




Oil and water resistant of packaging papers

ABSTRACT

The permeability of paper and cardboard materials is extremely important for packaging safety. However, paper and cardboards have poor liquid resistance due to their naturally porous structure and the hydrophilic character of cellulose fibers. When packaging papers are exposed to liquid effects both from inside (by the product) and outside, they may become deformed and cannot fulfill their duty of protecting the product. For this reason, the resistance of packaging papers to liquids (water-oil, etc.) should be well known in order to make a selection appropriate to the content of the products. In this study, the effects of air permeability, liquid contact angle and surface energy values of various papers and cardboards used in the packaging industry on water and oil resistance were experimentally investigated. After determining the air permeability of the papers, liquid contact angle and liquid absorption measurements were carried out using the Sessile water drop method for surface wetting characterization. The surface energies of the papers were calculated depending on the liquid contact angle. Then the oil absorption of these papers was determined by measuring the time-dependent absorption of Castor oil. Differences in liquid resistance between papers were demonstrated and evaluated based on the air permeability of the papers, water contact angle, surface energy and liquid absorption. It was concluded that the wettability and surface energy of papers are the determining factors in water absorption, while air permeability is the determining factor in oil absorption.

KEY WORDS

air permeability, liquid contact angle, packaging materials, surface energy, liquid resistance

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Introduction

Interest in eco-friendly, biodegradable, recyclable and renewable paper and cardboard packaging materials instead of synthetic packaging materials is increasing day by day.

Paper materials can be used as primary packaging that comes into direct contact with the foods or as a secondary packaging that provides mechanical resistance (Tutak, 2025)

The main function of packaging materials is to protect the contents from contaminants, leaks and damage during the production, storage, transportation and sales processes, and to provide safety, convenience and economic benefit (Shen et al., 2021).

Nowadays, the demand for ready-made and packaged food or beverages is increasing day by day, and these food items are mostly presented in non-biodegradable, single-use synthetic plastic packaging. Disposable plates, cups and straws account for approximately 70% of total global plastic production (Hossain et al., 2021). Since most of the plastic packaging waste cannot be biodegraded, it causes environmental pollution and endangers the ecosystem and life (Azmin & Nor, 2020; Bastance et al., 2021). Recently, with the greatly increased anti-plastic sentiment, there has been an increasing trend towards green packaging materials based on renewable resources that are recyclable, compostable and/or biodegradable (Aydemir, Yenidoğan & Tutak, 2023; Nechita & Roman 2020). Cellulose-based paper and cardboard materials, especially sustainable and eco-friendly, attract great attention in packaging.

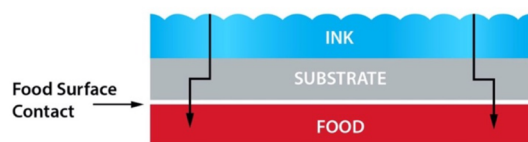
Compared to plastic materials, paper and cardboard materials have superior features in terms of being lightweight, biodegradable, recyclable, eco-friendly and low cost (Yenidoğan 2019; Herrera, Mathew & Oksman, 2017; Dai et al., 2021). When used as packaging materials, their good printability, mechanical strength, processability and ease of functionalization make them competitive.

Due to these properties, paper and cardboard materials are widely used in pharmaceutical packaging, food packaging, many other products such as paper bags, paper mats and paper placemat (Sheng, Li & Zhao, 2019).

However, depending on the content of the product, packaging paper and cardboard are expected to be resistant to liquids, especially water and oil. For example; Water and oil resistant paper and cardboard are needed for the packaging of refrigerated/frozen food labels, processed seafood, textile products, cosmetic products, paper filters and some other products. Therefore, knowing the water and oil resistance of paper and cardboard is critical for the safety of the product.

The liquid permeability of packaging paper is also important for printing processes. The resistance of papers to liquids determines the level of absorption of water, solvent and oil-based inks. The high liquid resistance of papers makes it difficult to absorb ink during the printing process and prolongs the drying time (Kandirmaz et al., 2020). In papers with low liquid resistance, ink is excessively absorbed by the paper layer (Yenidoğan, Aydemir & Ekinci Dogan, 2023).

In this case, the liquid phase of the ink (oil-water or solvent) penetrates the reverse side of the packaging paper (inside surface of the package) and may cause diffusion migration (Figure 1). Substances known as migrant ink components can penetrate from the printed side to the unprinted side of the packaging paper, and from there to the food due to their low molecular size (molecular weight <1000 daltons). This situation jeopardizes the safety of the product (contamination, etc.) in printed packages.



» **Figure 1:** Diffusion migration (Yenidoğan, Aydemir & Ekinci Dogan, 2023)

For all these reasons discussed above, the liquid resistance of packaging papers should be well known. In this study, the resistance of some papers to water and oil was tested in order to determine the right packaging material for different types of food and other products.

The effects of air permeability, liquid contact angle and surface energy parameters of papers on water and oil resistance were analyzed. Suggestions were made on choosing packaging paper suitable for the product content.

Experimental

Materials and Methods

The scope in study, the water and oil resistance of coated and uncoated papers (table 1), which are several commonly used in the packaging industry, were tested. As uncoated paper; Kraft Paper, Wrapping Paper, Sulphite Paper, as coated paper; Barrier Coated Paper (lacquered), Parchment Paper and Coated Paperboard were preferred.

Papers were obtained from companies operating in the packaging industry. After the samples were conditioned for 24 hours at $23\pm 1^\circ\text{C}$ and $50\pm 2\%$ relative humidity in accordance with the ISO 187:2022 standard, air permeability, water contact angles, water drop volume changes and oil absorption tests were performed and surface energies were calculated. All measurements were carried out in a conditioned laboratory environment.

Air Permeability

The surface properties of the paper are extremely important in the process of settling and absorption fluids on the paper surface (Abd El-Rahman et al., 2021).

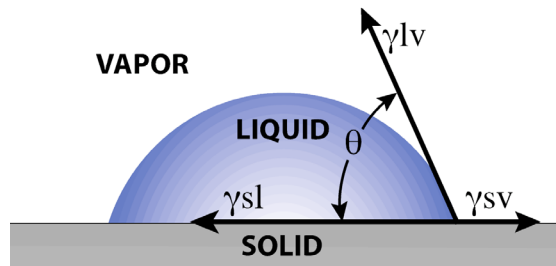
The permeability of paper and cardboard materials is related to their porous structure (Yenidoğan, Aydemir & Ekinci Dogan, 2023) and this porous structure is decisive for liquid permeability (Aydemir et al., 2021). Paper/boards have inherently poor barrier properties (low water and oil resistance) due to their porous structure and the hydrophilic character of cellulose fibers (Azmin & Nor, 2020). This limits the scope of application of papers.

Since the porosity of paper is not a directly measurable quantity, air permeability measurement is used to determine the porosity of paper. In this study, the air permeability of uncoated paper samples was measured with an L&W Air Permeance Tester (Lorentzen & Wettre, Kista, Sweden) according to TAP-PI T 460. Ten measurements were made from each sample and their averages were taken (Table 1).

Water Drop Volume Change and Contact Angle (Wettability)

A liquid droplet touching a paper surface tends to both penetrate into the paper structure and spread over the surface (Aydemir, Karademir & İmamoğlu, 2010).

When any liquid is dropped onto a paper surface, the droplet takes a curved shape when it first contacts the paper surface. An angle is formed between this curve and the paper surface; this angle is called the contact angle (Figure 2).



» **Figure 2:** Contact angle at the air-liquid-solid three-phase contact line

Liquid droplet contact angle measurement is a widely accepted method for analyzing the surface wettability and absorbency of polymeric materials. The rate of change in wettability (contact angle) is considered as a result of liquid absorption in the paper.

In this study, the wettability and water resistance of coated and uncoated papers were measured using a computer-integrated TMI Pocket Goniometer Model PG-X (FIBRO Systems AB Stockholm, Sweden).

First, distilled water droplets were placed on equal volumes of paper surfaces under constant experimental conditions. Then, the total volume and contact angle (TAPPI T 558 om-97) change values (Table 1) and images (Figure 3) of these sessile water drops on the papers were automatically measured and recorded for 4 minutes (50 data/minute) with a CCD video camera.

The changes of water drop volume and contact angle on the paper surfaces at 15 seconds, 45 seconds, 100 seconds and 240 seconds were analyzed.

Table 1

Test Paper Types and Properties

	Paper Type	Paper Symbol	Air Permeability (ml/min)	STDEV	Contact Angle (degree)	STDEV	Surface Energy (mJ/m ²)	STDEV
Uncoated Papers	Kraft Paper	P1	260	17,13	90,5	2,25	32,2	1,13
	Wrapping Paper	P2	220	3,28	92,8	1,86	31,3	2,55
	Sulphite Paper	P3	277	0	93,3	2,51	30,9	1,84
Coated Papers	Barrier Coated Paper	P4	-	0	83,6	1,26	36,5	0,17
	Parchment Paper	P5	-	0	71,9	3,7	43,2	2,54
	Coated Paperboard	P6	-	7,68	79,4	1,73	37,9	0,77

Surface Energy

The structure and surface energy of the paper greatly affect its interaction with liquids (Aydemir, Altay & Akyol, 2021). In this study, the surface energy of coated and uncoated papers was determined depending on the water contact angle according to the ASTM D5946 standard test method.

The relationship between static contact angle and surface energy forces is defined by Young–Dupré presented in Equation (1) from the interfacial tensions where γ_{sv} = solid–vapor interaction, γ_{sl} = solid–liquid interaction, and γ_{lv} = liquid–vapor interaction (Aydemir, Altay & Akyol, 2021; Altay et al., 2022).

Young's Equation

$$\gamma^{sv} = \gamma^{sl} + \gamma^{lv} \cos \theta \quad \cos \theta = \frac{\gamma^{sv} - \gamma^{sl}}{\gamma^{lv}} \quad (1)$$

θ is the contact angle

γ^{sv} is the solid/liquid interfacial free energy

γ^{sl} is the solid surface free energy

γ^{lv} is the liquid surface free energy

The surface energy of the test papers is listed in Table 1. Five samples were measured from each sample.

Measurements of Oil Resistance

Castor oil ($\gamma_{lv} = 39.0$ mN/m, $\eta = 889.3$ mPa/s, at 20°C) included in the TAPPI UM-557 test method procedure was used to determine the oil resistance of coated and uncoated papers. First, after the weights of the test papers were measured and recorded, Castor oil was dropped onto the surfaces of these papers in a fixed volume (from a height of approximately 13 mm) with a syringe. Castor oil on the surface of the papers was wiped with a tissue after 15 seconds, 45 seconds, 100 seconds and 240 seconds.

Then, the weights of these papers in each time period (15s, 45s, 100s and 240s) were measured separately (Figure 6) and their wettability was displayed separately (Figure 5). Thus, the absorption amounts of oil into papers and the resistance levels of papers against oil were analyzed.

Results and Discussions

The resistance to liquids of different types of paper and cardboard used in the packaging industry was tested using Distilled water and Castor oil. The measurement results and images are presented and discussed below.

Paper Surface Wettability

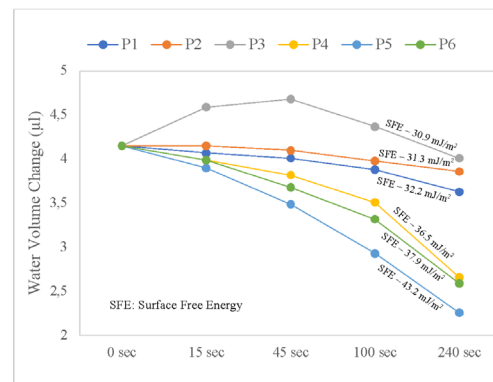
A liquid droplet dropped onto the paper surface, it shows spreading behavior on the paper surface and also tends to penetrate between the paper fibers. The extent of penetration or spreading depends on both the properties of paper and liquids (Aydemir, Karademir & İmamoğlu, 2010).

Contact angle measurement is a qualitative way to evaluate whether the surface has a hydrophobic or hydrophilic characteristic (Pasricha & Sachdev, 2017). Low contact-angle values demonstrate a tendency of the water to spread and adhere to the surface, whereas high contact-angle values show the surface's tendency to repel water (Huhtamäki et al., 2018; Aydemir et al., 2019).

When the wettability and absorbency of test papers are evaluated depending on the contact angle; it was determined that Parchment paper (P5), Coated Paperboard (P6) and Barrier lack coated (P4) paper surfaces had the highest wettability and absorbency. Sulphite Paper (P3), Wrapping Paper (P2) and Kraft Paper (P1) were found to be the papers with the lowest wettability and absorbency (Figure 3). These papers with low wettability also have low surface energy.

Water Absorbency of Paper Surfaces

According to the test results; The volume changes of the water droplet on different papers over time are shown graphically in Figure 4. These water absorption results are also compatible with the water-paper contact angles in Figure 3.



» **Figure 4:** Water drop volume change on paper surfaces

Time	0 s	15 s	45 s	100 s	240 s
Kraft Paper (P1)					
	90,5	90,1	87,1	85,98	80,24
Wrapping Paper (P2)					
	92,8	92,36	90,86	90,26	87,64
Sulphitic Paper (P3)					
	93,3	93,2	92,46	88,04	84,36
Barrier Coated Paper (P4)					
	83,6	81,1	75,88	70,08	63,36
Parchment Paper (P5)					
	71,9	52,98	42,72	37,44	32,5
Coated Paperboard (P6)					
	79,4	70,32	67,9	63,98	56,28

» **Figure 3:** Images and contact angles of sessile water drop on paper surfaces

According to the test results, Sulphite Paper (P3), Wrapping Paper (P2) and Kraft Paper (P1) are the papers with the lowest water absorbency (Figure 4). The common feature of these papers is that they are not coated. Among these papers, Sulphite Paper (P3) has the lowest wettability (high contact angle) and absorption rate, but hygro-expansion (also called swelling) has occurred on its surface (Figure 4). Swelling means that when water enters the cellulose fibers, the internal pore structure of the paper changes and the cellulose fibers swell, deteriorating the sheet form and functional properties (Scott & Abbott, 1995). This may be due to the fact that carboxymethylation, which is commonly used to add acidic groups to cellulose, increases the water retention value of the fibers. These papers have a slower absorption rate, indicating that swelling of the surface fibers may lead to the closure of the surface pores (Akinli-Kocak, 2001).

According to the test results, Parchment paper (P5), Coated Paperboard (P6) and Barrier Coated Paper (P4) showed the fastest water absorption. This shows that the water resistance of coated papers is weaker than uncoated papers. These papers have lower static contact angles and higher surface energies than coated papers.

Considering that paper with higher polar components has more affinity for water, it is thought that the high water absorbency of papers is due to the water-loving minerals in the coating structure (kaolin clay, calcium carbonate, titanium dioxide, talc, etc.). In addition, the coating binder level and the coating pore size can also affect the absorbency of the paper.

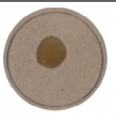
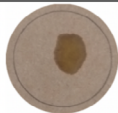
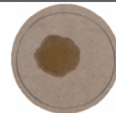
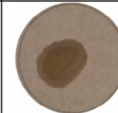
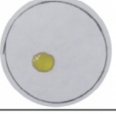
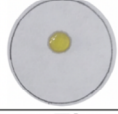
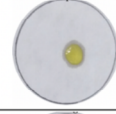
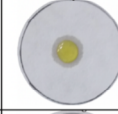
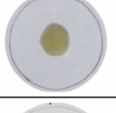
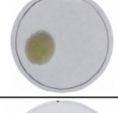
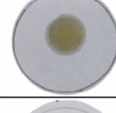
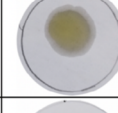
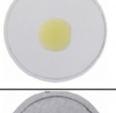
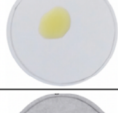
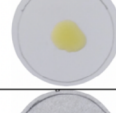
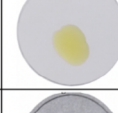
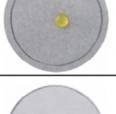
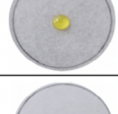
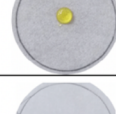
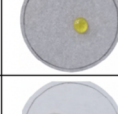




Oil Resistance of Papers

The large variety of grease-containing foods has motivated the production of oil-resistant paper materials used in food packaging in recent years (Wang et al., 2022).

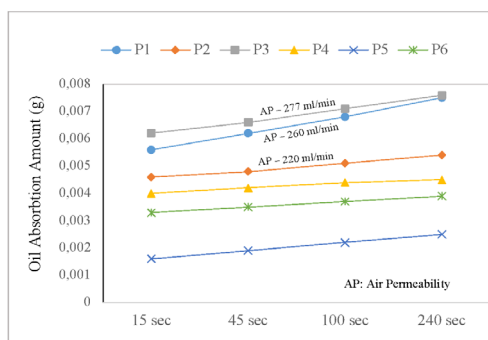
However, choosing the right packaging paper or board for fatty foods has become important due to reasons such as migration, contamination, and food spoilage. The oil absorption rate is a function of the surface absorption of the substrate. Castor Oil absorption test results of the papers used in packaging in this study are given below.

According to the test results; It has been determined that Sulphite Paper (P3) and Kraft Paper (P1) are the paper types with the highest oil absorption (Figure 5 – 6) and that the oil absorption in these papers increases over time (Figure 6). It is thought that the low oil resistance of these papers is due to their high degree of porosity. In this case, it becomes easier for the oil to pass through the paper fiber structure.

The high oil absorption of Kraft Paper (P1) is; It is known that it is caused by the removal of most of the lignin in the wood during the pulping process in order to provide strength. Low lignin is important to the resulting strength of the paper, as the hydrophobic nature of lignin interferes with the formation of the hydrogen bonds between cellulose (and hemicellulose) in the fibers. Because of this reason, the oil absorbency of kraft paper increased (Małachowska et al., 2020; Chandra, Lehtonen & Ragauskas, 2024).

Paper Type	15 s	45 s	100 s	240 s
Kraft Paper (P1)				
Wrapping Paper (P2)				
Sulphitic Paper (P3)				
Barrier Coated Paper (P4)				
Parchment Paper (P5)				
Coated Paperboard (P6)				

» **Figure 5:** Papers oil absorption images



» **Figure 6:** Oil Absorption values of papers and cardboards

According to test results, it was determined that the oil resistance of Parchment paper (P5) - Coated Paperboard (P6) and Barrier Coated Paper (P4) was higher than uncoated papers (Figure 5 – 6). The common feature of these papers is that they are coated papers. High surface energy coatings also showed better oil resistance due to higher polar component content. Another reason for the high oil resistance of these papers is that the porous structure in the paper matrix is closed.

Conclusion

The paper to be used in packaging must provide a barrier against water and oils. Knowing the liquid resistance of papers is important for the reliability of packaging and determines the scope of application.

The liquid resistance of paper and cardboard is also important for the correct management of packaging printing production processes. Printability parameters such as adhesion, drying, color and gloss are related to the penetration of ink into the paper. For this reason, in packaging printing applications with water, oil and solvent-based inks and varnishes, the right paper must be selected by taking into account the liquid permeability of the papers.

In this study, the differences between papers are demonstrated and evaluated on the basis of air permeability, liquid-solid contact angle, surface energy and liquid absorption. According to these evaluations;

- Wrapping Paper (P2) and Kraft Paper (P1) are paper types that are more resistant to water than coated papers.
- Although Sulphite Paper (P3) has low wettability, its usage area must be determined correctly due to the surface deformation that may occur when its surface interacts with water.
- Parchment paper (P5), Coated Paperboard (P6) and Barrier Coated Paper (P4) are the paper types with the highest resistance to oil.

These papers can be used as food paper and baking paper or in the packaging of non-food products that require resistance to oil.

- Parchment paper (P5), Coated Paperboard (P6) and Barrier Coated Paper (P4) were the paper types with the lowest resistance to water, and Sulphite Paper (P3), Kraft Paper (P1) and Wrapping Paper (P2) were the paper types with the lowest resistance to oil.
- Parchment paper (P5) is the most oil-resistant type of paper. However, the water resistance of this paper is lower than all other paper types.
- As the wettability and surface energy of papers decreased, their resistance to water increased. Therefore, the water resistance of papers can be related to their wettability (contact angle) and surface energy.
- When the air permeability of uncoated papers increased, the oil absorbency also increased, and when the air permeability decreased, the oil absorbency also decreased. These results showed that the oil resistance of uncoated papers is correlated with air permeability. Therefore, the oil permeability of paper can be characterized by its air permeability.
- The liquid resistance of the paper and cardboard to be used in packaging should be well known. However, in packages where high oil resistance is desired, papers with low air permeability and high surface energy should be preferred, and in packages where high water resistance is desired, papers with low wettability and surface energy should be preferred.
- In general, the results have been shown that papers with high water resistance have low oil resistance, and papers with high oil resistance have low water resistance. This reveals that the resistance of papers to oil and water is different. It has been determined that uncoated papers are resistant to water, while coated papers are resistant to oil. This shows that the liquid resistance of papers depends on the surface structure, coating type, and the properties of the liquid in contact with its surface (density, etc.).
- In order to obtain water- and oil-resistant packaging, the surfaces of the papers can be coated with environmentally friendly and sustainable bio-containing materials (such as alginate and chitosan).

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