of cohesive violation in Tab. 8.

The peel adhesion test showed that the brown papers (Paper 2 and Paper 3) had the best result. The sizing paper was the worst (Paper 4) as this sample failed even the least demanding test. The white paper (Paper 1) achieved sufficient adhesion only in the case of the second test (peel test after 7 days under normal conditions).

DISCUSSION

According to the hygroscopicity testing and drying of paper samples, it was determined that the bleached paper (Paper 1) absorbs water faster and at the same time dries twice as slowly as the brown paper (Paper 2). Therefore, the unbleached paper (Paper 2) should be more usable in the production process, where it is essential that the moisture content is as low as possible to avoid interactions in the paper and between the paper and the glass surface. During the storage of glass sheets in the white paper (Paper 1), the glass surface was contaminated with elements that could correspond to the additives used in the bleaching process, as well as possible corrosion processes on the glass surface.

For the second test, the same papers were used as in the first test (Paper 1 and Paper 2) and two more samples (Paper 3 and Paper 4) were added to ensure comparability of results and to expand the sample portfolio. These added samples were another unbleached brown paper (Paper 3), which was a cheaper variant of the Paper 2, and a sizing paper (Paper 4), which was supposed to excel in its low water and moisture absorption. Based on the results of the XRF analysis in Fig. 10, it was stated that the largest amount of residues remained on the glass

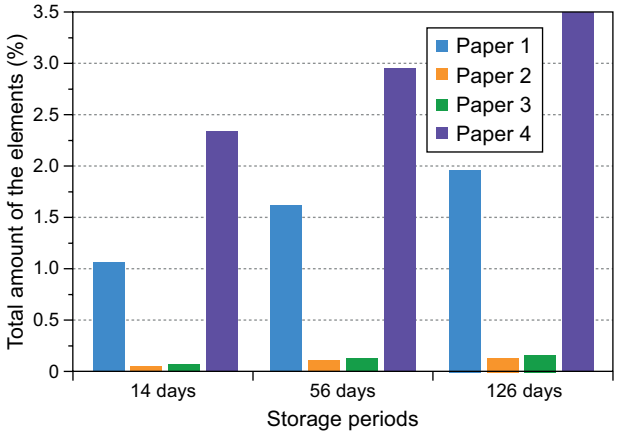

Fig. 10. Summary results of XRF analysis of the wiped surface after 14 days, 56 days and 126 days of storage after subtracting the clean pulp reference sample composition for all tested samples in weight percent

sheets wrapped in the sizing paper (Paper 4) and the white paper (Paper 1) left slightly less contamination. The least residues remained on the glass using brown unbleached papers (Paper 2 and Paper 3). After plotting the data in graphs Fig. 6-9, it was possible to observe a trend where the number of majority elements increased with the length of storage. A similar trend as in the results of the wiped surface analysis was observed during the peel test, in which the quality of adhesion was evaluated. Samples wrapped in the sizing paper (Paper 4) showed the worst adhesion, poor adhesion was also in the case of the white paper (Paper 1), on the contrary, good results were obtained with brown papers (Paper 2 and Paper 3).

According to the above results and based on the research, it is very likely that the bleached paper (Paper 1) has a negative effect on the surface properties of glass. Due to the properties of this paper, it can be concluded that this product supports corrosion processes and interacts negatively with the glass surface.

Corrosion and weathering produce corrosion products (silicates, sulfates, nitrates, etc.). The speed of the corrosion process depends on the rate of temperature changes and other environmental influences. The mechanism of corrosion (Fig. 11) involves the leaching of alkalis from the glass surface and the exchange for H^+ (H_3O^+) ions (interdiffusion of $Na^+ \leftrightarrow H^+$); dissolution of the SiO_2 network; and precipitation of corrosion products on the glass surface. The resulting corrosion effect is affected by the speed and kinetics of each process. Corrosion of glass depends on the composition of the glass, the composition and characteristics of the corrosive environment, the duration of the corrosive environment effect and the temperature. If the temperature and humidity of the air change rapidly (alternating night and day), water condenses on the surface of the glass. Other components of air (SO_2, NO_x, CO_2, UV radiation, etc.) also affect these processes. Corrosion products are formed by the chemical reaction of the glass material with the environment. The layers can be formed by the diffusion of alkalis from the surface of the glass (SiO_2 layer), being part of the glass material. Furthermore, there are layers not belonging to the original material, these corrosion products are formed by precipitation of glass components with the surrounding components. Diffusion of alkalis to the glass surface produces soluble alkali salts (e.g., sodium bicarbonate, sodium sulfate, sodium carbonate) which can be washed with water [38-40].

Deterioration of adhesion could be caused by the presence of impurities on the glass surface. If an adhesive promoter and a polymer are applied to the glass, it could bind to the glass and to impurities on its surface. Therefore, it is important that the glass surface is clean, free of dirt, grease, and other impurities. The possible principle of breaking the adhesion of the adhesive is shown in Fig. 12.

$$\underset{\text{(water)}}{H^+} + \underset{\text{(glass)}}{\equiv Si{-}O^-Na^+} \leftrightarrow \underset{\text{(glass)}}{\equiv Si{-}OH} + \underset{\text{(water)}}{Na^+} \quad (1)$$

$$\underset{\text{(glass)}}{\equiv Si{-}O{-}Si\equiv} + H_2O \leftrightarrow 2\ \underset{\text{(glass)}}{\equiv Si{-}OH} \quad (2)$$

$$\underset{\text{(glass)}}{H_2O} + \underset{\text{(glass)}}{\equiv Si{-}O^-Na^+} \leftrightarrow \underset{\text{(glass)}}{\equiv Si{-}OH} + \underset{\text{(water)}}{Na^+OH^-} \quad (3)$$

Fig. 11. Example of reactions that can occur during the glass corrosion process: an ion exchange process (1), the hydrolysis of the silicate network (2) and the dissociation of molecular water on the non-bridging oxygen sites (3) [40]

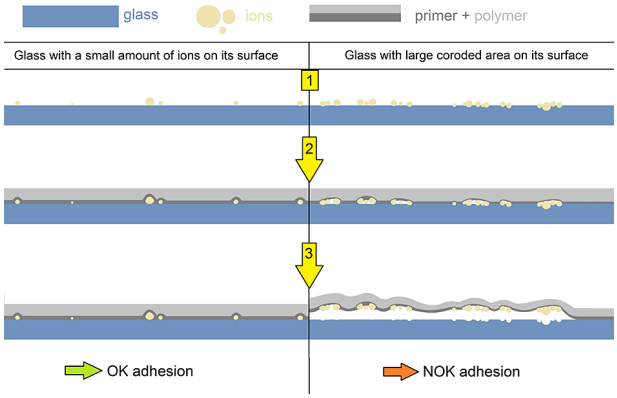

Fig. 12. A possible principle of adhesion failure due to the presence of impurities on the glass surface (illustrative picture)

CONCLUSIONS

The precondition for impaired adhesion when using a standard white paper was the interaction between the packaging material and the glass under storage conditions. Empirically obtained practical experience has shown that the use of an unbleached brown paper does not cause so many adhesive failures. The idea of this work was to determine and verify the differences between the papers used, to determine their effect on the glass surface and on the quality of the resulting adhesion when used for gluing adhesives to glass.

Based on the results of the experiment, it is possible to assume that corrosion processes of glass occur when it is wrapped in a packaging paper. Worse bleached paper (Paper 1) results could be associated with the fact that this sample absorbed more humidity and dries more slowly than the unbleached paper sample (Paper 2). The brown paper samples (Paper 2 and Paper 3) also contained more cellulose than the white paper (Paper 1). Ion exchange appears to occur, with the products of interactions settling on the glass surface and forming a visible haze. Comparing all four papers, which included one bleached paper (Paper 1), two unbleached papers (Paper 2 and

Paper 3) and one sizing paper (Paper 4), it was observed that the amount of residues on the glass surface increases over time. The total content of contaminants depended on the parameters and purity of the packaging paper.

The results and knowledge achieved in this work can significantly improve the quality of production in which a material is glued to the glass surface. In addition, the choice of quality packaging materials can save the cost of cleaning and pre-treatment of the glass surface before further operations. For the automotive industry, the quality of adhesion is a key factor, as automotive glass also plays a role in the safety of the car. To achieve good adhesion, not only a clean glass surface is required before the gluing process, but emphasis must also be placed on the quality of the materials in which the glass sheets are packaged.

REFERENCES

1. De Felice, F.; et al. Optimization of automotive glass production through business process reengineering approach. *Procedia – Social and Behavioral Sciences* **2013**, *75*, 272–281.
2. Pariafsai, F. A review of design considerations in glass buildings. *Frontiers of Architectural Research* **2016**, *5* (2), 171–193.
3. Shand, E. B. *Glass Engineering Handbook*, 2nd ed.; McGraw-Hill handbooks: New York, 1958.
4. Petrie, E. M. *Handbook of Adhesives and Sealants*, 2nd ed.; McGraw-Hill handbooks: New York, 2007.
5. Marshall, S. J.; et al. A review of adhesion science. *Dental Materials* **2010**, *26* (2), 11–16.
6. Strachotova, K. Ch.; et al. Effect of the presence of corrosion products on the corrosion rate of lead during the exposition with paper packaging materials. *Koroze a Ochrana Materiálu* **2018**, *62*(3), 87–96.
7. Backhouse, D. J.; et al. Corrosion of the International Simple Glass under acidic to hyperalkaline conditions. *Npj Materials Degradation* **2018**, *2* (1).
8. Goût, T. L.; et al. Diffusive processes in aqueous glass dissolution. *Npj Materials Degradation* **2019**, *3* (1).
9. Neeway, J. J.; et al. The dissolution behaviour of borosilicate glasses in far-from equilibrium conditions. *Geochim. Cosmochim. Acta* **2018**, *226*, 132–148.
10. Boksay, Z.; et al. Diffusion processes in surface layers of glass. *Phys. Chem. Glasses* **1967**, *8*, 140–144.
11. Sanders, D. M.; Hench, L. L. Mechanisms of glass corrosion. *J. Am. Ceram. Soc.* **1973**, *56*, 373–377.
12. Dove, P. M. The dissolution kinetics of quartz in aqueous mixed cation solutions. *Geochim. Cosmochim. Acta* **1999**, *63*, 3715–3727.
13. Dove, P. M.; Crerar, D. A. Kinetics of quartz dissolution in electrolyte solutions using a hydrothermal mixed flow reactor. *Geochim. Cosmochim. Acta* **1990**, *54*, 955–969.
14. Dove, P. M.; Nix, C. J. The influence of the alkaline earth cations, magnesium, calcium, and barium on the dissolution kinetics of quartz. *Geochim. Cosmochim. Acta* **1997**, *61*, 3329–3340.
15. Jollivet, P.; et al. Forward dissolution rate of silicate glasses of nuclear interest in clay-equilibrated groundwater. *Chem. Geol.* **2012**, *330*, 207–217.
16. Vienna, J. D.; et al. Current understanding and remaining challenges in modeling long-term degradation of borosilicate nuclear waste glasses. *Int. J. Appl. Glass Sci.* **2013**, *4* (4), 283–294.
17. Gin, S. Open Scientific Questions about nuclear glass corrosion. *Procedia Mater. Sci.* **2014**, *7*, 163–171.
18. Collin, M.; et al. Impact of alkali on the passivation of silicate glass. *Npj Materials Degradation* **2018**, *2* (1).
19. Matter, J. M.; Kelemen, P. B. Permanent storage of carbon dioxide in geological reservoirs by mineral carbonation. *Nature Geoscience* **2009**, *2* (12), 837–841.
20. Heimann, R. B. Weathering of ancient and medieval glasses – potential proxy for nuclear fuel waste glasses. *Int. J. Appl. Glass Sci.* **2018**, *9* (1), 29–41.
21. Heimann, R. B. *Classic and Advanced Ceramics: From Fundamentals to Applications* [online]; Wiley-VCH Verlag GmbH & Co. KGaA: Federal Republic of Germany, 2010. (accessed Dec 14, 2021).
22. Plesingerova, B.; et al. Extraction of sodium from the surface of glass by the products of decomposition of ammonium sulphate. *Ceramics* **1994**, *38* (3-4), 137–141.
23. Mochel, E. L.; et al. Strengthening of Glass Surfaces by Sulfur Trioxide Treatment. *J. Am. Ceram. Soc.* **2006**, *49* (11), 585–589.
24. Zhao, J.; et al. Glass Delamination: a Comparison of the Inner Surface Performance of Vials and Pre-filled Syringes. *AAPS PharmSciTech* **2014**, *15* (6), 1398–1409.
25. Rybarikova, L. Reduction of lead leaching from lead crystal glass. *Ceramics* **1993**, *37* (4), 193–197.
25. Wang, Y.; et al. Nonlinear dynamics and instability of aqueous dissolution of silicate glasses and minerals. *Scientific Reports* **2016**, *6* (1).
26. Dran, J. C.; et al. Mechanism of aqueous dissolution of silicate glasses yield by fission tracks. *Nature (London)* **1986**, *319* (6053), 485–487.
28. Doremus, R. H. Interdiffusion of hydrogen and alkali ions in a glass surface. *J. Non-Cryst. Solids* **1975**, *19*, 137–144.
29. Petit, J. C.; et al. Hydrated-layer formation during dissolution of complex silicate glasses and minerals. *Geochim. Cosmochim. Acta* **1990**, *54*, 1941–1955.
30. Gin, S.; et al. Origin and consequences of silicate glass passivation by surface layers. *Nature Communications* **2015**, *6* (1).
31. Putnis, A. Sharpened interface. *Nature Materials* **2015**, *14*, 261–262.
32. Hellmann, R.; et al. Nanometre-scale evidence for interfacial dissolutionreprecipitation control of silicate glass corrosion. *Nature Materials* **2015**, *14*, 307–311.
33. Ruiz-Agudo, E.; et al. Mechanism of leached layer formation during chemical weathering of silicate minerals. *Geology* **2012**, *40* (10), 947–950.
34. Geisler, T.; et al. The mechanism of borosilicate glass corrosion revisited. *Geochim. Cosmochim. Acta* **2015**, *158*, 112–129.
35. Lefevre, R. A.; et al. Modern and ancient glass in the polluted atmosphere. What is the prevailing phenomenon? Leaching, corrosion, soiling, encrusting...?. *Cultural Heritage Conservation and Environmental Impact Assessment by Non-Destructive Testing and Micro-Analysis*, 1st ed.; CRC Press: London, 2005; pp 77–89.
36. Geisler, T.; et al. Aqueous corrosion of borosilicate glass under acidic conditions: a new corrosion mechanism. *J. Non-Cryst. Solids* **2010**, *356* (28).

37. Deng, L.; et al. Ion-exchange mechanisms and interfacial reaction kinetics during aqueous corrosion of sodium silicate glasses. *Npj Materials Degradation* **2021**, *5* (1).
38. Devreux, F.; et al. Insight into silicate-glass corrosion mechanisms. *Nature Materials* **2008**, *7* (12), 978–983.
39. Tournié, A.; et al. Glass corrosion mechanisms: A multiscale analysis. *Solid State Ionics* **2008**, *179* (38), 2142–2154.
40. Alloteau, F.; et al. New insight into atmospheric alteration of alkali-lime silicate glasses. *Corros. Sci.* **2017**, *122*, 12–25.