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A Review of the Gas and Vapor Transport Through Single Polymer Films: Implications for Their Use in Book and Paper Conservation

Überblick über Gas- und Dampftransport durch einzelne Polymerfilme: Implikationen für deren Einsatz in der Buch- und Papierrestaurierung

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Abstract: Permeability is broadly defined as the ability of a material to allow the transportation of liquids, gases, or vapors through it. Although numerous references can be found in the literature giving the permeability values of polymers, there is no standard unit of gas or vapor transport, making comparisons difficult. This review summarizes the permeability of single polymer films used in book and paper conservation, specifically addressing the permeability of storage materials and processes such as encapsulation. In particular, the permeability of the polymer films to oxygen, nitrogen, hydrogen sulfide, water vapor, and acetic acid is summarized. The permeability was found to differ by over 7 orders of magnitude when comparing different gases and vapors diffusing through the polymer films. The permeability is dependent on the molecular size of the diffusing gas or vapor, with smaller molecules diffusing faster than larger molecules. In addition, the chemistry of the film plays a role. Hydrophilic polymers have a much greater permeability of polar diffusants such as water vapor than hydrophobic polymers. In addition, the permeation increases significantly with temperature. The review provides data of the permeability properties of the films used by conservators and will enable them to make an informed decision of the best material for their application.

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Zusammenfassung: Die Permeabilität ist allgemein definiert als die Fähigkeit eines Materials, den Transport von Flüssigkeiten, Gasen oder Dämpfen durch das Material zu ermöglichen. Obwohl in der Literatur zahlreiche Angaben für die Permeabilitätswerte von Polymeren zu finden sind, gibt es keine Standardeinheit für den Gas- oder Dampftransport, was einen Vergleich erschwert. Diese Übersicht fasst die Permeabilität einzelner Polymerfolien zusammen, die in der Buch- und Papierrestaurierung verwendet werden, und befasst sich mit der Permeabilität von Materialien, die bei Verpackung, (Langzeit)aufbewahrung oder zur Stabilisierung eingesetzt werden. Insbesondere wird die Durchlässigkeit der Folien für Sauerstoff, Stickstoff, Schwefelwasserstoff, Wasserdampf und Essigsäure zusammengefasst. Beim Vergleich verschiedener Gase und Dämpfe, die durch die Polymerfolien diffundieren, wurde festgestellt, dass sich die Durchlässigkeit um mehr als 7 Größenordnungen unterscheidet. Die Durchlässigkeit hängt von der Molekülgröße des diffundierenden Gases oder Dampfes ab, wobei kleinere Moleküle schneller diffundieren als größere Moleküle. Darüber hinaus spielt auch die chemische Zusammensetzung der Folie eine Rolle. Hydrophile Polymere haben eine viel größere Durchlässigkeit für polare Diffusionsmittel wie Wasserdampf als hydrophobe Polymere. Darüber hinaus nimmt die Permeation mit der Temperatur deutlich zu. Die Übersicht liefert Daten zu den Durchlässigkeitseigenschaften von in der Restaurierung verwendeter Folien und liefert eine fundierte Entscheidungshilfe bei der Auswahl von Folienmaterial.

Schlüsselwörter: Permeabilität; Polymere; Polyethylenterephthalat; Gas; Dämpfe

1 Introduction

Polymer films and coatings are used in many areas of conservation (Chu et al. 2023; Taylor 1985; “The Conservation and Art Materials Encyclopedia Online (Cameo)” 2024). For example, polymers are used for the repair and protection of artwork and to separate two materials that might otherwise corrode or react when in contact. In book and paper conservation, polymer films are used in encapsulation (McGath et al. 2017; Minter 1983; Polyester Film Encapsulation 1980), lamination (McGath 2017; McGath, Hall, and McGuiggan 2017), as consolidants and adhesives (Phelan, Baer, and Indictor 1971), and as an interleaf. Often, polymer films are used to give support and mechanical strength to a material and may be used to protect or isolate the material from the environment.

Some polymer films provide a barrier to the transport of specific gases through the film, others are semi-permeable to gases while others are permeable.

Permeability is broadly defined as the ability of a material to allow the transport of liquids, gases, or vapors through it. It is important in various science and technology applications including geology, gas separation, food and electronic packaging, coatings, and biomedical devices (Oreski et al. 2017; Robertson 1993). Because of its industrial importance, many references on the permeation of polymers can be found in the literature (Extrand 2008; Keller and Kouzes 2017; Kjeldsen 1993; McKeen 2017b; Norton 1957; Pauly 1999; Rogers 1985). Permeation occurs in most materials. For example, although glass is impermeable to liquids and most gases, helium can travel through glass because of its small atomic size (Norton 1953, 1957). Helium also diffuses through many polymer films and this is readily apparent in helium balloons which need to be metalized to hold the helium and still slowly deflate due to the helium diffusing out of the balloons (Mapes, Hseuh, and Jiang 1994; Murray et al. 2016).

Although there are numerous studies in the literature that give permeabilities of various polymers to a number of different gases and vapors, often these are taken with different measuring techniques and the results are published with different units, making a comparison difficult (Huglin and Zakaria 1983; Metz et al. 2005). One study found 29 different units were used to define permeability (Huglin and Zakaria 1983). For gases, the permeation is generally reported in terms of volume flux multiplied by the film thickness divided by the partial pressure difference while for vapors it is generally given in terms of mass flux times the film thickness. Therefore, vapor permeabilities do not always contain a pressure differential, whereas gas permeability measurements do (Massey 2003). In addition, permeability is often described by various terminologies including gas transmission rate, gas permeance, and the gas permeability coefficient. For water vapor, moisture vapor transmission rate, MVTR, is often given for permeability. Many references can be found with tables to allow conversion between the units (Keller and Kouzes 2017; Pauly 1999) and an online calculator (Abbott 2023) has also been published. However, conversions problems arise if no pressure is given or if the conditions of the test, i.e., relative humidity and temperature, are not given.

Therefore, this study was undertaken to summarize the permeability properties of many of the polymer films used in conservation using standard units to enable a direct comparison. Specifically, the permeability of various uncoated single polymer films to oxygen, nitrogen, hydrogen sulfide, and water vapor is summarized. A few polymer films not typically used in conservation are also included for comparison. The intention of this paper is to summarize the permeability of single polymer films used in conservation. With this information, conservators will be able to choose the best polymer film for their specific needs.

2 Encapsulation

In paper conservation, one method used to protect documents from the environment as well as to mechanically support brittle or damaged documents is to encapsulate the document by placing the document in a plastic envelope (“Care, Handling, and Storage of Works on Paper” 2024; Shahani and Wilson 1987; “The Physical Protection of Brittle and Deteriorating Documents by Polyester Encasement” 1975). Encapsulation mechanically stabilizes the document and also protects it from exposure to liquids, fingerprint grease, insects, or other environmental hazards.

Encapsulation is one of the preferred methods used to protect documents since it is easily reversible and does not involve bonding with the material (McGath et al. 2017; Minter 1983; Polyester Film Encapsulation 1980). Other conservation methods such as silking (Marwick 1964), lining (Zihrul 2010), and lamination (Barrow 1939; McGath et al. 2015; Poole 1976) involve chemical and physical attachment of the polymer film to the document. In encapsulation, the two opposing films weakly adhere and this keeps thin or light objects from shifting within the envelope, thereby protecting the document from damage due to friction (Preservation Office Research Services 1980).

Although the purpose of a barrier film is to protect the material from physical and chemical damage from the environment, even in typical environments, materials degrade over time. For example, paper degrades due to oxidation and hydrolysis of the cellulose fibers within the paper as well as possible degradation reactions of the additives and sizing in the paper affecting the cellulose (Hubbe et al. 2017; Łojewska et al. 2005; Shahani and Harrison 2002; Zou, Uesaka, and Gurnagul 1996). The degradation of cellulose leads to the production of volatile acidic compounds such as formic and acetic acid which can further catalyze degradation reactions within the document (Fenech et al. 2010; Jablonsky et al. 2012; Olivier et al. 2009; Shahani and Harrison 2002; Smedemark et al. 2020; Tétreault et al. 2013). Because of these reactions, it is desirable to remove or isolate volatile degradation products from the environment of the paper. However, if the paper is encapsulated, the gases formed during cellulose degradation might become trapped within the encapsulate, causing further damage to the paper. The degradation of the cellulose fibers leads to the paper becoming brittle and is one of the leading causes that books are removed from general use in libraries and archives.

Although a variety of polymer films are used by hobbyists and conservators, many are not considered archival and even archival films should not be used with all materials. For example, encapsulation is not recommended for photographs and chalk or charcoal drawings (Reilly et al. 1989). One reason is the electrostatic charge on the film can attract dust which might scratch the material or the friction and electrostatics between the polymer film and the material can lift off flaking or friable media, causing damage to the document (Polyester Film Encapsulation 1980).

The specific film selected to protect a material is dictated by the desired chemical and physical properties of the polymer film. Biaxially oriented poly(ethylene terephthalate) (PET) is the polymer film recommended for encapsulation (Polyester Film

Encapsulation 1980). PET was chosen since it has good clarity, mechanical strength, and is chemically stable. PET is also used as a barrier material to separate two contacting surfaces. This interleaf minimizes transfer of compounds from one surface to another (Tétreault 2017).

Other polymeric materials are also used for storage of paper based materials, including polyethylene and polypropylene bags and sleeves specifically made to hold photographs, baseball cards, and single paper sheets (Reilly et al. 1989). Various other polymer films that are or have been used in conservation include cellulose based polymers: cellophane, nitrocellulose, cellulose acetate, glassine, and ethyl cellulose and oil-based polymers: nylon and polytetrafluorethylene (Teflon®). In particular, glassine is often used for the short-term storage of stamps and negatives. Nylon Film (Nylon 6/6) is used as a wrap for art, a transparent dust cover, and lining for paintings ("Masterpak." n.d.). Note that some of these polymer films are not considered archival, but nonetheless are sometimes used as short-term storage materials for paper-based materials.

In determining the archival nature of a polymer film, the mechanical, optical, aging, and off-gassing properties of the polymer films are carefully considered (Reilly et al. 1989). However, because the film may also be used as a gas barrier to protect the document from the environment, the ability of the film to limit or allow the transport of gases and vapors needs to be considered. It should be noted that Cameo, The Conservation and Art Materials Online Encyclopedia lists the oxygen and water vapor transmission of many polymer materials, highlighting the need of this data for conservators ("The Conservation and Art Materials Encyclopedia Online (Cameo)" 2024).

3 Permeation

Permeation is often described as a three-step process: adsorption of gas or vapor on one side of the film, diffusion of the gas or vapor through the film, and evaporation of the gas or vapor on the other side of the film (Doty, Aiken, and Mark 1946). Generally, diffusion is the slowest, rate-limiting step. In order for gas or vapor molecules to diffuse through the polymer film, there must be some internal empty spaces, also called polymer free volume, for the gas or vapor molecules to move into. The empty spaces that are large enough for a gas or vapor molecule only occur in the amorphous part of the polymer film. It is easier for the gas or vapor molecules to fit into larger spaces, and therefore the diffusion will depend on the structure and available free volume of the polymer film (McGonigle et al. 2002). At temperatures above the glass transition temperature, T_g, these empty spaces within the polymer are generally larger and transient as the polymer chains have mobility (Michaels, Vieth, and Barrie

1963; Sha and Harrison 1992; Stannett 1978). The diffusion will act in the direction from the higher partial pressure to the lower partial pressure for each specific gas.

The barrier properties of polymer films are dependent on the composition of the film (polymer type, presence of plasticizers and additives) as well as the processing conditions such as cooling rate and amount of drawing, that is, whether or not the film is cast or stretched and oriented, as this affects the crystallinity and structure of the film. Cast films generally have a greater permeability than drawn films since drawing generally increases the crystallinity of the film (McGonigle et al. 2004; Pauly 1999). Besides permeability, processing conditions can alter the crystallinity and density which affects tensile strength and clarity. In addition, permeability may also be dependent on the relative humidity (RH), especially for hydrophilic polymers.

The permeability is also dependent on the molecular size and polarity of the diffusing gas or vapor molecule. The kinetic diameter, σ, is an estimate of the effective size of one diffusing molecule. The smaller the kinetic diameter, the easier and faster the gas or vapor can diffuse through the material since it can fit into the free volume within the polymer film (Stannett 1968). The kinetic diameters, σ, of the diffusing gases studied are given in Table 1. Specifically, for H_2O, O_2, H_2S, and N_2, the size of the gas or vapor molecule varies according to:

$$\sigma_{H_2O} < \sigma_{O_2} < \sigma_{H_2S} \approx \sigma_{N_2}$$

Gases such as O_2 and N_2 are considered to be non-reactive gases, will generally not react with the polymer and will generally diffuse as a single molecule. Reactive gases and vapors such as H_2O, H_2S, and acetic acid are polar, have high chemical activity, and have large absorption or solubility within polar polymers.

The transport of gases or vapors through a polymer film can be described as the amount (mass) of the gas diffusing per area per time and can be mathematically described as a mass flux J_M according to (Stannett and Szwarc 1955; Waack et al. 1955):

$$J_M = \frac{(\text{mass of permeant})}{(\text{area}) \times (\text{time})} = \frac{1}{A}\frac{dM}{dt} \tag{1}$$

where M is the mass of the diffusing gas or vapor, A is the area of the film where permeation occurs, and t is the time.

The flux is also dependent upon the film thickness and the force driving the gas or vapor through the film, i.e. the difference in partial pressure of the diffusing vapor or gas. These variables combine to describe the permeability coefficient P given by (Stannett and Szwarc 1955):

$$P = J_M \frac{x}{(P_2 - P_1)} = \text{DS} \tag{2}$$

where x is the film thickness, P_2 and P_1 are the relative partial pressures on either side of the polymer film, D is the diffusion coefficient, and S is the solubility coefficient. A larger permeability coefficient means that more gas is transported through the film. It should be noted that permeation can occur in either direction, either into or out of an encapsulated material. The driving force ($P_2 - P_1$) in equation (2) will dictate the direction of the flow. As shown in equation (2), the permeability coefficient can be described in terms of a mass flux, J_M, or in terms of the product of the diffusion and solubility. Since smaller molecules diffuse faster than larger molecules, the permeation is expected to decrease as the molecular size of the diffusant increases. Equation (2) shows that the permeability coefficient P also depends on the solubility, S. Since the polar gases and vapors such as H_2S and H_2O are expected to be more soluble in polar, hydrophilic polymers, it is expected that the permeability will be larger for these two molecules in hydrophilic polymers. Therefore, the permeation depends on both the diffusivity and the solubility (Mulder 1996) of the diffusing molecule in the polymer. Generally, the diffusion coefficient is assumed to be constant with concentration of the diffusant. However, the value of the diffusion coefficient for water vapor is known to be concentration dependent for polar polymers, so sometimes the value is extrapolated to 0 concentration when the diffusion constant or permeability is reported (Rouse 1940). The gases and vapors with higher solubility might also swell the polymer or act as a plasticizer and decrease the T_g of the polymer, again increasing the permeability.

4 Values of the Permeability Taken from the Literature

The permeability coefficients P for a variety of uncoated single polymer films are given in Table 1. The gases diffusing include O_2, N_2, and H_2S. A range of permeability values are given for each polymer as found in the literature. This variation in reported permeability values is not surprising as the physical properties of the films, e.g., crystallinity, amount of drawing, amounts of plasticizers, and humidity, were likely different. Therefore, the values should be considered to be approximate. In addition, the permeability may be concentration dependent which is not always stated in the literature and polar vapors may aggregate, i.e., forming dimers, slowing the diffusion and permeation which would lead to different permeability values. Although Mylar® is a trade name for Dupont poly(ethylene terephthalate), since the literature values specifically mention Mylar®, it is listed separately from poly(ethylene terephthalate).

Table 1: The permeability coefficient *P* of single polymer films often found in conservation practices with O_2, N_2, and H_2S as the diffusing gases. The temperature of the permeation was generally 22–30 °C. The kinetic diameter (σ) of the diffusing species and the T_g values were taken from published values (Hiemenz and Lodge 2007; Keller and Kouzes 2017).

Polymer with most common trade name, and/or abbreviation, and T_g	Permeability coefficient ($\times 10^{-13}$ cm^3 (STP) cm/cm^2/Pa/s)		
	O_2 (σ = 0.346 nm)	N_2 (σ = 0.364 nm)	H_2S (σ = 0.360 nm)
Poly(ethylene terephthalate) PET (Tg = 70 °C)	0.013–0.035 (Bhadha 1999; Gilbert 2017; Massey 2003; McGonigle et al. 2004; Pauly 1999)	0.002–0.038 (Gilbert 2017; Massey 2003; Pauly 1999)	0.0435 (Kjeldsen 1993)
Poly(ethylene terephthalate) Mylar®	0.023–0.03 (Kjeldsen 1993; Massey 2003; Pauly 1999)	0.0038–0.0045 (Pauly 1999; Massey 2003)	0.044 (Kjeldsen 1993)
Poly(ethylene terephthalate) Hostaphan®	0.014 (Pauly 1999)	0.0034 (Pauly 1999)	0.014 (Pauly 1999)
Poly(ethylene terephthalate) glycol PET G	0.11 (Massey 2003)	0.045 (Massey 2003)	
Cellulose hydrate Cellophane	0.0016–0.009 (McKeen 2017a; Pauly 1999)	0.0024–0.0140 (McKeen 2017a; Pauly 1999)	0.000045–0.0086 (McKeen 2017a; Pauly 1999)
Cellulose nitrate	1.46 (Hsieh 1963; Pauly 1999)	0.087 (Hsieh 1963; Pauly 1999)	
Cellulose acetate (some plasticized) CA (T_g = 207 °C)	0.585–0.98 (Gilbert 2017; McKeen 2017a; Pauly 1999)	0.019–0.21 (Gilbert 2017; McKeen 2017a; Metz et al. 2005; Pauly 1999)	2.63 (Pauly 1999)
Ethyl cellulose (T_g = 130 °C) (Lai, Pitt, and Craig 2010)	11 (Hsieh 1963; Pauly 1999)	3.3 (Hsieh 1963; Metz et al. 2005; Pauly 1999)	
Low density polyethylene LDPE (T_g = −100 °C)	1.2–2.3 (Bhadha 1999; Marais et al. 2002; Massey 2003; Pauly 1999)	0.45–0.9 (Bao 2015; Massey 2003; Pauly 1999)	27 (Pauly 1999)

Table 1: (continued)

Polymer with most common trade name, and/or abbreviation, and T_g	Permeability coefficient ($\times 10^{-13}$ cm^3 (STP) cm/cm^2/Pa/s)		
	O_2 (σ = 0.346 nm)	N_2 (σ = 0.364 nm)	H_2S (σ = 0.360 nm)
High density polyethylene HDPE (T_g = −100 °C)	0.23–0.9 (Bhadha 1999; Gilbert 2017; Massey 2003; Pauly 1999)	0.11–0.27 (Gilbert 2017; Massey 2003; Pauly 1999)	6.5 (Pauly 1999)
Polypropylene PP (T_g = −10 °C)	0.4–4 (Bhadha 1999; Gilbert 2017; Kjeldsen 1993; Massey 2003; Pauly 1999)	0.225–0.33 (Kjeldsen 1993; Metz et al. 2005; Pauly 1999)	2.4 (Pauly 1999)
Polytetrafluoroethylene PTFE (T_g = 130 °C)	2.6–4.1 (Bhadha 1999; Kjeldsen 1993; Massey 2003; Pauly 1999)	1.0 (Massey 2003; Pauly 1999)	
Nylon 6,6 (T_g = 55 °C)	0.0067–0.026 (Kjeldsen 1993; Massey 2003; McKeen 2017b)		
Nylon 6 (T_g = 47 °C)	0.012–0.029 (Gilbert 2017; Kjeldsen 1993; Massey 2003; Pauly 1999)	0.0033–0.075 (Gilbert 2017; Kjeldsen 1993; Massey 2003; Metz et al. 2005; Pauly 1999)	0.26 (Pauly 1999)
Poly(ethyl methacrylate) PEMA (T_g = 65 °C)	0.89 (Pauly 1999)	0.17 (Pauly 1999)	2.88 (Pauly 1999)
Poly(vinyl acetate) PVA (T_g = 30 °C)	0.367 (Pauly 1999)		
Polyvinyl alcohol PVOH (T_g = 90 °C)	0.00005–0.00665 (Massey 2003; Pauly 1999)	0.0001–0.004 (Metz et al. 2005; Pauly 1999)	0.0052 (Pauly 1999)
Poly(vinyl chloride) PVC (T_g = 75 °C)	0.034–0.09 (Gilbert 2017; Pauly 1999; Tikhomirov et al. 1968)	0.009–0.03 (Gilbert 2017; Pauly 1999; Tikhomirov et al. 1968)	0.14 (Pauly 1999)

Table 1: (continued)

Polymer with most common trade name, and/or abbreviation, and T_g	Permeability coefficient ($\times 10^{-13}$ cm^3 (STP) cm/cm^2/Pa/s)		
	O_2 (σ = 0.346 nm)	N_2 (σ = 0.364 nm)	H_2S (σ = 0.360 nm)
Poly(vinylidene dichloride) Saran® PVCl2 (T_g = 15 °C)	0.004 (Gilbert 2017; Pauly 1999)	0.0007 (Gilbert 2017; Pauly 1999)	0.027 (Pauly 1999)
Polycarbonate (Lexan™) PC (T_g = 149 °C)	0.5–1.2 (Extrand 2008; Kjeldsen 1993; Massey 2003)	0.1–0.24 (Kjeldsen 1993; Massey 2003; Metz et al. 2005)	
Poly cis isoprene (natural rubber) PI (T_g = −70 °C)	17.5–17.8 (Bhadha 1999; Extrand 2008; Gilbert 2017; Kjeldsen 1993; Pauly 1999)	6.06–6.52 (Gilbert 2017; Kjeldsen 1993; Massey 2003; Metz et al. 2005)	
Polystyrene PS (T_g = 100 °C)	0.825–2.0 (Extrand 2008; Gilbert 2017 Massey 2003; Pauly 1999;)	0.22–1.9 (Gilbert 2017; Massey 2003; Metz et al. 2005; Pauly 1999)	

The permeability values given in Table 1 show greatest variation for hydrophilic polymers such as nylon and polyvinyl alcohol (PVOH) where the relative humidity might swell the polymer film allowing faster permeation. Because the relative humidity is not always reported, the values for hydrophilic polymers show large variation.

The permeability of the polymer films to O_2 and N_2 gas are shown in Figures 1 and 2. In Figure 1, the data is shown by a bar chart. Note that the permeability values are on a log scale, so the permeability of O_2 in natural rubber is approximately 1,000 times more than the permeability in nylon 6,6. The permeability of O_2 is generally larger than the permeability of N_2 as expected since the O_2 molecule is smaller than the N_2 molecule. A least squares analysis of the average permeability values for each of the films is calculated to be $P_{N2} \approx 0.34\, P_{O2}$, showing that P_{N2} is approximately 1/3 that of P_{O2}. PVOH, PET, Saran®, cellophane, PVC and Nylon 6 have low oxygen and nitrogen permeabilities whereas natural rubber and ethyl cellulose have relatively high oxygen and nitrogen permeabilities. HDPE and cellulose acetate have lower oxygen permeability than PP and LDPE.

Figure 2 is a log-log plot of the average values of O_2 and N_2 permeability of each of the polymer films given in Table 1. The name of the polymer film is positioned over the average value of the permeability of that polymer film. This plot is modeled after Material Property Charts or Ashby plots which aid in material selection (Ashby 2005) and can be used to quickly identify a material suitable for a specific need. The inset in Figure 2 shows a more traditional log-log plot of P_{O2} versus P_{N2}.

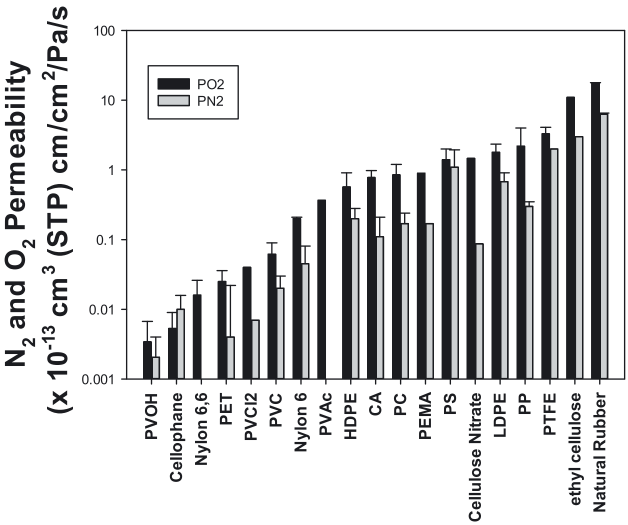

Figure 1: Semi-log plot comparing the permeabilities of O_2 and N_2 for each of the polymer films. The length of the bar represents the average of the permeability values given in Table 1. The black bars represent P_{O2} whereas the gray bars represent P_{N2}. The full polymer names are given in Table 1.

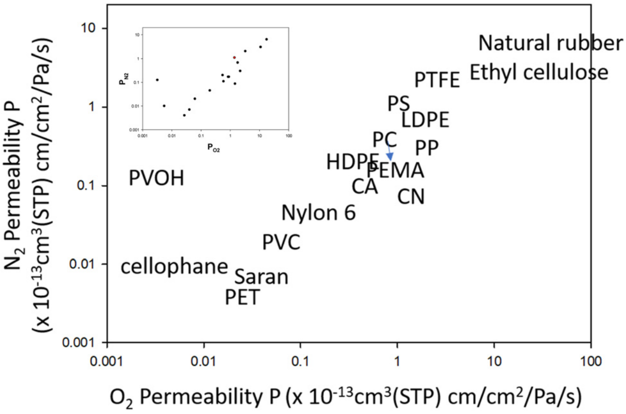

Figure 2: Log–log plot comparing the permeabilities of O_2 and N_2. The polymers listed are centered on the average values of the permeability given in Table 1. The chemical names are listed in Table 1. The inset shows a log–log plot of the average values of the permeabilities of each of the polymer films given in Table 1.

Figures 1 and 2 show that the permeabilities of specific polymer types are not clustered together, i.e., permeabilities of the non-polar hydrophobic polymers and polar hydrophilic polymers are not clustered. This indicates that there is a similar interaction and solubility of the gases with the polymer and the differences in the permeation between specific polymers are due to the specific structure of each of the polymers, notably the free volume, crystallinity, plasticizers, and the T_g.

The water vapor permeabilities and water vapor transmission rates are given in Table 2. As expected based on the smaller size of the water molecule, the permeability of water vapor is much larger than the permeability of N_2 and O_2 gases. For hydrophilic polymers, the higher solubility of water into the polymer also influences the permeability. Hydrophilic polymers such as PVOH also show large variation in permeability values. Most likely, this is due to differences in the relative humidity which is not reported. Because of this large variation, two ranges of the permeability of PVOH are given reflecting the different numbers reported.

Plots of oxygen, hydrogen sulfide, and water vapor permeabilities are shown in Figures 3, 4, and 5. The more hydrophilic polymers such as cellophane, PVOH, ethyl cellulose, cellulose (glassine), polyvinyl acetate (PVAc), and cellulose acetate (CA) have the highest water vapor permeabilities whereas the hydrophobic polymers such as LPDE, PP, HDPE, and Nylon 6,6 have the lowest water vapor permeabilities. In hydrophilic polymers, the concentration of water in a film is dependent on the

Table 2: Water vapor permeability coefficient *P* and water vapor transmission rate (WVTR) of single polymer films often found in conservation practices. The temperature of the permeation was generally 22–30 °C. The kinetic diameter (σ) of a water molecule is 0.265 nm (Keller and Kouzes 2017).

Polymer with most common chemical and trade name	**Water vapor permeability coefficient ($\times 10^{-13}$ cm^3 (STP) $cm/cm^2/Pa/s$)**	**Water vapor transmission rate WVTR (g mm/day m^2)**
	σ = 0.265 nm	
Poly(ethylene terephthalate) PET	97–398 (Barrie 1986; Bhadha 1999; Gilbert 2017; Hall et al. 2020; Keller and Kouzes 2017; McGath et al. 2017; Pauly 1999; Rogers 1985)	0.21–0.71 (Bhadha 1999; Massey 2003; Tock 1983)
Poly(ethylene terephthalate) Mylar®	98–114 (Hubbell, Brandt, and Munir 1975; Pauly 1999; Rogers 1985)	
Poly(ethylene terephthalate) Hostaphan®	114 (Pauly 1999)	
Poly(ethylene terephthalate) glycol PET G	0.02 (Massey 2003)	1.5–1.6 (Massey 2003)
Cellulose hydrate Cellophane	18,900 (Pauly 1999)	1–19 (Tock 1983)
Cellulose nitrate	4,720 (Hsieh 1963; Pauly 1999)	
Cellulose acetate (some plasticized)	4,125–7,500 (Barrie 1986; Gilbert 2017; Metz et al. 2005; Pauly 1999; Rogers 1985)	4–16 (Tock 1983)
Ethyl cellulose	6,700–15000 (Barrie 1986; Hsieh 1963; Metz et al. 2005; Pauly 1999; Rogers 1985)	1.8–5.6 (Tock 1983)
Low density polyethylene LDPE	51–68 (Bhadha 1999; Keller and Kouzes 2017; Marais et al. 2002; Pauly 1999; Rogers 1985)	0.11–2.5 (Bhadha 1999; Tock 1983; Massey 2003)

Table 2: (continued)

Polymer with most common chemical and trade name	Water vapor permeability coefficient ($\times 10^{-13}$ cm^3 (STP) cm/cm^2/Pa/s)	Water vapor transmission rate WVTR (g mm/day m^2)
	$\sigma = 0.265$ nm	
High density polyethylene HDPE	6.8–9.8 (Bhadha 1999; Gilbert 2017; Keller and Kouzes 2017; Metz et al. 2005; Pauly 1999; Rogers 1985)	0.12–0.2 (Massey 2003; Tock 1983)
Polypropylene PP	18–51 (Bhadha 1999; Gilbert 2017; Keller and Kouzes 2017; Metz et al. 2005; Pauly 1999; Rogers 1985; Togashi and Hara 2011)	0.06–1.2 (Bhadha 1999; Massey 2003; Tock 1983)
Glassine (wood pulp)	9,627–17,880 (Carson 1937)	
Polytetrafluoro-ethylene PTFE	2.25–6 (Bhadha 1999; Keller and Kouzes 2017; Pauly 1999; Rogers 1985)	0.0425–0.008 (Bhadha 1999; Massey 2003)
Nylon 6,6	510 (Rogers 1985)	1.1–7.4 (Massey 2003)
Nylon 6	133–525 (Barrie 1986; Gilbert 2017; Metz et al. 2005; Myers et al. 1960)	0.8–16 (Massey 2003; Tock 1983)
Poly(ethyl methacrylate) PEMA	2,380 (Pauly 1999)	
Poly(vinyl acetate) PVA	4,125 Barrie (1986)	
Polyvinyl alcohol PVOH	5.25–14.3 or 6,800–7,000 (Barrie 1986; Metz et al. 2005; Pauly 1999) or (de Andrade et al. 2022; Xianda, Anlai, and Suqin 1987)	590–280,000 (Tock 1983)

Table 2: (continued)

Polymer with most common chemical and trade name	**Water vapor permeability coefficient ($\times 10^{-13}$ cm^3 (STP) $cm/cm^2/Pa/s$)**	**Water vapor transmission rate WVTR (g mm/day m^2)**
	***σ* = 0.265 nm**	
Poly(vinyl chloride) PVC	46–210 (Barrie 1986; Doty, Aiken, and Mark 1946; Gilbert 2017; Metz et al. 2005; Pauly 1999; Rogers 1985; Tikhomirov et al. 1968)	0.45–1.2 (Bhadha 1999; Massey 2003; Tock 1983)
Poly(vinylidene dichloride) Saran® PVCl2	1.1–7.5 (Gilbert 2017; Pauly 1999; Rogers 1985)	0.01–0.14 (Massey 2003; Tock 1983)
Polycarbonate (Lexan™) PC	825–1,050 (Barrie 1986; Metz et al. 2005; Moon and Extrand 2009; Pauly 1999)	1.5–4 (Massey 2003; Tock 1983)
Poly cis isoprene (Natural rubber) PI	1718–2025 (Barrie 1986; Bhadha 1999; Metz et al. 2005)	3.76 (Bhadha 1999)
Polystyrene PS	728–840 (Barrie 1986; Doty, Aiken, and Mark 1946; Metz et al. 2005; Pauly 1999)	0.8–3.9 (Massey 2003; Tock 1983)

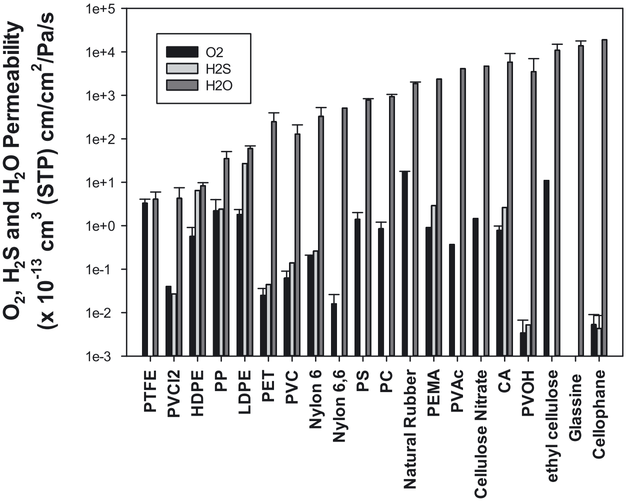

Figure 3: Semi-log plot comparing the permeabilities of O_2, H_2S and H_2O. The length of the bar represents the average of the permeability values given in Tables 1 and 2. The black bar, light gray bar and dark gray bar represent P_{O2}, P_{H2S}, and P_{H2O}, respectively. The full polymer names are given in Table 2.

relative humidity, RH. A higher RH drives more water into the films, increasing the water vapor solubility S in hydrophilic polymers. As shown in equation (2), if the solubility S is greater and the diffusion constant D is constant, then the permeability must increase. In addition, the absorbed water can swell the polymer which

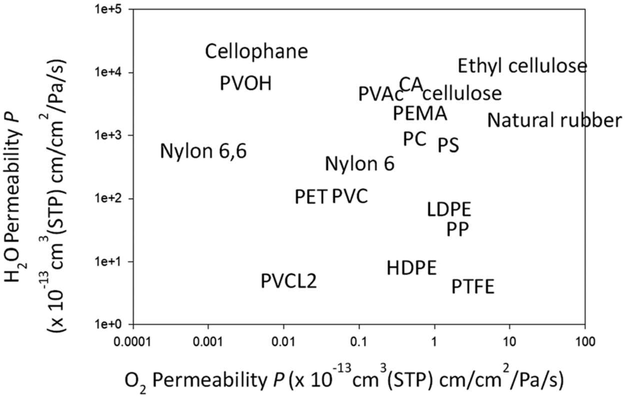

Figure 4: Log–log plot comparing the permeabilities of O_2 and H_2O. The polymers listed are centered on the average values of the permeability given in Table 2. The full polymer names are given in Table 2.

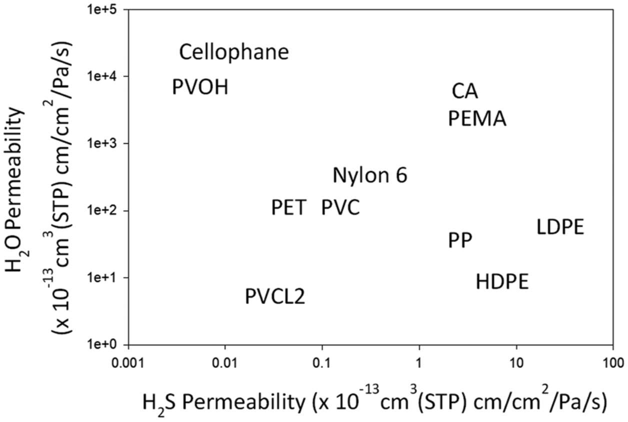

Figure 5: Log–log plot comparing the permeabilities of H_2O and H_2S. The polymers listed are centered on the average values of the permeability given in Table 1. The full polymer names are given in Table 1.

increases the diffusion coefficient (Hubbell, Brandt, and Munir 1975). Therefore, the analysis of the permeation of water vapor in hydrophilic films is problematic as highlighted by the large variation in permeability values.

Plots of the permeabilities of H_2O versus O_2 and H_2O versus H_2S are given in Figures 4 and 5. The plots look similar, except that the H_2S permeability is greater than the permeability of O_2. Since the size of the H_2S molecule is larger than the size of the O_2 molecule, this is a bit surprising. However, since the H_2S is considered a reactive gas and it is polar, it likely has a higher solubility within the polymer film giving a higher permeability than expected. The water vapor permeability is generally much greater than the H_2S permeability, as expected since the water vapor molecule is much smaller than the H_2S molecule. As shown in Figures 4 and 5, the non-polar polymer films, HDPE, LDPE, PTFE, PP, show low water vapor permeability and high H_2S and O_2 permeability. For the hydrophilic polymers, the H_2O permeability is relatively large but the O_2 and H_2S permeability does not show a strong dependence with the type of polymer.

Other vapors such as acetic and formic acid are formed during cellulose degradation. If the paper is encapsulated, these might become trapped inside the encapsulate further degrading the paper. Therefore, the permeation of volatile organic compounds such as acetic acid and formic acid should be included in this summary. Unfortunately, data of the permeability of acetic acid through single polymer films is limited. However, a few references could be found which give not only the permeability at one temperature, but at multiple temperatures. The

Table 3: Vapor transmission rate (TR) of acetic acid vapor through various polymer films. The permeability increases with temperature. The kinetic diameter of an acetic acid molecule is 0.436 nm (Bowen, Noble, and Falconer 2004). Chemical names for the polymer films are given in Tables 1 and 2. Note that nylon is a polyamide, but the exact type was not given in the paper.

Polymer film trade name	**TR (g mm/m^2 d) $T \approx 21$ °C**	**TR (g mm/m^2 d) $T \approx 54$ °C**	**TR (g mm/m^2 d) $T \approx 74$ °C**
LDPE (Rogers 1985)	1.22	25.9	119
HDPE (Rogers 1985)	0.39 (T = 22.8 °C)	2.36 (T = 49 °C)	
PET (McGuiggan et al. 2023; Oreski et al. 2017)	<0.026	0.29 (T = 65 °C)	7.5 (T = 85 °C)
Polyamide (Oreski et al. 2017)	20	173 (T = 65 °C)	381 (T = 85 °C)

transmission rate of acetic acid vapor in four polymer films is given in Table 3. As a comparison, the Water Vapor Transmission Rate for PET as given in Table 2 is WVTR ≈ 0.5 (Bhadha 1999; Massey 2003; Tock 1983), which is approximately 20 times greater than the transmission rate of acetic acid vapor. This is expected since the kinetic diameters of the acetic acid and water molecules are 0.436 nm and 0.295 nm, respectively (Bowen, Noble, and Falconer 2004). The transmission rate of O_2, N_2, H_2O, and acetic acid through LDPE and HDPE are similar, reflecting a similar diffusant interaction.

The transmission rate of acetic acid vapor is much lower for PET films than for either polyamide or polyethylene films. Specifically, the transmission rate of acetic acid vapor through PET is 50 times slower than through low density polyethylene. Therefore, if trapping of acetic acid is a concern for encapsulation due to aging and off-gassing of materials, switching to polyamide or polyethylene would increase the gas flow out of the enclosure.

The values in Tables 1 and 2 are measured at approximately 20 °C. The permeability can increase significantly with temperature since the permeability increases approximately 5 % per °C rise in temperature (Doty, Aiken, and Mark 1946; Keller and Kouzes 2017; Tock 1983). As shown in Table 3, the transmission rate increases as least 10 times as the temperature increases from 21 to 74 °C. This increased transmission rate has an impact when evaluating the results of artificial aging of encapsulated materials. Since artificial aging is done at elevated temperatures, gases and vapors will be transported through the films much faster at higher temperatures than if they were done at room temperature. Therefore, artificial aging of encapsulated materials performed at elevated temperatures will not reflect the true aging properties of the document within the encapsulate.

5 Discussion

Tables 1, 2, and 3 show a wide range of permeability coefficients and transmission rates between different polymer films and a wide range of values for the same film but for different gases and vapors. As previously stated, the permeability depends on the type of polymer, specific crystallinity, and additives present in the polymer film as well as the testing conditions such as temperature, humidity, and diffusant size, polarity, and concentration. In addition, there are a variety of methods used to measure the permeability coefficient (Giacinti Baschetti and Minelli 2020). Therefore, Tables 1, 2 and 3 show a range of reported values, highlighting the variation in the films measured, measuring conditions, and reported measurements. In spite of the differences, generalizations about the data can be made.

Comparing the permeability of the non-reactive gases O_2 and N_2, Table 1 and Figures 1 and 2 show that the permeability of N_2 is slightly smaller than the permeability of O_2: P_{N2} is approximately 1/3 that of P_{O2} and this ratio holds consistently for all the polymers investigated. It is expected that the permeability of N_2 is less than the permeability of O_2 since O_2 has a smaller molecular size than N_2. Besides the small difference in the permeability due to the molecular size, it is likely that the O_2 and N_2 molecules each interact with the polymers via van der Waals interactions. Since there is little difference between the interaction of O_2 and N_2, the permeability is likely determined by the free volume in the polymer.

The permeation properties of non-polar hydrocarbon polymers such as LDPE, HDPE, and PP to water vapor is small. Hydrophilic polymer films have much higher water vapor permeability than the hydrophobic polymers. In addition, the permeation of hydrophilic polymers such as nylon, cellulose, and polyvinyl alcohol are highly dependent on the humidity; the permeation of water vapor increases dramatically with concentration of water in the film. The higher solubility of water vapor in the film increases the amount of water molecules diffusing through the film, as given in equation (2). The absorbed water vapor can swell and plasticize the polymer, allowing faster permeation (Hubbell, Brandt, and Munir 1975). Therefore, the analysis of the permeation of water vapor in hydrophilic films is problematic. Because of this, the relative humidity should be noted when the permeability is tested.

Looking at the data of acetic acid vapor shown in Table 3, the permeability of polyamide and polyethylene films is much greater than the permeability of PET films. Therefore, polyamide and PE films might be considered as alternates to PET films if acetic acid transport is to be maximized.

The ratio of the permeability of two specific gases for a single polymer film is known as the selectivity of the film. For example, the ratio of the permeability of water and nitrogen in cellulose acetate (P_{H2O}/P_{N2}) is 24,000 (Metz et al. 2005). This means that when a mixture of nitrogen and water vapor is on one side of a cellulose acetate film, water permeates through cellulose acetate much faster than nitrogen, effectively giving the ability of the polymer film to selectively separate the gases. Conversely, the ratio of the permeabilities in polyethylene is not as selective since $P_{H2O}/P_{N2} = 6$ (Metz et al. 2005).

Besides single polymer films, countless other films are used by conservators. Some of these include non-woven spunbound materials such as Hollytex® (a non-woven spunbound polyester fabric) and Tyvek® (spunbound HDPE). Because the pore size of a spunbound material is much larger than the small free volume regions within single cast or oriented polymer films, the permeability of spunbound films is much larger than the permeability of cast or oriented polymer films. Goretex®, while not spunbound, contains sub-microscopic pores due to a stretching process and readily allows vapors and gases to flow through.

Polymer films can also be made by mixing two polymers thereby making a polymer blend or by attaching two polymers on the same polymer chain, thereby making a co-polymer. Examples of films include isobutyl methacrylate polymer (Acryloid B-67) and poly(ethylene-co-vinyl acetate) (EVA) and BEVA® 371 which is composed mainly of ethylene vinyl acetate. The permeability of the films will depend on the ratio of each monomer (Marais et al. 2002) and, for blends, it will depend on the phase behavior, i.e., whether they phase separate or are miscible (Kamal, Jinnah, and Utracki 1984; Shirvani et al. 2019). Polymer blending is often used to tune the selectivity of the film.

Single polymer films can be further modified by laminating two films together, i.e., having a double layer of two different polymer films, or the film can be coated or metalized. Laminated vapor barrier films used in conservation have been previously described (Burke 1992). Single-layer films are permeable to most simple gases to some extent, so added layers or coatings will decrease the transport of gases and vapors and therefore improve the barrier strength. Beva Tex® is Hollytex® coated with ethylene vinyl acetate, and Marvelseal®, an aluminized heat sealable polyethylene and nylon barrier film. Conservators use a variety of films to limit oxygen transmission. Escal Neo® and TECHBARRIER® are polymer laminates containing a layer of a silica containing polymer to give high oxygen and moisture barrier films. For example, Escal Neo® is coated with a ceramic polyvinyl alcohol film (polypropylene/silica deposited PVOH/LDPE).

To obtain a material which limits the diffusion of gases, the film should have the following properties:

(a) high glass transition temperature (T_g) leading to low polymer segment mobility, less voids, and a more tortuous path for the gas moving through the polymer

(b) higher film crystallinity since diffusion only occurs in the amorphous parts of the polymer film
(c) inertness with the diffusant or environment, such as the RH. Moisture absorbed by the polymer has a plasticizing effect and can lower the T_g of the polymer and thus increase the permeability (Robertson 1993)
(d) small polymer free volume

In addition, the diffusant can swell the polymer, leading to a larger solubility and larger permeability polymer. If high permeability is desired, then the opposite properties are desired.

The goal of this work was to review the permeation of some single polymer films commonly used in conservation and compile the permeability using standard units so that the permeability can be directly compared. Using the data in Tables 1, 2, and 3, the polymer can be specifically chosen according to the need. The polymers given in Tables 1, 2 and 3 show a wide range of values for the different gases and vapors. It is important to note that there is not one best polymer for all applications. It will depend on the specific use and environmental conditions. Since the gases or vapors can permeate in either direction, the question to ask is what is the important gas in the system that needs to be limited? If the environment has a large concentration of a certain gas, such as H_2S, then the polymer film should be chosen so as to minimize the permeation of H_2S from the environment. However, if degradation products of the material need to be removed from the environment of the artifact, then high permeability of gases and vapors is desired. Smaller molecules generally give the larges permeability according to $P_{\mathrm{H2O}} > P_{\mathrm{H2S}} > P_{\mathrm{O2}} > P_{\mathrm{N2}} > P_{\mathrm{acetic\ acid}}$. Specifically, for encapsulation, polyamide and LDPE have a greater permeability to acetic acid vapor than PET. Therefore, if acetic acid transport is desired, polyamide and LDPE appear to be viable candidates based on permeability. Other properties such as mechanical strength, clarity, presence of plasticizers, and off-gassing and stability need to be evaluated to determine if these materials can be used as an alternative to PET.

6 Conclusions

Single polymer films used in conservation are breathable materials and allow the flow of vapors and gases through them. This review summarizes the permeability of single polymer films used in book and paper conservation using standard units for easy comparison. The polymer films chosen to study represent single polymer films generally used for storage materials and processes such as encapsulation. The permeability was found to differ by over 7 orders of magnitude when comparing different gases and vapors diffusing through the polymer films. The permeability is dependent on the molecular size σ of the diffusing gas, with $\sigma_{H_2O} < \sigma_{O_2} < \sigma_{H_2S} \approx \sigma_{N_2} < \sigma_{\text{acetic acid}}$. The smaller molecules diffuse faster than the larger molecules. In addition, the chemistry and structure of the polymer film influence the permeability. Hydrophilic polymers have a much greater permeability of polar diffusants such as water vapor than hydrophobic polymers. The commonly used polymer, poly(ethylene terephthalate) PET, shows a much slower transmission of acetic acid than polyethylene. In addition, the permeation increases with temperature. The review provides data of the permeability properties of the films used by conservators and will enable them to make an informed decision of the best material for their application.

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