# INFLUENCE OF LIGHT AND TEMPERATURE ON OPTICAL PROPERTIES OF PAPERS

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Paper still represents one of the most significant carriers of information. In order to preserve printed information, the resistance of paper should be considered. The aim of the present study was to examine the cumulative effect of light and temperature on the optical properties of four often used paper grades. Office, recycled, synthetic and permanent papers were examined, and their properties were evaluated based on spectrophotometric measurements after exposure to xenon light at two different temperatures to simulate the conditions of use and storage of documents. Results showed that the effect of temperature alone was not negligible, however, light caused the most evident changes. Paper samples that contained cellulosic and recycled fibres were more affected by the destructive influence of light, a significantly smaller impact was observed on synthetic and permanent papers. Colour differences, which occurred on papers after exposure to light, were calculated using CIEDE2000 equation and they were in agreement with the changes in whiteness and yellowness of the papers.

***Keywords***: paper, optical properties, stability, light, temperature, CIEDE2000

## INTRODUCTION

Paper still represents one of the most important carriers of written and printed information.[1-3] Due to the constant strive for a better printing quality and fast reproduction, manufacturers are endeavouring to improve the quality of paper.[4-5] Consequently, the market offers a broad spectrum of papers for different purposes. A vast multitude of contemporary materials is available, which could eventually become a part of important documents.

Conventional paper is mostly made of cellulose with a small amount of organic and inorganic additives.[6] Users are increasingly aware of ecological aspects and to contribute to a sustainable world they often choose environmentally friendly recycled papers. During the last few years, synthetic papers became more present due to their specific advantages. Synthetic papers are made of synthetic fibres alone, as well as in combination with cellulose fibres and additives to complement the deficiency in compatibility between polar natural and non-polar polymer fibers.[7]

Several studies have been carried out in relation to the permanence and durability of paper.[1,6,8-16] Permanence refers to the chemical stability of paper, durability, on the other hand, relates to the stability of paper as regards its physical and mechanical properties.[17] Paper has a low rate of degradation, therefore, the purpose of durability tests is to speed up the process of degradation and to establish standardised and reproducible conditions, as well as to predict the long-term effect of external factors on materials.[12,18] Different methods were proposed to predict the deterioration of synthetic and cellulosic polymer materials.[3-4,6-8,10-12,14,17-19]

In order to initiate a photochemical reaction, the energy of light should be present. Different wavelength regions induce photochemical deterioration of cellulose-based materials.[12] Photodegradation is not the only process responsible for the degradation of cellulose-based materials, the process of degradation also includes acid hydrolysis, oxidation, and biodegradation.[6] The presence of other factors contributes to the level of degradation of paper.[6,12,20] For example, the rise in temperature alone does not cause a photochemical reaction, but higher temperature accelerates the degradation process.[6,12] During its lifecycle, the paper may be exposed to higher temperature for a longer or shorter period of time. Therefore, for the preservation of paper, temperature conditions are very important. All the damage caused by light is irreversible and cumulative. Changes that occur due to external factors can be estimated through various analytical methods. Colorimetric measurements represent a non-destructive way to establish the changes of the material and allow monitoring the process of degradation.[6,12,21-24]

The purpose of the present research was to study the cumulative effect of light and temperature on the optical properties of paper under conditions similar to those of real use and storage of documents. Samples of four often used paper grades, which differed as to chemical structure, colour, grammage and manufacturer, were investigated: office, recycled, synthetic and permanent paper. They were exposed to controlled conditions of simulated daylight, temperature and relative humidity in order to evaluate the influence of different external factors and to determine to what extent a higher temperature affects paper's resistance to light.

Changes of optical properties after exposure were evaluated using CIELAB colour coordinates and CIEDE2000 equation for colour differences, together with parameters of whiteness and yellowing index. The roughness of paper was measured since it was recognized as one of the most important paper properties, which also affects its optical properties.[4] The aim of the research was to examine the stability of the four paper grades under different surrounding conditions in order to give recommendations regarding their use and storage.

## EXPERIMENTAL

### Materials

#### *Paper*

Four different categories of paper were chosen for the experimental part. The first sample represented the most widely used office paper (OP); it was uncoated paper with grammage of 80 g/m$^2$.

The second paper type (RP) represented the category of recycled papers with a coated surface and grammage of 100 g/m$^2$. Paper RP is made of 100% recycled fibres, matt-coated, natural white without optical brightening agents (OBA) and according to the manufacturer's assurances, it has a broad range of use.

The third category, synthetic papers, was represented by paper (SP), which has a coated surface and is made from selected pulps and synthetic fibres (polyamide and polyester), using a special impregnation method. According to the manufacturer's statement, paper SP is thermally and dimensionally stable, weather-proof and has several other favourable characteristics.

The fourth category represents permanent papers. Paper PP is made in accordance with the standard permanence and durability requirements on a pilot paper mill.[25-26] It contains no optical brightening agents.

### Methodologies

#### *Exposure of paper samples to light*

Light fastness of papers was estimated in two separate procedures, at temperature T = 35 °C and T = 60 °C. Xenotest Alpha (Atlas, USA) was used, with a xenon arc lamp, which simulates intensive daylight at selected temperature and constant relative humidity, RH = 35%. The paper samples were exposed to xenon light for 24, 72 and 144 hours. In order to establish the influence of heat alone, one half of the samples were protected with non-transparent material.

#### *Measurements of optical properties of paper*

The measurements were performed using a Datacolor SF 600 spectrophotometer (Datatacolor, Switzerland) under the following conditions: SAV (2r = 9 mm), d/8°, D65/10°.

Among colorimetric properties, the CIELAB colour coordinates L$^*$, a$^*$, b$^*$, chroma, C$^*$, and hue, h, were determined. For each paper sample, 10 measurements were performed. All measurements were made on the upper side of the papers only. The colour differences (ΔE) between the exposed and non-exposed samples were calculated according to the CIEDE2000 equation (*cf.* Eq. 1).[27] CIE whiteness (W) was determined according to standard ISO 11475:1999 (*cf.* Eq 2).[28] Yellowing index (YI) was calculated according to ASTM method 313 (*cf.* Eq. 3).[29]

$$\Delta E_{00}^{*} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T\left(\frac{\Delta C'}{k_C S_C}\right)\left(\frac{\Delta H'}{k_H S_H}\right)} \quad (1)$$

$$W_{10} = Y_{10} + 800 \times (x_{n,10} - x_{10}) + 1700 \times (y_{n,10} - y_{10}) \quad (2)$$

$$YI = 100 \times \left[1 - \frac{0.847 \times Z}{Y}\right] \quad (3)$$

Specular gloss was measured using a reflectometer ZGM 1022 Gloss meter 20°-75° (Zehntner, Germany). All measurements were made only on the exposed side of papers in accordance with ISO 8254-1 standard.[30] The results represent the mean value of ten measurements at an angle of 75° to the normal to the paper surface for all four directions.

#### *Measurements of roughness*

Roughness was measured according to ISO 4287 standard with a TR200 profilometer (Time Group Inc., USA).[31] All measurements were made on the exposed side of the papers. The measured values (Ra) are the average of ten measurements and represent the arithmetic average deviation of paper surface.

***Scanning Electron Microscopy (SEM)***

To obtain information about changes on the surface of papers, a 6060LV scanning electron microscope (JEOL, Japan) was used. The samples were cut to a diameter of 10 mm and mounted on a 10 mm cylindrical sample holder. To create conductive gold coating on the surface of paper samples, a sputter coater was used. Image scanning started after loading the sample into the properly evacuated chamber of the microscope, at 500× magnification and accelerating voltage of 15 kV.

## RESULTS AND DISCUSSION

Figure 1 shows the measured reflection of the samples before and after exposure to light under controlled conditions of temperature (T = 35 °C or T = 60 °C) and relative humidity (RH = 35%).

The most evident changes can be observed on office (OP) and recycled (RP) paper. In both cases, the most affected is the short-wave part of the spectrum and this can be seen as a colour change. The colour of papers is described by the coordinates of the CIELAB colour space. They were calculated from the measured spectra and are shown in Table 1. The changes of colour, which occur during exposure to light, can be calculated using different equations for colour differences. Figure 2 shows colour differences CIEDE2000 (Eq. 1) for the four paper samples. It is evident that the office paper (OP) exhibits the biggest changes. The colour differences on OP gradually increase with time and after 144 hours of exposure to light at temperature T = 60 °C, they reach slightly more than 9 units, which means that such changes would be clearly visible. At lower temperature (T = 35 °C), the effect of light is less destructive, but still visible as the colour difference after 144 hours is close to 7 units. Such changes are comparable to the differences observed on the recycled paper (RP) already after a shorter period of exposure, *i.e.* 72 hours, or after only 24 hours of exposure to light at higher temperature (T = 60 °C). The differences on RP occur very quickly, but afterwards remain practically the same. Only moderate colour differences ($\Delta E < 2$) were observed on the synthetic paper (SP). In spite of the fact that changes occur already after 24 hours, the colour differences do not increase with time. The effect of higher temperature (T = 60 °C) is not as evident as that seen on OP and RP. Much smaller colour differences were calculated for permanent paper (PP) and they would probably not even be noticed as the values $\Delta E$ are below 1 unit.

Table 1

CIE L*, a*, b*, C*, whiteness (W), yellowness (YI) for exposed and non-exposed papers before (standard) and after 144 hours of exposure to light at temperatures of 35 °C and 60 °C

| | | T (°C) | L* | a* | b* | C* | W | YI |
|---|---|---|---|---|---|---|---|---|
| OP | standard | | 93.23 | 3.17 | –15.45 | 15.77 | 153.63 | –25.28 |
| | exposed samples | 35 | 92.42 | 0.41 | –6.48 | 6.50 | 111.56 | –9.14 |
| | | 60 | 92.44 | –0.29 | –3.14 | 3.16 | 96.21 | –3.47 |
| | non-exposed samples | 35 | 93.24 | 3.19 | –15.57 | 15.89 | 154.12 | –25.50 |
| | | 60 | 93.11 | 2.91 | –14.72 | 15.00 | 150.23 | –23.94 |
| RP | standard | | 91.49 | –0.08 | –1.43 | 1.43 | 86.24 | –0.85 |
| | exposed samples | 35 | 89.53 | –0.44 | 6.79 | 6.81 | 42.85 | 12.27 |
| | | 60 | 88.81 | 0.31 | 6.41 | 6.42 | 42.94 | 11.77 |
| | non-exposed samples | 35 | 91.60 | –0.15 | –0.96 | 0.97 | 84.25 | –0.07 |
| | | 60 | 91.64 | –0.20 | –0.56 | 0.60 | 82.48 | 0.57 |
| SP | standard | | 95.84 | –0.16 | 4.98 | 4.98 | 67.17 | 9.08 |
| | exposed samples | 35 | 96.30 | –0.07 | 2.78 | 2.78 | 78.33 | 5.82 |
| | | 60 | 95.69 | –0.18 | 2.62 | 2.63 | 77.52 | 5.59 |
| | non-exposed samples | 35 | 95.95 | –0.18 | 4.89 | 4.90 | 67.79 | 8.95 |
| | | 60 | 95.62 | –0.16 | 5.48 | 5.48 | 64.35 | 9.82 |
| PP | standard | | 97.79 | –0.19 | 2.83 | 2.84 | 81.85 | 5.83 |
| | exposed samples | 35 | 97.77 | –0.13 | 2.07 | 2.07 | 85.19 | 4.72 |
| | | 60 | 97.65 | –0.05 | 2.07 | 2.07 | 84.81 | 4.73 |
| | non-exposed samples | 35 | 97.64 | –0.31 | 2.75 | 2.77 | 81.76 | 5.72 |
| | | 60 | 97.55 | –0.31 | 3.03 | 3.05 | 80.30 | 6.13 |

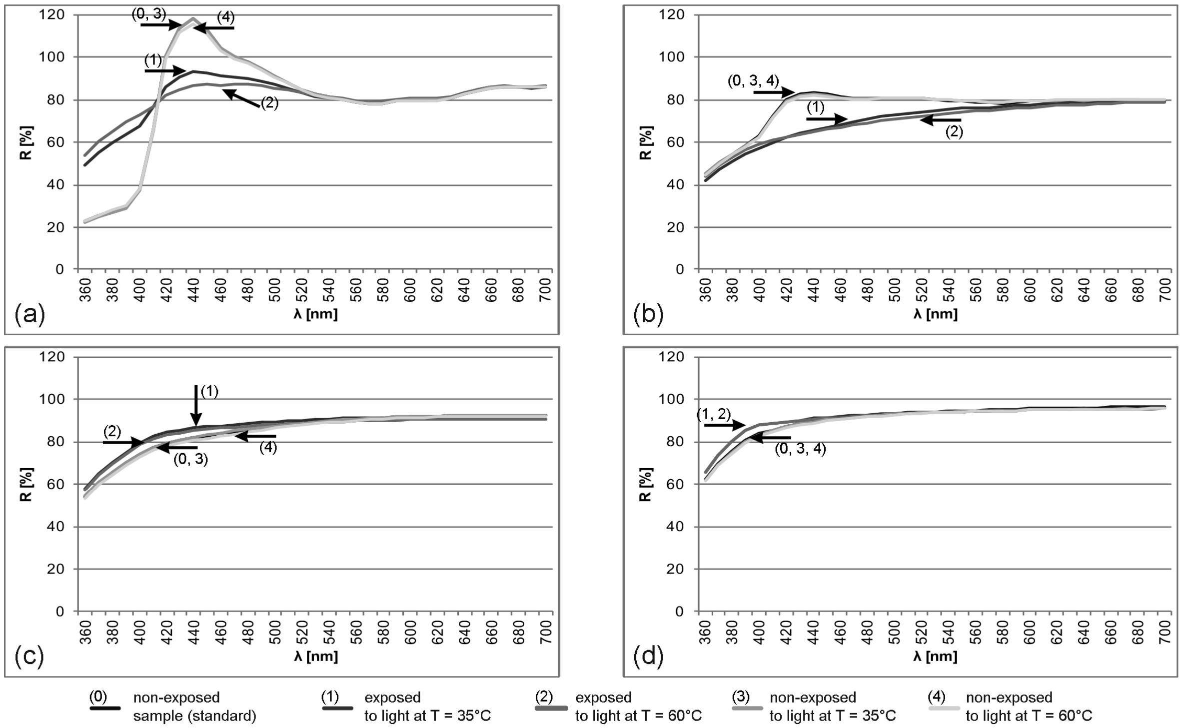


Figure 1: Reflectance of non-exposed and exposed papers after 144 hours of exposure to light at temperature T = 35 °C or T = 60 °C; (a) office; (b) recycled; (c) synthetic; (d) permanent paper

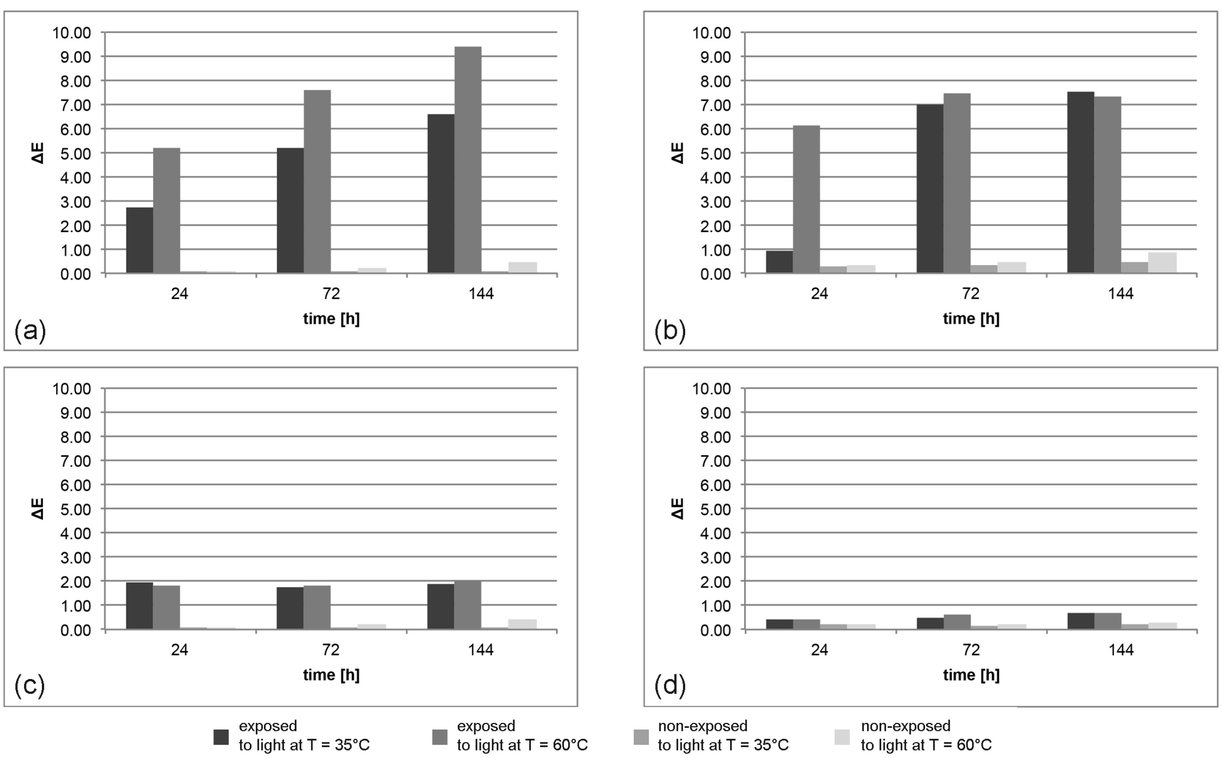


Figure 2: Colour difference (ΔE) of (a) office; (b) recycled; (c) synthetic; (d) permanent paper

The characteristic behaviour of the paper samples is strongly connected to their composition. Optical brightening agents (OBAs) are often added to the office papers to achieve demanded high whiteness. Evidently, the presence of OBAs in OP (Fig. 1) affects its resistance to light. It is very likely that OBA was disintegrated during illumination.[5] The temperature alone has no influence on the reflectance curves and consequently on the colour of office papers. From Table 1, it is evident that the changes in chroma occurred mostly due to the influence of light, as temperature alone did not have such an effect on the colour of recycled paper. According to the manufacturer, the recycled paper was made of 100% recycled fibres and did not contain any OBAs. It is known that a reduction in paper stability is caused by many factors and another endogenous factor can be the presence of lignin in paper.[32–33] The most undesirable feature of lignin is its effect on brightness properties, which is shown as discoloration after exposure to light. However, the lignin-free papers lost their optical properties more slowly.[32] Synthetic and permanent papers were designed for special purposes and they indeed exhibited better resistance to light. Paper SP contains cellulosic as well as synthetic fibres (polyamide (PA) and polyester (PES)). PES fibres have better light resistance than PA and cellulosic fibres, the destructive reactions would take place at higher temperature.[14,34] Discoloration of paper SP occurred after exposure to light and higher temperature. Permanent paper PP was made according to the standards for permanence and durability that require paper to be alkaline, to contain an alkaline reserve, such as $CaCO_3$, and to have no more than 1% lignin content.[33]

The comparison of colour differences that occurred on the samples, which were kept in the dark under conditions of constant temperature and relative humidity (RH = 35%), shows that even at higher temperature (T = 60 °C) the value $\Delta E$ does not exceed 1 unit, regardless of the type of paper, so it is very likely that in the absence of light, a moderate elevation of temperature alone would not cause visible changes on paper. In the dark, however, the recycled paper (RP) was the one that was the most affected by the influence of higher temperature.

Figure 3 shows CIE whiteness, W, which was calculated from the reflectance spectra (Fig. 1) of the paper samples before and after exposure to light.

Among the paper samples presented, the highest whiteness was observed for OP (W = 153.63), which is the consequence of added OBAs. During the exposure to light, the whiteness of OP gradually decreases. The decrease is more evident at higher temperature (T = 60 °C) and after 144 hours of illumination its whiteness is only W = 96.21. The whiteness of recycled paper (RP) decreases substantially after 144 hours of exposure to light from the starting value W = 86.24 to W = 42.85 (T = 35 °C) or W = 42.94 (T = 60 °C). The changes occur mostly within the first 24 or 72 hours; afterwards the whiteness remains the same. Completely different characteristics can be observed on the synthetic paper (SP), as after 24 hours of exposure its whiteness slightly increases (from W = 67.17 to W = 78.33 (T = 35 °C) or W = 77.52 (T = 60 °C)). Such behaviour is probably the consequence of partial degradation of some of its coloured components.[1] This effect does not enhance with time, however. A similar but not as obvious effect can be detected on the permanent paper (PP). There is no significant decrease of its whiteness after exposure to light, although the starting value was not very high (W = 81.85). If we compare the whiteness of samples, which were kept in the dark, we can conclude that the effect of moderately increased temperature is only minor. Some changes in whiteness can be observed on OP and RP at T = 60 °C as the values decrease by 3.4 units. At T = 35 °C, the whiteness of PP and SP remains practically the same after 144 hours. Ideally, white paper would exhibit very high lightness and very low chromaticity, therefore the decrease in whiteness can be the consequence of decreased lightness or increased chromaticity. According to Figure 3, the value of lightness, $L^*$, for the majority of the samples, changes very little. The only exception is RP, where the decrease of lightness can be observed, especially at higher temperature ($\Delta L^* = -2.68$). Evidently, the paper becomes a little darker. A similar but less obvious effect can be observed for OP ($\Delta L^* = -0.79$).

The decrease in whiteness is very often the consequence of increased yellowness, which can occur due to different degradation processes in paper.[33] The process of yellowing under the influence of light can be evaluated on the basis of yellowing index, YI, or merely by CIELAB coordinate $b^*$ (Fig. 4). According to the results, the calculated values of YI and $b^*$ for OP were negative, meaning that before exposure to light this paper was slightly bluish; this is also evident from its spectral curve (Fig. 1). During the exposure to light, OP became less bluish and the yellowing index increased, but its whiteness remained relatively high (W = 111.56 (T = 35 °C), W = 96.21 (T = 60 °C)), compared to the other three paper grades. The recycled paper (RP) also exhibited an increase in yellowness during the exposure to light. Compared to OP, this paper is more yellowish, but the values of YI and $b^*$ do not increase with time after 72 hours. Also, the effect of slightly higher temperature is less pronounced than on OP. According to the results, neither synthetic nor permanent paper exhibits further yellowing, as the calculated values YI and $b^*$ slightly decrease during the exposure to light.

However, those two papers were yellowish already before exposure. In the dark, all of the four selected paper grades exhibited slight yellowing at higher temperature (T = 60 °C) after longer time of exposure.Specular gloss is one of the optical properties of paper that significantly influence its appearance. Higher specular gloss is connected to a more oriented reflection of light from a smoother surface. External factors, such as heat, humidity and light, can change the surface properties of paper. According to the results (Fig. 5), the smallest changes were observed on the office paper (OP), which had the lowest specular gloss before the exposure. Paper RP exhibited the highest specular gloss at the beginning, which can be ascribed to the presence of a special coating. After exposure to light, however, its specular gloss was significantly lower, especially on the sample that was treated at higher temperature (T = 60 °C). In the dark, the decrease in specular gloss was not so evident.

The light affected also the specular gloss of the permanent paper (PP) and under the influence of both light and temperature the specular gloss of PP almost halved. Because photochemical damage is largely a surface phenomenon, and it depends on the wavelength of radiation as well as on the diffusion of oxygen into the material, the changes of specular gloss considerably differ among samples.[12]

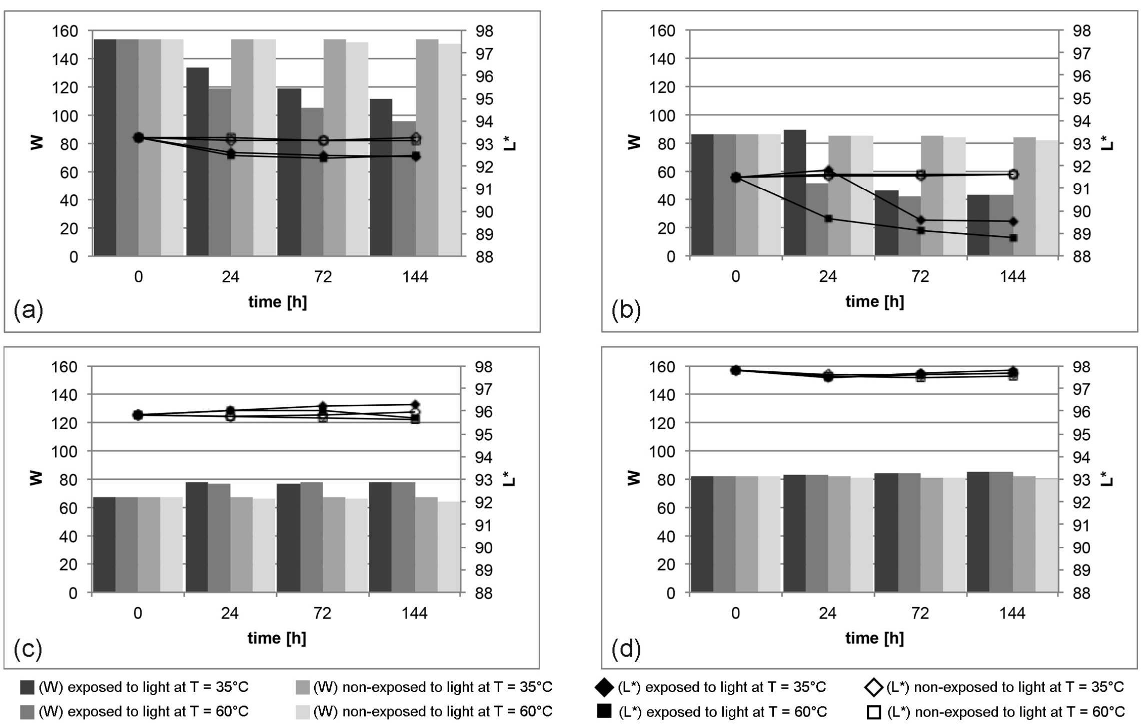


Figure 3: CIE whiteness (W) and lightness (L*) of (a) office; (b) recycled; (c) synthetic; (d) permanent paper; before and after 24, 72, 144 hours of exposure to light at temperatures of 35 °C or 60 °C

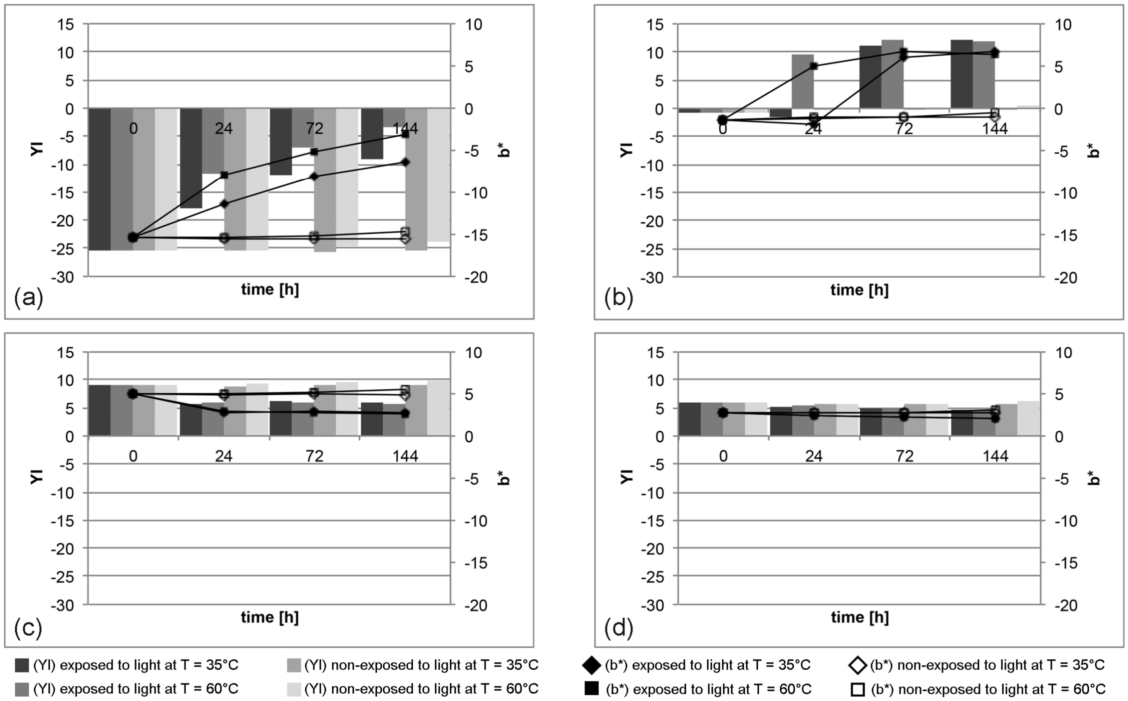


Figure 4: Yellowing index (YI) and CIELAB coordinate (b*) of (a) office; (b) recycled; (c) synthetic; (d) permanent paper; before and after 24, 72, 144 hours of exposure to light at temperatures of 35 °C or 60 °C

Changes of gloss can be connected to changes of the surface structure. Table 2 shows that temperature has a considerable influence on the roughness of papers. The roughness (Ra) of papers increases as a function of the time of illumination. At higher temperature, slightly greater changes in roughness of papers are observed. The amount of coating influences the roughness properties of papers,[4] therefore the roughness of uncoated office paper (OP) is higher in comparison with that of coated recycled paper (RP). The roughness of paper RP is generally lower because of the coated surface of the paper, as can be also seen from the SEM photos (Fig. 6). The roughness of synthetic paper (SP) significantly increased in time. As it was expected, higher temperature in combination with light had an even greater effect (Fig. 6). For paper SP, standard deviation (σ) of roughness measurements was very high, which indicates a high influence of light and temperature, as well as the inhomogeneity of the paper surface. However, light and temperature had almost no effect on the roughness of permanent paper (PP).

The surface of the four papers before and after exposure is shown on SEM images (Fig. 6). Different coating components are used in order to improve paper printability and optical properties. The amount of coating influences the roughness/smoothness of the surface and, in addition, it has a significant impact on important properties of paper and print, such as gamut, sharpness, water-fastness.[35–36] According to the SEM images in Figure 6, papers OP and PP were uncoated and after 144 hours of illumination, no changes were detected on their surface. However, changes appeared on coated papers RP and SP. With an increase of temperature, the most obvious changes occurred on the surface of paper SP. We can assume that under the influence of light, especially at moderately elevated temperature, the smoothness of papers can be decreased. This effect is more pronounced on the papers that originally exhibit higher smoothness or specular gloss.

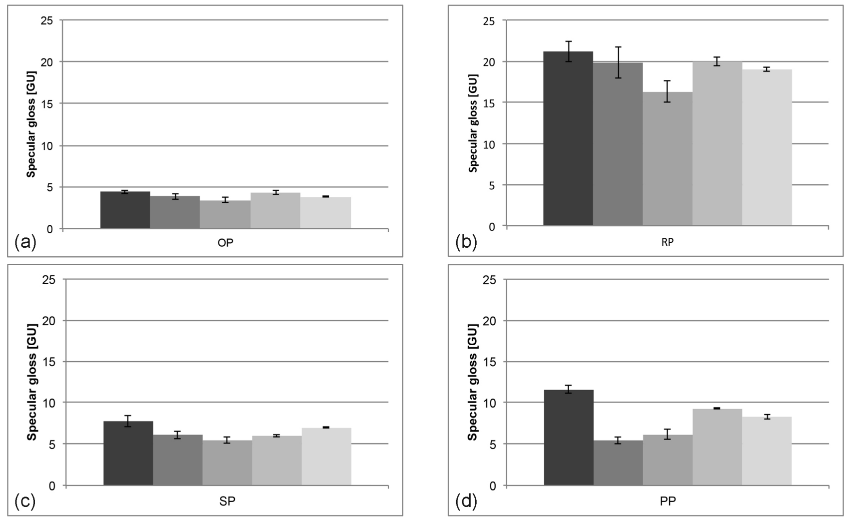


Figure 5: Specular gloss of non-exposed and exposed papers after 144 hours of exposure to light at temperatures of 35 °C or 60 °C; (a) office; (b) recycled; (c) synthetic; (d) permanent paper

Table 2
Roughness measurements (Ra) with standard deviation (σ) for exposed and non-exposed papers before (standard) and after 24, 72 and 144 hours of exposure to light at temperatures of 35 °C and 60 °C

| T (°C) | | | OP | | | RP | | | SP | | | PP | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Hours | | | 24 | 72 | 144 | 24 | 72 | 144 | 24 | 72 | 144 | 24 | 72 | 144 |
| Standard | | Ra (µm) | | 2.91 | | | 1.60 | | | 4.72 | | | 3.06 | |
| | | σ | | 0.17 | | | 0.26 | | | 0.77 | | | 0.29 | |
| Exposed samples | 35 | Ra (µm) | 3.06 | 3.11 | 3.13 | 1.74 | 1.76 | 1.75 | 5.38 | 5.39 | 5.44 | 3.07 | 3.08 | 3.23 |
| | | σ | 0.27 | 0.08 | 0.37 | 0.24 | 0.17 | 0.18 | 0.40 | 0.56 | 0.31 | 0.21 | 0.16 | 0.27 |
| | 60 | Ra (µm) | 3.05 | 3.26 | 3.26 | 1.81 | 1.85 | 1.92 | 5.44 | 5.51 | 5.63 | 3.11 | 3.12 | 2.99 |
| | | σ | 0.17 | 0.24 | 0.28 | 0.30 | 0.12 | 0.26 | 0.68 | 0.83 | 0.45 | 0.29 | 0.29 | 0.18 |
| Non-exposed samples | 35 | Ra (µm) | 3.03 | 3.06 | 3.16 | 1.71 | 1.74 | 1.74 | 5.17 | 5.22 | 5.21 | 2.57 | 2.49 | 2.79 |
| | | σ | 0.30 | 0.42 | 0.42 | 0.17 | 0.19 | 0.20 | 0.87 | 0.71 | 0.56 | 0.18 | 0.23 | 0.21 |
| | 60 | Ra (µm) | 3.05 | 3.20 | 3.21 | 1.71 | 1.71 | 1.80 | 4.81 | 5.02 | 5.11 | 2.95 | 2.68 | 2.81 |
| | | σ | 0.41 | 0.25 | 0.25 | 0.20 | 0.20 | 0.24 | 0.51 | 0.56 | 0.46 | 0.06 | 0.14 | 0.27 |

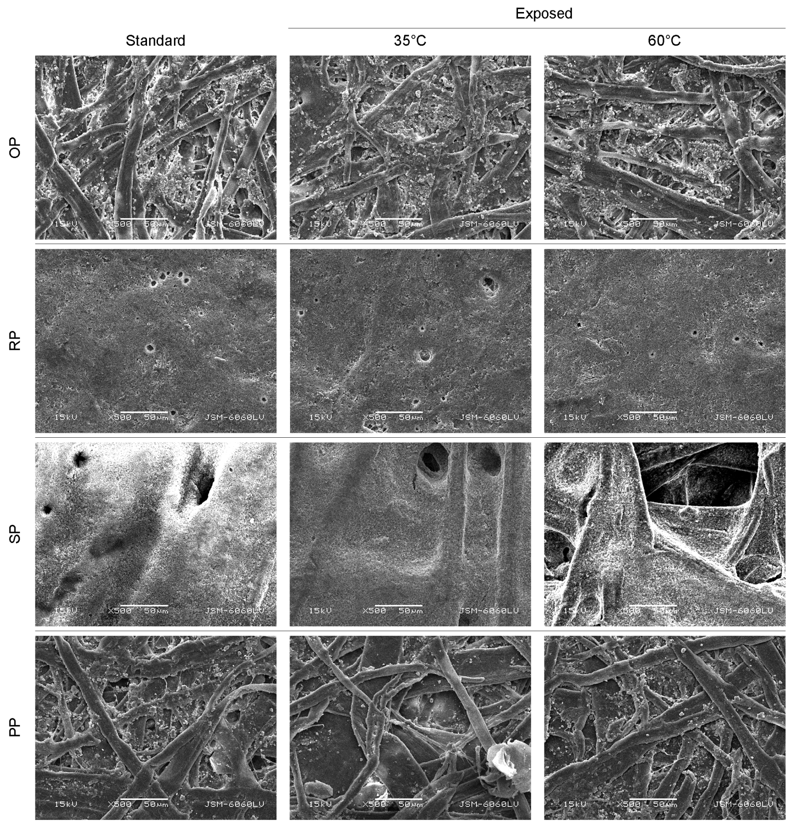


Figure 6: SEM images of papers at 500× magnification before (standard) and after 144 hours of illumination with xenon light at a temperature of 35 °C and 60 °C

## CONCLUSION

In the present research, the influence of temperature and light on optical properties of four selected papers was studied under conditions similar to those of real use and storage of documents. According to the results, changes of optical properties on papers are induced predominantly by light. Nevertheless, the influence of temperature is not negligible as the differences in the optical properties of the papers became more evident already at moderately increased temperature.

Results have shown that light has an extremely negative influence on the stability of papers that contain cellulosic fibres in combination with optical brightening agents (OBA) or recycled fibres. It can be concluded that the presence of OBA strongly aggravates the stability of paper and can substantially reduce the durability of documents. According to the results, recycled papers are not suitable for documents intended for a longer storage period. The influence of moderately elevated temperature in the absence of light is less destructive and the colour differences on the papers examined were mostly not noticeable. Among the papers tested, the synthetic paper clearly stands out, as its optical properties were very good, but under the influence of light and higher temperature, its surface properties became significantly poorer. As expected, the permanent paper achieved better results in comparison with office paper. Its whiteness and colour, as well as other optical properties, only slightly changed even after exposure to light at 60 °C. The calculated colour differences were in accordance with the changes in whiteness and yellowness of the papers.

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