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Ayhan GENÇER, Ahmet CAN, Rıfat KURT, Cengiz KEŞMER

NON-DESTRUCTIVE TESTING USING UV-INDUCED COLOUR CHANGES TO PREDICT MECHANICAL PROPERTIES OF KRAFT PACKAGING PAPER

This study measured the decrease in some mechanical properties of kraft paper used in packaging and bag production by examining the colour changes occurring in accordingly prepared standard samples subjected to UV influence. Thus, it is aimed to predict the mechanical properties of packages that have been exposed to sunlight over long periods of time with the colour change measurement, which is a non-destructive test, without the packaging deformation. Since there are mechanical differences between the cross-machine width and machine length of fabricated papers, these were measured separately. Measurements were made on papers exposed to UV for 0, 120, 240, 360, 480, 600, and 720 h. Tensile strength, strain at break, and tensile energy absorption (TEA) values were measured and also predicted for the kraft paper. For the prediction, the time and colour change values were used as independent variables, whereas the mechanical properties were taken as the dependent variables. The relationship between the time and colour change and the tensile strength, strain at break, and TEA variables in both the cross-machine and the machine directions were significant at the 5% level, and the R^2 values were within acceptable limits. The results showed that the mechanical properties of the papers under UV effect decreased proportionally with the increase of the UV time.

Keywords: packaging paper, UV effect, colour change, mechanical properties, regression analysis

Ayhan GENÇER✉ (*agencer@bartin.edu.tr*), https://orcid.org/0000-0002-0758-5131, Ahmet CAN (*acan@bartin.edu.tr),* https://orcid.org/0000-0001-5926-6039, Rıfat KURT (*rkurt@bartin.edu.tr*), https://orcid.org/0000-0002-7136-7665, Department of Forest Industrial Engineering, Faculty of Forestry, Bartın University Bartın, Turkey; Cengiz KEŞMER (*ckesmer@cu.edu.tr*), Aladağ Vocational High School Forestry Department, Çukurova University, Adana, Turkey

Introduction

Depending on the marketing network of the retailing company, packaging and bag paper used in product packing can be subjected to diverse climates during distribution to different geographical regions. As a result of exposure to external influences such as temperature, relative humidity, rain, light, and friction in these environments, changes may occur in the physical, chemical, and mechanical properties of the paper in direct proportion to the degree and duration of the effect. Lignin content in pulp has no significant impact on the aging rate of paper [Małachowska et al. 2020]. Humidity of the air during the aging of the paper significantly changes the chain length of the cellulose [Małachowska et al. 2021]. One of the most important among these effects is UV radiation, which is often difficult to avoid. Therefore, in this study, we tried to measure the deformation of mechanical properties with UV.

Light transfers energy and thus, when sunlight hits paper, it activates the free electrons in the paper and raises them to a higher level, and when the light effect decreases, these electrons tend to return to their orbitals. When the oxidation energies do not allow rotation, this indicates that a change has occurred [Łojewski et al. 2010]. This can be a colour change or a change in the chemical, physical, or mechanical sense, either alone or in combination. The mechanical properties of the paper may be reduced as a result. In this case, if the paper cannot fulfil the mechanical strength requirements at the place of use, this signifies that its packaging feature has been lost, which is an undesirable situation in terms of the safety of the product inside.

Paper products obtained from different wood species exhibit different photochemical behaviour because of the differences in their wood structure. Because coniferous and broad-leaved tree woods have different lignin structures, their photochemical reactions to visible and invisible rays may cause brightness/darkness differentiation in their natural colours [Şahin 2002]. Thus, depending on the type of lignocellulosic raw material used in the production of the paper, the colour change may also differ under the influence of UV, which might indicate decrease in the mechanical strength properties of the paper as a result of the decomposition of its structure.

Kraft paper is usually sold in rolls and is generally used in various industries for dry product packaging and bagging. For bag manufacture, this paper is semi-finished and sized according to the dimensions of the bag to be produced. These bags are mostly used to package cement, lime, flour, seeds, etc.

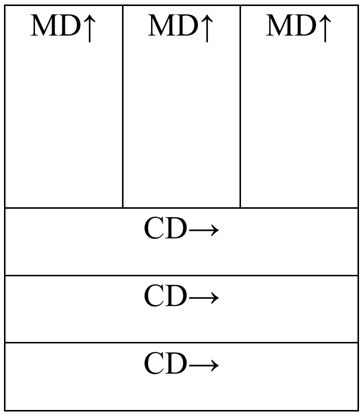
In this study, roll kraft paper used in the production of packaging and paper bags was subjected to 0, 120, 240, 360, 480, 600, and 720 h of UV, and the effects on the paper were measured and also predicted in terms of the machine direction (MD) and cross-machine direction (CD) tensile strength, tensile index, strain at break, total energy absorption (TEA), and tensile stiffness. Measurement of colour change on paper can be achieved non-destructively using existing measuring devices. However, tests for tensile strength, tensile index, strain at break, TEA, and tensile stiffness values are destructive procedures and require separate standard samples to be prepared. Therefore, these measurements cannot be carried out on the packaging. The significance of this study lies in predicting mechanical values by applying non-destructive means to measure the colour changes of bags and packaging paper after long-term exposure to sunlight and in gathering information about the durability of the packaging.

Materials and methods

The kraft paper used in this study was taken from a roll of 80 ±2 g/m^2 weight, containing 80% pine wood pulp and 20% broad-leaved tree wood pulp. The average moisture content of the paper was calculated as 7.20%.

Local weight differences may occur in the paper because of uneven fibre distribution during production. For this reason, grammage (weight) tolerance in the range of 56-125 g/m^2 (±2.5%) is recognized for paper [Bostancı 1984]. The weight tolerance of the paper used in the study was calculated as ±1.70%.

The mechanical properties of paper differ in production in terms of machine (longitudinal) and cross-machine (transverse) directions, e.g., the shear strength is always higher in the machine direction than in the cross-machine direction [Casey 1980]. This is because when the pulp coming from the headbox flows over the sieve, the fibres are directed to the machine in the longitudinal direction. When taking a sample from a roll, the first three layers of the roll should be separated from the intact top layer and the sample should be taken from the fourth layer [Bostancı 1984]. The breaking strength changes from the edge to the middle due to different stresses in the cross-machine direction of the roll. Therefore, in order to determine the actual breaking strength difference between the cross-machine (transverse) direction and the machine (longitudinal) direction, the sampling was taken, according to the TAPPI T 400 sp-02 [2002] standard, from the outer third layer of the roll and from precisely the middle on the same line of the cross-machine direction, as shown in Fig. 1. In this part of the publication the research subject matter is described in a concise way.



Machine Direction (length): MD↑
Cross-machine Direction (width): CD→
Fig. 1. Directions of samples taken from the paper roll

Paper samples cut for breaking strength were kept for 24 h in an air-conditioned laboratory at 23 ±2 °C and 50 ±2% relative humidity according to TAPPI T 402 [2003] standard. Tensile strength, tensile index, strain at break, TEA, and tensile stiffness measurements were performed on the same device according to TAPPI T 494 om-01 [2001] standard.

The accelerated weathering test was carried out in a Q-Lab testing chamber in the laboratory of the Forest Industrial Engineering Department of Bartin University in Turkey. Test parameters included a UV wavelength of 340 nm at 0.65 W/m^2 for 24, 48, 96, 120, 240, 360, 480, 600, and 720 h. The reason for choosing these parameters was determined according to the results of our previous studies. In previous studies, it was concluded that a maximum of 150 hours of weathering was sufficient for paper samples. It is also known that the weathering process takes place intensively in the first 72 hours [Gencer et al. 2016; Gencer et al. 2019]. Colour measurement of the test samples was carried out before and after the accelerated weathering with ISO 7724-3 standard using Konica Minolta CD-600 colour meter (Osaka, Japan). For three replicates in each variation, the colour measurements were measured from three different points on the paper samples, and their mean value was calculated [ISO 7724-3:2021].

The Commission Interationale de l'Éclairage (CIELab) system consists of three variants (ISO 7724): L* refers to light stability and a* and b* to chromatographic coordinates (+a* indicates red, -a* green, +b* yellow, -b* blue). The values of L*, a*, and b* were measured on the samples and the colour changes were determined according to the following formulae:

$$\Delta L^* = Lf^* - Li^* \quad (1)$$
$$\Delta a^* = af^* - ai^* \quad (2)$$
$$\Delta b^* = bf^* - bi^* \quad (3)$$
$$\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{½} \quad (4)$$

Results and discussion

Mechanical Properties

In paper production, many variables such as the orientation of the fibres, the degree of wet pressing, and the stresses upon drying affect the ultimate tensile strength and elongation properties of the paper. Fibre orientation and drying stresses affect the amount of elongation in the machine and cross-machine directions. In the machine direction, the strength of the paper is greater and the elongation is less than in the cross-machine direction [Eroğlu 2003]. The reason may be that the fibre attachment is lateral in the cross-machine direction and the microfibrils open like scissors before breaking during loading. The elongation determines the amount of energy absorbed to tear the paper. It is more important than tensile strength in many applications because as the energy absorption increases, the strength of the paper also increases. Elongation is 10% in bag paper but can be increased by creping [Bostancı 1984]. In this study, the elongation in the cross-machine direction was higher than in the machine direction. The mechanical properties of the MD and CD samples declined as the UV time was increased. Therefore, the packaging properties would certainly be weakened. In terms of reducing possible risks, it is important to use colour change to monitor this situation.

Breaking strength is important for cement bags and packaging paper [Eroğlu 1990]. Long-term exposure of paper-packaged products to sunlight causes significant reductions in breaking strength. The values of the breaking strength samples taken as shown in Figure 1 are given in Table 1 for machine direction (MD) and cross-machine direction (CD), revealing a robust difference between them.

Table 1. Values for cross-machine direction (CD) and machine direction (MD)

Duration (h)	Tensile Strength (kNm)	Tensile Index (Nm g^{-1})	Strain at Break (%)	TEA (J m^{-2})	Tensile Stiffness (kN m^{-1})
Cross-machine Direction					
0	4.56	55.93	11.20	344	367
120	4.45	55.64	8.98	287	344
240	3.99	49.09	7.50	222	340
360	3.86	47.83	7.04	206	331
480	3.68	44.01	6.41	174-	320
600	3.56	45.90	5.80	161	316
720	3.46	43.38	4.96	155	314
Machine Direction					
0	5.56	55.64	7.16	268	427
120	5.36	55.00	7.00	250	427
240	5.25	55.00	6.52	240	424
360	4.80	56.18	5.43	189	425
480	4.57	52.36	5.32	181	419
600	4.39	55.62	5.11	177	416
720	4.47	50.25	4.98	164	418

The colour change values in the test and control samples were calculated using the ⊿L* (light intensity), ⊿a* and ⊿b* chromatographic coordinates (+a red, -a green, +b yellow, -b blue) determined according to the CIELab system. Figure 2 shows the ΔL*, Δa*, Δb*, and total colour change of the samples.

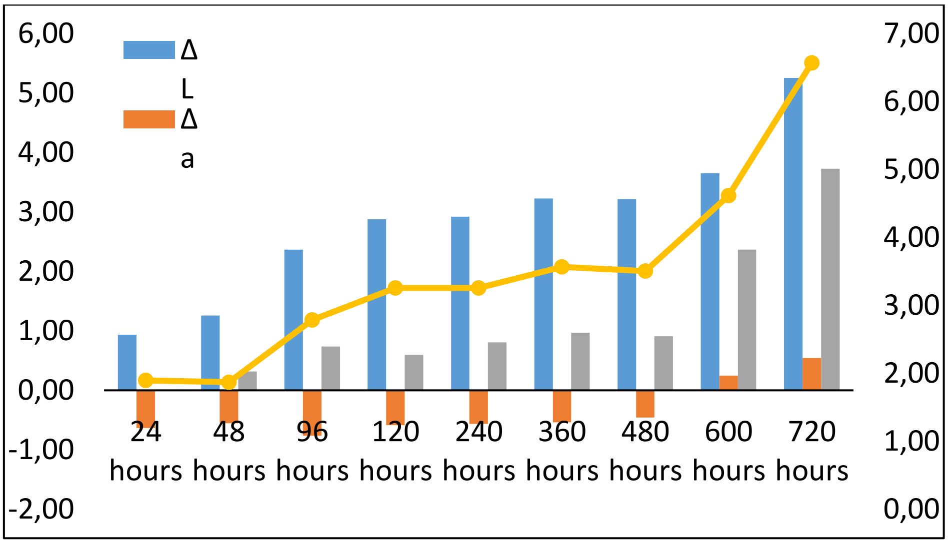


Fig. 2. **ΔL*, Δa*, Δb* and total colour changes of samples**

According to the results, the ΔL^*, Δa^*, and Δb^* values increased in parallel with the increase in time. The colour stability was preserved for the first 480 h, but the total colour change increased significantly with increasing time. The increase in the ΔL^* value in the paper samples indicated that the whiteness of the sample surfaces had increased. When the Δa values (Fig. 2), which represent the red-green colour coordinates of the samples, were examined, they were negative (green) for the first weathering time. As the weathering time increased the Δa^* value tended to be positive. The Δb value (Fig. 2) represents the yellow-blue colour coordinates of the samples. Because the lignin content in the unbleached pulp was high, the weathering effect led to oxidation in the lignin and the yellow colour tendency increased. There was only a minute amount of lignin in the bleached pulp and because of this, there was no tendency to become yellow. As the bleaching substances are blue, the tendency towards blue is increased by oxidation.

Statistical analysis and prediction

The tensile strength, strain at break, and TEA values of the kraft paper used in packaging and bag paper production were also predicted in the study. For the prediction, the time and colour change values were used as independent variables, whereas the mechanical properties were taken as the dependent variables. The correlation (R) and determination (R^2) values and the analysis of variance (ANOVA) results of the established models given in Table 2 show that the relationship between the time and colour change and the tensile strength, strain at break, and TEA variables in both the cross-machine and the machine directions were significant at the 5% level, and the R^2 values were within acceptable limits.

Table 2. ANOVA results for the models

Mechanical Properties	Sum Squares (SS)	Mean Square (MS)	F Ratio	P-Value	R (%)	R-sq (%)
Cross-machine Direction (CD)						
Tensile Strength	22.677	11.338	49.888	0.000	78.3	61.3
Strain at Break	179.330	89.665	75.218	0.000	84.0	70.5
TEA	301726.121	150863.061	121.443	0.000	89.1	79.4
Machine Direction (MD)						
Tensile Strength	12.963	6.481	93.439	0.000	86.5	74.8
Strain at Break	49.160	24.580	90.443	0.000	86.1	74.2
TEA	103033.503	51516.751	73.286	0.000	83.6	69.9

After determining that the results were statistically significant and the models valid, the regression equations given in Table 3 for tensile strength, strain at break, and TEA variables were established and the change in mechanical properties was predicted at 600 to 720 h. The negative effects of both the time and colour change variables on all mechanical properties are clearly seen in the established equations.

Table 3. Regression prediction models

Cross-machine Direction	Machine Direction
Regression Model	Regression Model
$Y_{Tensile}=5.140-0.038_{colourchange}-0.002_{Time}$	$Y_{Tensile}=5.815-0.146_{colourchange}-0.001_{Time}$
$Y_{Strainbreak}=10.987-0.655_{colourchange}-0.003_{Time}$	$Y_{Strainbreak}=7.777-0.343_{colourchange}-0.002_{Time}$
$Y_{TEA}=361.987-17.804_{colourchange}-0.188_{Time}$	$Y_{TEA}=293.575-14.114_{colourchange}-0.085_{Time}$

The predicted and the measured values regarding the mechanical properties between 600 and 720 h are given in Table 4. The results showed that in both the predicted and the measured values, the decrease in the mechanical properties continued with time. In order to measure the performance of the prediction process, mean squared error (MSE) and mean absolute percentage error (MAPE) values of the variables were calculated (Table 5).

The error performance values in Table 5 demonstrate that the tensile strength (with very low measured error values) and the predicted values were very close to each other. In the literature, it is stated that models with MAPE values below 10% are classified as “very good” models [Lewis 1982; Witt and Witt 1992; Çuhadar et al. 2009]. However, the strain at break and TEA prediction results were also at acceptable levels. Similarly, models with MAPE values between 10% and 20% are classified as “good” [Lewis 1982; Witt and Witt 1992; Çuhadar et al. 2009].

Figure 3 was created in order to demonstrate the predictions made with the values measured for tensile strength, strain at break, and TEA values comparatively and to provide clearer visual information. The success of the prediction for tensile strength values is also clearly seen in Figure 3. Again, in the predictions of strain at break and TEA values, the results obtained in some time periods were very close to the real values. This is an indication that the established models could be used successfully to predict the stated mechanical properties.

Table 4. Predicted and measured values

Time	Cross-machine Direction						Machine Direction					
	Tensile strength		Strain at break		TEA		Tensile strength		Strain at break		TEA	
	M	P	M	P	M	P	M	P	M	P	M	P
600	3.57	3.76	5.86	6.07	149	164	4.48	4.52	5.24	4.94	167	175
600	3.69	3.79	6.69	6.60	178	179	4.79	4.64	5.70	5.22	186	187
600	3.33	3.72	4.35	5.37	112	146	3.93	4.37	5.05	4.58	104	160
600	3.56	3.75	4.8	5.91	134	160	4.38	4.49	5.20	4.86	166	172
600	3.33	3.72	4.72	5.41	130	146	4.31	4.37	5.05	4.60	166	161
600	3.61	3.78	5.95	6.47	164	175	4.49	4.61	5.35	5.16	174	184
600	3.65	3.78	6.51	6.50	171	176	4.51	4.62	5.41	5.17	182	185
600	3.97	3.81	7.04	6.97	197	189	4.88	4.72	6.17	5.42	189	195
720	3.76	3.49	6.09	5.13	154	126	4.57	4.27	5.67	4.40	189	153
720	3.59	3.48	5.83	5.08	148	125	4.53	4.26	5.13	4.38	187	152
720	3.29	3.41	5.08	3.83	140	91	4.15	3.98	4.42	3.72	157	125
720	3.4	3.47	5.36	4.82	148	118	4.43	4.20	4.96	4.24	159	146
720	3.32	3.43	5.25	4.11	144	98	4.27	4.04	4.40	3.87	157	131
720	3.31	3.40	4.75	3.72	138	88	4.14	3.96	4.13	3.66	136	122
720	3.52	3.48	5.37	4.97	148	122	4.48	4.23	5.08	4.32	187	149
720	3.34	3.45	5.3	4.54	146	110	4.4	4.14	4.92	4.09	159	140

*M: Measured, P: Predicted

Table 5. Error performance values

Time	Cross-machine Direction						Machine Direction					
	Tensile Strength		Strain at Break		TEA		Tensile Strength		Strain at Break		TEA	
	MAPE	MSE	MAPE	MSE	MAPE	MSE	MAPE	MSE	MAPE	MSE	MAPE	MSE
600	5.29	0.04	3.51	0.04	10.29	235.21	0.87	0.00	5.68	0.09	4.98	69.06
600	2.71	0.01	1.29	0.01	0.55	0.94	3.15	0.02	8.35	0.23	0.49	0.83
600	11.68	0.15	23.54	1.05	29.95	1125.01	11.07	0.19	9.30	0.22	54.24	3182.14
600	5.34	0.04	23.21	1.24	19.57	687.75	2.41	0.01	6.48	0.11	3.65	36.61
600	11.74	0.15	14.58	0.47	12.67	271.47	1.46	0.00	8.95	0.20	2.92	23.51
600	4.78	0.03	8.78	0.27	6.96	130.11	2.67	0.01	3.63	0.04	5.80	101.73
600	3.67	0.02	0.17	0.00	2.99	26.18	2.35	0.01	4.45	0.06	1.46	7.02
600	4.00	0.03	1.02	0.01	4.12	66.01	3.27	0.03	12.24	0.57	3.05	33.22
720	7.31	0.08	15.84	0.93	18.18	783.65	6.57	0.09	22.42	1.62	19.25	1324.34
720	2.98	0.01	12.79	0.56	15.61	533.72	5.94	0.07	14.68	0.57	18.86	1243.99
720	3.66	0.01	24.51	1.55	35.05	2408.14	4.04	0.03	15.78	0.49	20.51	1036.84
720	1.99	0.00	10.07	0.29	20.46	916.92	5.15	0.05	14.54	0.52	8.15	167.98
720	3.21	0.01	21.68	1.30	31.63	2073.92	5.29	0.05	12.10	0.28	16.71	687.95
720	2.83	0.01	21.75	1.07	36.43	2527.66	4.45	0.03	11.36	0.22	10.10	188.77
720	1.25	0.00	7.51	0.16	17.77	691.84	5.48	0.06	15.05	0.58	20.22	1429.40
720	3.33	0.01	14.41	0.58	24.66	1295.79	5.94	0.07	16.87	0.69	12.00	363.97
Mean	***4.73***	***0.04***	***12.79***	***0.60***	***17.93***	***860.89***	***4.38***	***0.05***	***11.37***	***0.41***	***12.65***	***618.58***

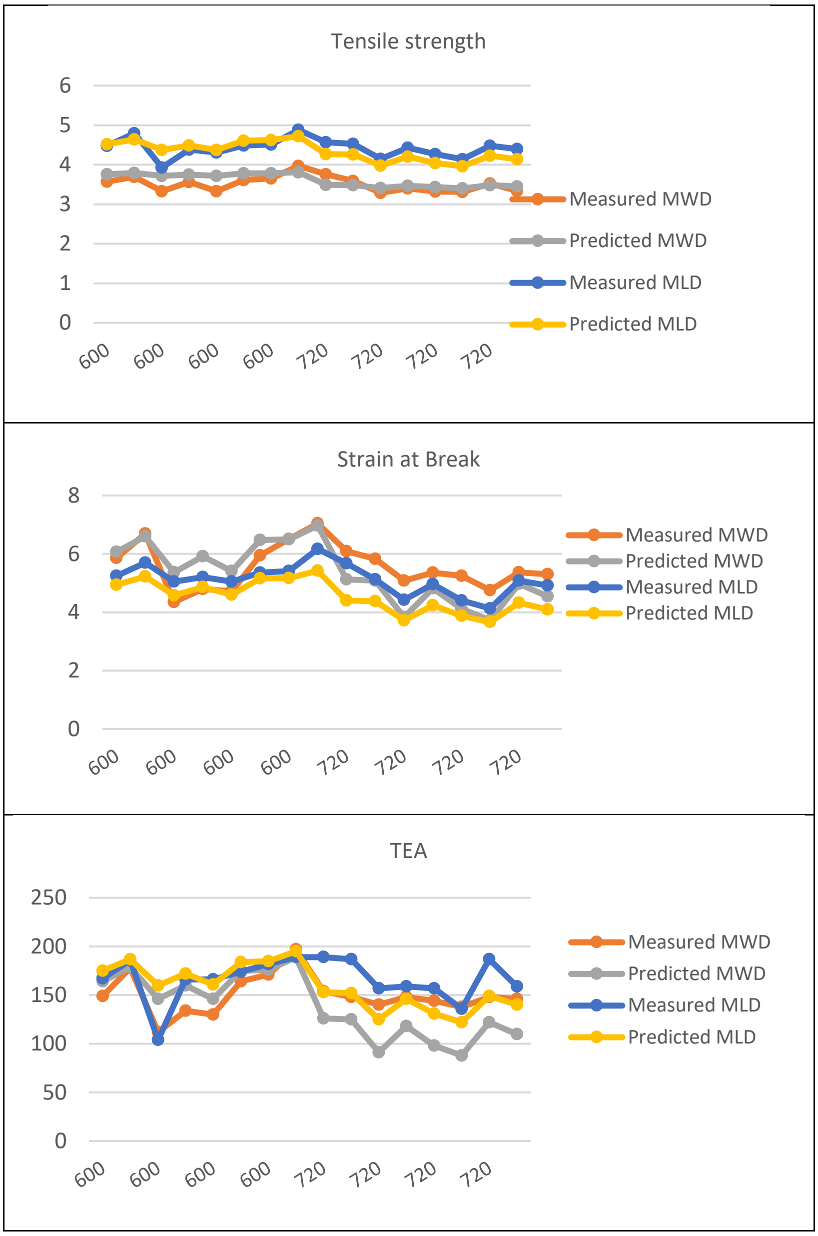


Fig. 3. Predicted and measured tensile strength values

Conclusions

In the study, tensile strength, tensile index, strain at break, TEA, and tensile stiffness values of kraft paper used in packaging and bag production were measured. In addition, prediction of these data were carried out. For the prediction, the time and colour change values were used as independent variables, whereas the mechanical properties were taken as the dependent variables.

The correlation (R) and determination (R^2) values and ANOVA results of the established models demonstrated that the relationship between the time and colour change and the tensile strength, strain at break, and TEA variables in both the cross-machine and machine directions was significant at the 5% level and that the R^2 values were within acceptable limits. As a result, the tensile strength (with error values measured very low) and the predicted values were very close to each other. Because the MAPE values were below 10%, they could be classified as “very good” models. As a result of measurements made for strain at break and TEA, the MAPE values were between 10% and 20%, and therefore could be classified as “good”. As a result of this study, the mechanical reduction was found to be proportional to the increase of UV time and exposure of the papers to the UV effect. Without damaging the package, measuring the colour change, as a non-destructive method, and predicting the reduction of the mechanical properties can reveal information about the durability of the packaging in which the product is enclosed and about the security of the paper package during transport.

The kraft paper used in this study was obtained from a mixture containing 80% softwood pulp and 20% hardwood pulp. The lignin structures of softwoods and hardwood are different. To examine the effect of photochemical reactions on paper obtained from coniferous and hardwood, the UV effect on paper obtained from the pure pulp was also examined separately. In addition, the values were predicted for paper samples exposed to 600-720 h of UV exposure. In future studies, it could prove useful to examine the prediction values for different times of exposure.

In the case of bags produced from 80 g/m^2 kraft paper, we measured the carrying capacity as 25 kg. The minimum tensile strength values required in the market are 5.5 kNm in the machine direction and 4.5 kNm in the cross-machine direction. Because the desired durability properties can change depending on the volume of the paper packaging and the density of the product inside, in this study, no conclusion was presented about packaging safety. This is directly related to the bag volume and the density of the product in it, and therefore, product safety should be calculated separately.

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board, pulp handsheets, and related products
TAPPI T 494 om-01:2001 Tensile Properties of Paper and Paperboard (Using Constant Rate of Elongation Apparatus)

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