

Article

Parameters Affecting the Water Vapour Permeability of Gelatin Films as Evaluated by the Infrared Detecting Method ASTM F1249

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Abstract: The purpose of this study was to assess testing parameters for measurement of water vapour permeability (WVP) properties of bovine gelatin films by ASTM F1249. This method utilises an infrared sensor to determine the WVP of conventional plastic-based films and has been widely applied within the packaging industry, but has had very limited application with hydrophilic compostable/edible packaging materials. These films have low WVP properties with highly variable WVP values (as studied by ASTM E96); consequently, this parameter has to be carefully controlled. Assessment of the module was carried out using bovine gelatin films with different thicknesses of film, and gelatin was chosen as an example of such film types using the following Mocon Permatran testing parameters: 50 or 70% relative humidity (RH), 100 or 150 cm³ nitrogen gas flow rates for the module's RH-cell (100 cm³ for all other cells). In all cases, WVP increased with an increase in gas flow rate. This method showed that WVP increased with increasing gelatin film thickness, and categorically supports previous—and often unexplained—WVP findings for other hydrophilic, biopolymer films. The current study is a helpful guide to the examination of water barrier properties of hydrophilic edible/biodegradable/compostable packaging materials in research and industry.

Keywords: gelatin; edible/biodegradable/compostable films; water vapour barrier; ASTM F1249; gas transport; water vapour permeability



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1. Introduction

The use of sustainable sources of biopolymers which could function as effectively as conventional plastics when applied to food products, yet also be biodegradable/compostable, would help reduce or eliminate conventional plastic use in a wide range of packaging applications. Edible, biodegradable and compostable (EBC) films can be derived directly from by-products of food manufacturing waste streams which are high in proteins or polysaccharides or both. However, the major disadvantage of their use for food packaging is due to their sensitivity to high levels of humidity [1–6]. Gas barrier properties are critical in EBC packaging material development and the understanding of gas transport mechanisms within different packaging materials is pivotal in order to understand the role that numerous factors play in impacting upon a material's ability to serve as an adequate gas barrier. High water vapour penetration through EBC films can encourage microbial growth and food spoilage, can destroy ingredient structures and their functionalities, and can impact food texture and produce unsightly pack fogging or misting (if vapours condense). For these reasons, the control of water vapour permeability through food packaging films is a pivotal matter within the industry and needs to be tested precisely and effectively.

Gelatin has been used to form edible films and coatings since the 1960s [1,7–9]. Films formed using gelatin sources have good mechanical properties and can be manufactured in different ways [7,10,11]. However, as an EBC biopolymer, gelatin suffers like

all hydrocolloid-based films in terms of its hygroscopicity. In the literature it has been found that WVP values for gelatin films are hard to compare with each other. For this reason, we previously reviewed the WVP values of gelatin films reported in the literature whose manufacturing processes differed in terms of plasticizers used, concentrations, thicknesses, relative humidities applied, temperatures and test methods for vapour permeability (WVP) values for which were normalized to the same units [12]. It was found that WVP values varied widely depending on plasticizer used, film thickness and gelatin content, and method preparation (Table 1). In addition, molecular weight can influence the results of gelatin WVP because it varies depending on the manufacturing process used. In fact, WVP values reported in the literature for gelatin films are widely diverse: 481.5 ± 7.30 [13]; 0.5 ± 0.05 to 5.6 ± 0.05 g mm/m² d atm [14]; 3959.7 ± 45.60 to 9976.5 ± 623.15 mm/m² d atm [7,10]; 37.6 ± 3.42 mm/m² d atm [5]; 26438.5 ± 1225.63 mm/m² d atm [15]; 2.8 ± 0.33 to 196.1 ± 0.89 mm/m² d atm [16]; 2188.62 mm/m² d atm [17] depending on plasticizer used, film thickness and gelatin content (Table 1) [12].

Table 1. Normalized WVP property comparison of gelatin films depending on biopolymer source, plasticizer type and concentration, thickness value, relative humidity, temperature and test method (Adopted from [12]).

Bovine Gelatin Content of Film	Plasticizer Type and Concentration	Thickness, (μm)	WVP (g mm/m ² d atm)	Test Relative Humidity RH, (%)	Temperature, (°C)	ASTM
Gelatin 10%	Glycerol 4.5% to gelatin	80.0 ± 4.0	481.5 ± 7.30	50	25	E96
Gelatin 1%	Sorbitol 15% to gelatin	43 ± 9	413.406	100	22	E96
	Sorbitol 45% to gelatin		705.222			
	Sorbitol 65% to gelatin		924.084			
Gelatin 12%	Malic acid 20% to gelatin	21.0 ± 0.6	0.5 ± 0.05	50	25	E96
	Polyethylene glycol 20% to gelatin	23.2 ± 0.7	3.3 ± 0.15			
	Sorbitol 20% to gelatin	23.7 ± 0.4	0.7 ± 0.05			
	Ethylene glycol 20% to gelatin	22.4 ± 0.7	5.6 ± 0.05			
	Diethylene glycol 20% to gelatin	23.0 ± 0.8	4.7 ± 0.05			
	Triethylene glycol 20% to gelatin	22.5 ± 0.5	4.6 ± 0.07			
		23.2 ± 0.7	3.0 ± 0.02			
	Ethanolamine 20% to gelatin	21.5 ± 0.5	2.4 ± 0.05			
	Diethanolamine 20% to gelatin	23.4 ± 0.5	3.1 ± 0.05			
Gelatin	Glycerol 0.25% water	56.5 ± 7.43	$10,993.8 \pm 531.96$	50	23 ± 2	E96
		43.0 ± 9.50	7802.0 ± 531.96			
		40.8 ± 5.36	6383.5 ± 106.39			
		50.3 ± 3.79	9023.0 ± 106.39			
		46.9 ± 4.15	9976.5 ± 623.15			
		55.0 ± 4.06	7014.7 ± 373.89			
Gelatin	Glycerol 0.2% to gelatin	20.7 ± 3.2	4063.1 ± 137.80	50	23 ± 2	E96
	Glycerol 0.5%	23.3 ± 2.2	4352.9 ± 15.20			
	Glycerol 0.8%	21.1 ± 6.6	3959.7 ± 45.60			
	Glycerol 1.1%	23.4 ± 2.5	4869.7 ± 122.60			
Gelatin 5%	Glycerol 33% to gelatin	57 ± 6	37.6 ± 3.42	50	23 ± 2	E96
Gelatin 3.34%	Sorbitol 2% to solution	N/A	26438.5 ± 1225.63	50	25	E96
Gelatin	Glycerol 40% to gelatin	125 ± 25	2.8 ± 0.33	35	23	F1249
			14.7 ± 1.18	50		
			77.8 ± 0.48	70		
			196.1 ± 0.89	90		
Gelatin	Glycerol 30% to gelatin	58 ± 4	2188.62	65	25	E96
Gelatin (only fish gelatin) 20%	-	None	1630 ± 300	50	25	F1249

As can be seen from the table, even when the same conditions apply, there is a variability of WVP values across the different studies. Typically, these results can be compared only within experimental batches of individual studies; in most cases, they cannot be compared

to the other sources. The same situation can be found concerning other EBC films. There is an absence of a standardized WVP method for hydrophilic films [12].

Currently, two ASTM standard test methods exist to evaluate water barrier properties of packaging materials. ASTM E96 [18], commonly referred to as the cup method, is a gravimetric assay usually adopted for the WVP testing of EBC biopolymeric films. The other method is ASTM F1249, commonly known as the infrared detecting method, which provides much more accurate results compared to the E96 method [19]. ASTM F1249 [20] is widely used as the ‘gold standard’ for industrial assessment of WVP in conventional plastic-based polymers used for packaging applications. The application of this method using Mocon Permatran (USA) equipment is rather complex and requires a specific instrumentation setup procedure, and adjustment of many testing parameters depending upon the test materials under scrutiny. The application of the Permatran for EBC biopolymeric films has been limited within the scientific literature, (probably attributable to the high cost of purchase of this equipment). The only two gelatin-related references which we were able to source with regard to the application of this method were reported by Ciannamea et al. [16], and Yi et al. [21]. Therefore, there is a requirement to investigate and determine the appropriate conditions necessary for WVP testing of EBC films, using the Permatran 3/33 module.

For this reason, the objective of this study was to optimise Permatran operating conditions for optimal measurement of WVP properties of gelatin films and further WVP controlling of hydrophilic films, e.g., pectin and others. This approach might serve as a guide for future studies concerning WVP measurement in gelatin and other biopolymer-based films for research and industrial WVP control. The module can be set up to manipulate the following: different values of relative humidity (RH) from 35 to 100%; gas flow rates on each of the cells A, B, C, Ref, and RH from 10 to 150 cm³ (film testing area 1.1 cm²); and temperature (usually 25 or 37.9 °C). Because of this, the instrument offers an ideal means of establishing how well this testing system operates with EBC films. Therefore, another objective of this study was to determine if the ASTM F1249 [20] method for the Permatran module was suitable for WVP assessment of gelatin film and, if so, to determine whether it was possible to optimize the test conditions necessary to produce reliable WVP properties. The effect of film thickness on the water barrier properties of gelatin film was also investigated.

2. Materials and Methods

2.1. Reagents

Gelatin from bovine skin, gel strength 225 Bloom, type B gelatin and glycerol (ReagentPlus® ≥99.9% (GC), both sourced from SIGMA-ALDRICH (St. Louis, MO, USA), were used for preparation of films. Nitrogen gas “pure grade” (Air Products and Chemicals, Inc., Walton on Thames, UK) was used for conducting the ASTM F1249 method.

2.2. Method of Selection and Preparation of Gelatin Films

Wang et al. [2] showed that packaging-related properties of edible/biodegradable/compostable films were dependent upon ingredient sources and their inherent composition. All of the physical tests conducted on protein-based films were carried out on films with ingredient concentrations ranging from 4 to 16% (*w/v*). The use of lower protein concentrations resulted in poor protein interaction; conversely, higher protein contents produced too much interaction, thereby producing very viscous protein solutions. A specific viscosity range was chosen by Wang et al. [2] in order to prevent physical defects within films (pinholes, cracking and distortion), and to prevent dissolving difficulties with proteins and the formation of bubbles. Gelatin-based films manufactured using 4% and 8% forming solutions (*w/v*) exhibited good results for the following qualities: tensile strength, elongation at break point, puncture strength, tear strengths, water resistance, acid resistance, alkali resistance, oxygen imperme-

ability, and oil impermeability [2]. Wang et al. [2] also assessed different production temperatures, but all optimal films were obtained from 80 °C heated solutions, thereby guaranteeing a compact film structure possessing less internal free space and, consequently, possessing greater impermeability properties. After considering all of the above, a solution of 4.69% gelatin with 33% *w/w* of glycerol (added in proportion to gelatin content) heated at 90 °C was chosen to create the gelatin films employed for this study.

Films were prepared according to the method developed by Clarke et al. [5] with minor modification. Gelatin (5 g) was dissolved in 100 mL solution in distilled water at 90 °C and stored in a shaken bath (Julabo SW 23, Seelbach, Germany) at 90 rpm for 30 min. An amount of 1.65 g of glycerol heated at 90 °C was added into the mixture (33% *w/w* dry gelatin matter). The final concentration of gelatin in film-forming solution was 4.69%. The film-forming solution was cooled down to 40 °C and a specific volume of 4–18 mL, depending on the necessary film thickness, was placed onto polystyrene Petri dishes (92 × 16 mm, (Sarstedt Inc. Numbrecht, Germany)) in order to obtain the desired gelatine film thickness (50, 100, or 150 µm). Petri dishes were then placed inside a constant climate chamber at 50% RH at 25 °C (Binder Series KBF, Tuttlingen, Germany) with fan assistance permanently supplied, as recommended by Delgado et al. [22]. In Table 2 different volumes of the film-forming solution and the appropriate film thicknesses obtained are represented.

Table 2. Gelatin film forming solution volume per Petri dish and resulting film thickness.

V, (mL)	Average Film Thickness Obtained (µm)
4	20 ± 2
7	50 ± 1
13	100 ± 6
18	150 ± 10

2.3. Film Thickness

In order to obtain a desired film thickness, different trials were carried out to achieve this. In particular, different amounts of gelatin solution were poured into Petri dishes (9.2 cm diameter) and, following plasticization, film thickness was measured using a digital micrometer. Based on these studies, an optimised pouring gelatin solution was determined (Table 2).

As reported by Bertuzzi et al. [23], the thickness of edible, starch-based films results from a linear function of the film-forming solution poured into Petri dishes. A non-dependence of film thickness at RH values below 60% during conditioning was also demonstrated by the same authors. Consequently, all gelatin films manufactured as part of this study were placed inside a climate chamber and held at 50% RH, a value commonly reported in numerous similar studies [2,7,11,14].

Film thickness was measured using a digital micrometer screw gauge (Käfer, Villingen-Schwenningen, Germany). The test areas on the film were marked and thickness values were expressed as an average of four measurements taken within the marked film area. For the measurement of film thickness for the whole film sample taken from the Petri dish, on average, 5 measurements were taken from each different film area.

2.4. Film Uniformity

Film uniformity was studied by optical microscopy cross-section on an Olympus BX 61 microscope (Tokyo, Japan) equipped with a digital camera (Supplementary Materials, Figure S1). We did not carry out any SEM studies of our films because this method can describe only one tiny part of the film. However, we have reported SEM studies of gelatin films in laminate structures previously (Supplementary Materials, Figure S2).

2.5. WVP Studies

WVP properties of gelatin films were measured in accordance with ASTM F1249 [20]; all the measurements taken using this standard were obtained using a MOCON Permatran 3/33 module (Ametec Inc., Minneapolis, MN, USA). A gelatin film square (2×2 cm) was placed between two aluminium masks (Labthink Instruments Co., Ltd., Jinan, China). Then, masks with the sample inside were mounted between two test cells within the module. Inside the two masks and on the sealing rims of the two cells, high vacuum grease (Dow Corning Corporation Midland, Michigan, MI, USA) was spread where the test film was placed. This was carried out to ensure that a vacuum environment was created while the test was conducted. The effective test area was 1.131 cm^2 or 5 cm^2 ; environmental conditions in cells were set at 37.9°C or 25°C ; RH was 50 or 70%, and nitrogen gas flow rate (Mocon manual [24]) was set to 20, 100 or 150 sccm for the RH cell.

Before all experiments were carried out, a one-hour conditioning period (at test conditions) was employed for the testing apparatus. The choice of the temperature used was based on the default mode of the module (37.8°C) and on the temperature conditions reported previously in the literature (25°C) [16].

The RH of gases was set by rotating the RH pressure regulator [23]. Permeation values were provided by WinPerm UI software v.2.72 (Brooklyn Park, MN, USA). Three to five replicates for each condition and film thickness were performed. Values were based on a mean value and standard deviation shown by figures on columns. Data were analysed with ANOVA one way ($\alpha = 0.05$) along with t test ($\alpha = 0.05$).

As was recommended previously [12], the unit of $\text{g} \times \text{mm}/\text{m}^2 \text{ d atm}$ was used for WVP in order to make it more easily comparable to other published WVP data. All of the experimental condition combinations applied are reported in Table 3.

Table 3. Condition combinations of Permatran 3/33 module for RH cell gas flow rate, film test area, RH and temperature.

RH Cell Gas Flow Rate, (sccm)	Cells A, B, Z, Ref Gas Flow Rate, (sccm)	Test Area, (cm^2)	RH, (%)	Temperature, ($^\circ\text{C}$)
20	10	1.131	50	37.8
100	100	1.131	50 or 70	37.8
150	100	5	50 or 70	37.8
100	100	5	50	25

2.6. The Effect of RH and Flow Rate on WVP Measurements Using Gelatin Films

Different film thicknesses were chosen to measure WVP at different RH values, and several combinations of gas flow rates were applied to specific cells. On the module it is possible to operate different handles representing different cell gas flow rate regulators. Specifically, the first trial was carried out by setting 10 sccm gas flows for cells A, B, Z and R, while for the RH cell a 20 sccm gas flow rate was employed.

These conditions (10 sccm gas flows for cells A, B, Z, and R; and 20 sccm for the RH cell) are usually applied to packaging materials possessing high barrier properties with respect to water vapours (for example, polyolefin films), and are typically used by flexible packaging manufacturers to control WVP in commercial plastics, co-extrusions and laminates. The results determined by this study for conditions usually used for plastic-based films are shown in Table 4.

Table 4. WVP values from Permatran 3/33, (10 sccm gas flows for cells A, B, Z, and R; and 20 sccm for the RH cell, 50% RH, 37.9°C).

Gelatin Film Thickness, (mm)	WVP, ($\text{g mm}/\text{m}^2 \text{ d atm}$)
0.020 ± 0.0008	84.4 ± 10.99
0.030 ± 0.0004	91.8 ± 18.36
0.041 ± 0.0001	104.5 ± 24.15

Numerous tests failed owing to a too-high level of water vapour permeating through the film, thereby resulting in the automatic shutdown of the analytical instrument in order to avoid sensor damage. As described in studies conducted by Yi et al. [21] and Ciannamea et al. [16], gas flow rates were increased 10-fold within cells A, B, Z, and Ref, while rates of 100 and 150 sccm were used for the RH cell. In this scenario, WVP measurements were repeated for 0.020 mm films at 100 sccm gas flows for cells A, B, Z, and Ref, and at 100 or 150 sccm for the RH cell (Supplementary Materials, Table S1). Comparable results for WVP values were found only at 50% RH for 100 and 150 sccm RH cell gas flow rates, respectively, as follows: 12.57 g mm/m² day atm (at 100 sccm) and 12.64 g mm/m² day atm (at 150 sccm), 50% RH, 37.8 °C (Supplementary Materials, Table S1). With the increase of RH up to 70% for the 100 sccm RH cell flow rate, the average WVP value (based on 4 experiments) increased to 24.13 g mm/m² day atm, 37.8 °C. In this case, a higher range of WVP values was observed for experimental results, from 15.13 to 28.77 g mm/m² day atm. The WVP for 150 sccm RH cell gas flow rate at 70% RH was not possible to measure because of experimental failure after several attempts (Supplementary Materials, Table S1). We consider that the film thickness of 20 µm was too thin to measure WVP at the high humidity of 70% RH, and the film could melt or be damaged at high gas flow rates by faster water absorption. Thus, WVP measurements began with a higher film thickness of 50 µm, followed by 100 and 150 µm.

All tests were carried out in “Convergence by cycles” mode, which stops the machine when two comparable results are obtained sequentially. Before the commencement of the test, examination or testing time can be set between 2 and 240 min. However, transmission rates for films with poor barrier properties do not increase significantly with increments of time (Permatran 3/33 operator’s guide [23]). For this reason, testing times of 3 and 10 min were employed during these studies, while temperature was set at 37.9 °C. No significant differences were observed between the two testing times and, therefore, a test time of 3 min was chosen for all subsequent film testing experiments.

3. Results

The WVP values obtained for two different RH cell flow rates are shown in Figure 1. A positive trend was observed for WVP values, both RH flow rates and gelatin film thicknesses.

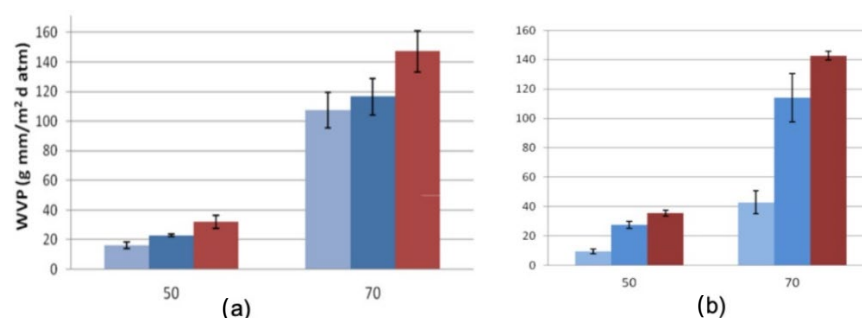


Figure 1. Gelatin film WVP (g mm/m² day atm) values depending on RH (50 or 70%), and gas flow RH-cell: (a) 100 sccm rate and (b) 150 sccm, for (■) 50 µm, (■) 100 µm and (■) 150 µm film thickness, ASTM F1249, 37.9 °C.

The WVP values obtained for the two RH values of 50 and 70% were statistically different ($\alpha = 0.05$) for all film thicknesses analysed (50, 100, 150 µm) and for both RH cell gas flow rates (100 and 150 cm³). This finding is due to the hydrophilic nature of gelatin. As RH increases, gelatin solubilisation increases and, as it does so, the value of WVP increases. Similar findings have been reported by other authors when employing gelatin films [16,21] and transglutaminase cross-linked gelatin films [25,26]. The results determined in this study are comparable with similar data reported by Ciannamea et al. [16]. These authors employed gelatin films with a thickness of 100 ± 25 µm and assessed WVP rates at 23 °C

on a Permatran module (RH gas flow rate of 150 cm³) and reported that WVP increased with an increase in RH.

According to Balaguer et al. [26], this behaviour is a result of enhancement of water absorption by gelatin film, where water molecules create hydrogen bonds with polar groups such as OH that characterize this type of film, thereby acting as plasticizers.

3.1. The Effect of Film Thickness on WVP Measurements in Gelatin Films

WVP refers to how easily water vapour can penetrate through a film. In the present investigation, a dependency of WVP upon gelatin film thickness was clearly shown. WVP values determined for gelatin film thicknesses of 50, 100 and 150 µm, tested at different RH values (50 and 70%), are reported in Figure 2 (for 20 µm film thickness comparison see Supplementary Materials, Figure S3). WVP values increased ($\alpha = 0.05$) with increasing film thickness under all examined and test conditions.

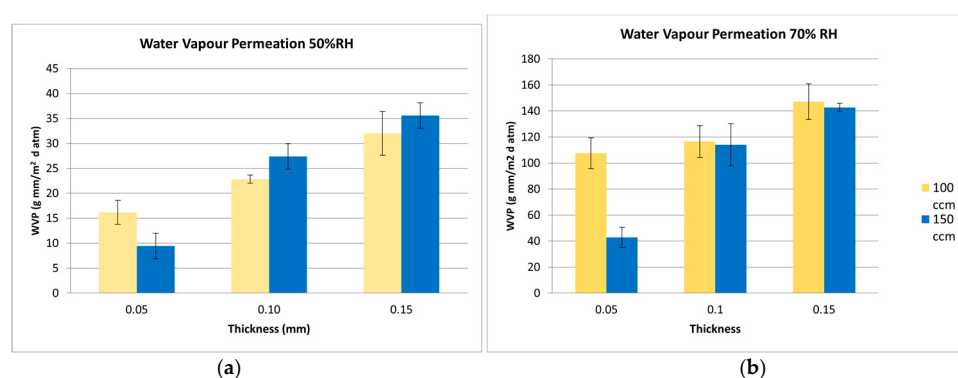


Figure 2. Gelatin film WVP values depending on film thickness at (a) 50% and (b) 70% RH; (■) 100 and (■) 150 cm³ RH cell gas flow rates, ASTM F1249, 37.9 °C.

These results are in line with those observed for other hydrophilic films, such as those reported by Roy et al. [27] who investigated cast wheat gluten films. This phenomenon of the thickness effect is also in accordance with Bourlieu et al. [28], who investigated lipid-based edible films; with Delgado et al. [22], who investigated yeast biomass-based films; and with Wang et al. [3], who studied gelatin films made from 4 and 8% film-forming solutions. The behaviour observed in gelatin film in this study is contrary to that reported for hydrophobic packaging films such as oil-sourced polyolefins (e.g., polyethylene or polypropylene films) in which water vapour barrier properties usually increase with increasing film thicknesses, thereby resulting in packaging materials with lower water penetration [29,30].

Gas transport through a film depends upon the penetrant nature of the gas, the film material itself, and the interaction between them. Gas transport through a film occurs in three stages. Initially, the polymer surface adsorbs the penetrant from the film side presented with the highest concentration of the gas. Then, the molecules of the penetrant gas are transported through the polymer matrix under gradient (diffusion mechanism) and finally, the penetrant gas is desorbed on the film side presenting the lowest concentration of the penetrant gas [31]. This system can be described through combining Fick's first law (on diffusion) and Henry's law (on solubility), where P , D , and S represent permeation, diffusion and solubility, respectively, as required for Equation (1) (Basics of Barriers [32]).

$$P = D \times S \quad (1)$$

Solubility of a gas represents the amount of the gas that can be retained by a material, and it is measured as a volume of gas per volume or mass of material. Diffusion of a gas expresses how quickly the gas moves through a material. Consequently, permeation can be

described as a steady-state property which determines the rate of a gas that passes under a gradient, and this can be expressed by Equation (2) (Basics of Barriers [32]).

$$P = (q \times l) / (A \times t \times \Delta p) \quad (2)$$

where q is the quantity of permeate; l is the thickness of the polymer; A is the area of the test material; t is the time and Δp is the difference of partial pressure between the two sides of the polymer.

Gelatin film is composed of hydrophilic peptide units and possesses low barrier properties against water vapour, which are further impacted by the addition of plasticizers (which are also hydrophilic in nature), thereby further reducing barrier properties [2,3,28]. Equation (1) does not truly explain hydrophilic film behaviour when water permeation of films is considered. Specifically, both diffusion and solubility vary with the degree of moisture absorption by films, which subsequently produces plasticization and swelling. Consequently, there is no one specific WVP value that can typify a hydrophilic film held under a wide range of environmental conditions. Therefore, WVP rates are dependent upon both the penetrant gas (water vapour) and the film material being assessed; as a result, RH and film thickness are factors which must be ascertained before any attempt to determine WVP for a specific packaging material. The non-linearity of water sorption is another issue that also needs to be addressed, because it defines the RH-dependent relationship with WVP measurement. Bourlieu et al. [28] reported a mathematical model on this topic from Buonocore et al. [33], employing some biopolymeric films (gelatin not included) which were validated within a range of 30–80% RH at 20 °C.

Film thickness affects water permeation in hydrophilic films. Such films show a positive relationship between WVP and film thickness values, as different film thicknesses can determine different film structures. Film homogeneity and greater film thicknesses means that more hydrophilic molecules reside inside the film material, thereby presenting the potential for greater interaction with water.

Two further reasons put forward to explain the relationship between WVP values and film thickness in hydrophilic films are: enhancement of the water vapour gradient on one side of the film (the cause of major resistance to permeation brought about by greater film thickness values) that finally results in a greater water vapour permeability; and the swelling of the matrix which results in hydrophilic films absorbing water and which promotes swelling on one side of the film. This occurs to a greater degree in thinner rather than thicker films and, consequently, water movement can be more affected and delayed [8].

3.2. ASTM F1249 Method Results Compatibility for Gelatin Film

In order to compare method conditions appropriate for WVP testing of gelatin films on a Permatran module, an additional test was carried out at 25 °C in order to compare the results with the data presented in the scientific literature (Table 5). For this assessment, films of similar thickness were employed.

Table 5. WVP values comparison of gelatin films analysed on Permatran 3/33 module (“exam time” set to 3 min on Permatran), 5 cm² mask.

Thickness, (μm)	Temperature, (°C)	WVP, (g mm/m ² day atm)	Reference (@ RH = 50%)
125 ± 25	25	12.9 ± 2.36	Current study: RH cell flow = 150 cm ³ , gas flow for other cells = 100 cm ³
125 ± 25	23	14.7 ± 1.18	[16]: RH cell flow = 150 cm ³ , gas flow for other cells = 100 cm ³ **

** In this research, results are given in g mm/m² day in the table and are given in g mil/m² day in Ciannamea et al. [16]. From communication with the authors of [16] the right values must be read as g mil/m² day atm, which are then normalized to g mm/m² day.

The difference between the WVP results obtained with Permatran module in the two studies was about 2 g mm/m² day atm, as can be seen from Table 5, thereby indicating good comparability between the studies. Such a difference can be explained by the temperature difference of 2 °C between the experiments conducted.

4. Conclusions

For the first time, specific Permatran 3/33 module conditions (at 50 and 70% RH, and room temperature and 37.9 °C) for the application of the standard method ASTM F1249 were optimized for WVP measurement of bovine-derived gelatin films. WVP properties for gelatin films were analysed at different film thicknesses, gas flow rates and relative humidities. The optimized conditions provided replicable and comparable results for RH, using an RH-cell system employing N₂ gas flow rates of 100 or 150 cm³. An increase in WVP, with an increase in film thickness, was observed for gelatin films, confirming the behaviour commonly reported for other hydrophilic biopolymeric films by ASTM E96. The current study may serve as a helpful guide for other barrier property studies and testing methods for application to biopolymeric films. This may prove to be important as the market begins to develop more commercially relevant, biopolymeric-based packaging materials for food applications in a bid to produce more sustainable forms of ‘Green Plastics’ over current conventional plastic-based forms and their applications.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/su14159018/s1>. Figure S1: Optical microscopy of gelatin films cross-section, Olympus BX 61. Figure S2: SEM image of Gelatin film (G-layer) structure in gelatin—sodium alginate (SA) duplex laminate. Adopted from Tyuftin et al. 2020 [34]. Figure S3: Gelatin film WVP values depending on film thickness at (a) 50% and (b) 70% RH; for 100 and 150 cm³ RH cell gas flow rates, ASTM F-1249, 37.9 °C. Table S1: WVP, g mm/m² day atm for 20 µm film, 37.8 °C.

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