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Water Vapor Permeation in Plastics

Paul E. Keller
Richard Kouzes

January 2017
Revision 1

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Executive Summary

Polyvinyl toluene (PVT) and polystyrene (PS) (referred to as “plastic scintillator”) are used for gamma ray detectors. A significant change in radiation detection performance has been observed in some plastic scintillator-based gamma-ray detectors in systems in outdoor environments as they age. Recent studies have revealed that plastic scintillator can undergo an environmentally related temporary or permanent material degradation that adversely affects gamma ray detection performance under certain conditions and histories.

The degradation of plastic scintillator over time is due to a variety of factors, and the term “aging” is used to encompass all factors. Some plastic scintillator samples show no aging effects (no significant change in sensitivity over more than 10 years), while others show severe aging (significant change in less than 5 years). Aging effects arise from weather (variations in heat and humidity), chemical exposure, mechanical stress, light exposure, and loss of volatile components. The damage produced by these various causes can be cumulative, causing observable damage to increase over time. Damage may be reversible up to some point, but becomes permanent under some conditions.

The objective of this report is to document the phenomenon of permeability of plastic scintillator to water vapor and to derive the relationship between time, temperature, humidity and degree of water penetration in plastic. An overview of diffusion and the properties of materials is also presented. Several conclusions are documented about the properties of water permeability of plastic scintillator.

Acronyms and Abbreviations

AAT	Accelerated Aging Test
C	Concentration
D	diffusion coefficient
HDPE	high-density polyethylene
J	diffusion flux
LDPE	low-density polyethylene
MVTR	moisture vapor transmission rate
PNNL	Pacific Northwest National Laboratory
P	permeability
PS	polystyrene
PVT	polyvinyl toluene
S	solubility
STP	standard temperature and pressure (exactly 273.15 K and 100 kPa)
VTR	vapor transmission rate
WVTR	water vapor transmission rate

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1. Introduction

Polyvinyl toluene (PVT) and polystyrene (PS) (referred to as "plastic scintillator") are used for gamma ray detectors. A significant change in radiation detection performance has been observed in some PVT-based gamma-ray detectors in systems as they age. The degradation of sensitivity of PVT over time is due to a variety of factors, and the term "aging" is used to encompass all factors. Some plastic scintillator samples show no aging effects (no significant change in sensitivity over more than 10 years) while others show severe aging (significant change in sensitivity in less than 5 years). Aging effects arise from weather (variations in heat and humidity), chemical exposure, mechanical stress, light exposure, and loss of volatile components. The damage produced by these various causes can be cumulative, causing observable damage to increase over time.

The degradation of PVT and PS in cold environments has been observed to be associated with internal "fogging" of the plastic. The photos in Figure 1.1 show a laboratory-induced example of a fogging phenomenon in a slab of PVT plastic (left), which fills the interior, compared to a clear piece of PVT (right). Pacific Northwest National Laboratory (PNNL) has made several studies of the fogging phenomenon [Cameron 2014; Fritz 2014; Kouzes 2015]. The conclusion reached was that water plays the key role in causing the observed fogging in PVT. Fogging may be temporary or permanent, where micro-cracks are produced inside the material. There may also be other phenomena at work in addition to this one in inducing the observed fogging, but water plays a dominant role. Studies at PNNL have found that only certain locations have the weather conditions necessary to cause the observed fogging, and the effect has been found in some studies to only occur after several years of exposure, apparently due to the slow permeation of water into the plastic.

Figure 1.1. Internal fogging of PVT (left) after rapid aging compared to unexposed PVT (right).

The objective of this report is to document the phenomenon of permeability of plastic to water vapor and to derive the relationship between time, temperature, humidity and degree of water penetration in plastic. The general assumption made is that these are idealized materials so that models like ideal gas behaviour can be assumed.

The first several Sections of this report provide information on theoretical aspects of diffusion, explanations of terminology and units, as well as information on material properties. The later Sections provide numerical results derived from measurements and results of the theoretical framework of diffusion.

1.1 Unit Conversions and Consistency

The two major issues in the field of liquid, gas and vapor diffusion, permeability, and transport within materials are the variation in terms used and the mixture of units often combining various metric, Système international (SI), and United States customary system (USCS) standard units in the same measurement. Numerous units are used to express permeability leading to conversion issues [Yasuda 1975]. This can lead to confusion in terms when comparing one published result to another. Another source of confusion is the term permeability. For gases, such as oxygen, permeability is given in units of volume of gas while for water vapor it is given in units of mass of water vapor. Sometimes, the amount of permeant is given in moles. Sometimes the term "diffusion coefficient" is used incorrectly instead of permeability, which are different properties, leading to additional confusion. Another complication involves the water vapor transmission rate (WVTR). It is given in units of mass per area per time with no reference to the pressure differential across the material, or often the temperature of measurement. In this report, these mostly historic issues are addressed, and literature values were converted and compiled into self-consistent tables using modern units. Unit conversions are also supplied to aid the reader.

1.2 Polymers

Table A.1 provides details on the properties of PVT. The tables in this report provide numbers and ranges for a variety of polymers. Table A.2 provides a list of polymers encountered in this report with IUPAC names, abbreviations, and common product names.

2. Fickian Diffusion

Frickian diffusion describes an idealized system, and while Frick's law describes most diffusion, there are some non-Frickian materials. Fick's first law states that under steady state the diffusion flux, J (also represented by some a F or q), is proportional to the product of the *diffusion coefficient*, D, and the concentration gradient, dC/dx, also known as *solubility*, S [Fick 1855a; Fick 1855b; Fick 1995; Lahtinen]. This is written as

$$J = -D\frac{dC}{dx} = DS\frac{dp}{dx} \qquad \text{(Eq. 2.1)}$$

Here, J is the rate of transfer per unit area, and p is the pressure. The diffusion coefficient, also known as diffusivity, is a measure of how much liquid, gas, or vapor (the permeant) can penetrate an area of a material in a given time period, so it is in units of area per time. It is often reported in units of $\text{cm}^{2}\cdot\text{sec}^{-1}$. In the steady state, the concentration gradient can be estimated by dividing the change in concentration of the liquid, gas, or vapor across a material by the path length through the material. For example, Figure 2.1 shows the change of concentration across a material in one dimension. In steady state, the concentration (C) gradient for that example between the backside and front side is given by

$$\frac{dC}{dx} = \frac{C_{BS}-C_{FS}}{x_0-x_1} \qquad \text{(Eq. 2.2)}$$

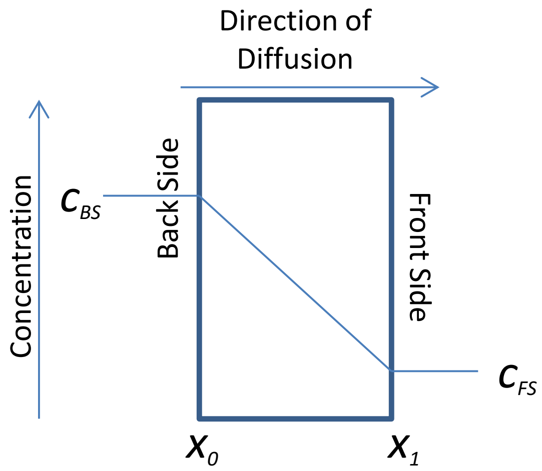

Figure 2.1. Change in concentration of a liquid, gas, or vapor diffusing through a material of thickness ($x_1 - x_0$) in the steady state.

Fick's second law describes normal diffusion across a membrane or barrier in one dimension in terms of the concentration of permeant as a function of time and distance. In one dimension, it is written as

$$D\frac{d^2C}{dx^2} = \frac{dC}{dt}. \qquad \text{(Eq. 2.3)}$$

Diffusivity depends on the polymer, the diffusing permeant, especially its molecular size, temperature, pressure, and in some cases, on the concentration. It is a measure of the diffusion mobility. It has units of

$$D(units) = \frac{area}{time}, \qquad \text{(Eq. 2.4)}$$

and common units are $\text{m}^2\cdot\text{s}^{-1}$ and $\text{cm}^2\cdot\text{s}^{-1}$.

Diffusivity is temperature dependent and the relationship to temperature is given by the Arrhenius [1889a, 1889b] equation as:

$$D = D_0 e^{-E_A/RT}, \qquad \text{(Eq. 2.5)}$$

where E_A is the activation energy for diffusion and D_0 is the maximal diffusion coefficient at infinite temperature. This Arrhenius equation is a formula expressing the temperature dependence of reaction rates [Arrhenius 1889a, 1889b]. Diffusion is dependent on a material's structure. The activation energy of a polymer in the rubber state is higher than in the glassy state (PVT is a glassy polymer). In the rubber state, the permeant must create channels and active chains to permeate the polymer. In the glassy state, where the temperature is above the glass temperature (T_g) of the polymer, the permeant moves through holes via local activation. This results in different diffusion coefficients between the rubber state and the glassy state. Some diffusion values are listed in Table 2.1. For oxygen permeating a polymer above the glass temperature, the permeability increases approximately 10%/°C [DeLassus 2002, pg. 392].

Another thing that affects diffusivity is the apparent diameter of the permeant as that determines how a permeant can travel through the material. Kinetic diameter, *d*, is an estimate of the effective target size of one particle at the atomic or molecule scale as it relates to the likelihood that it will collide with another particle. It is not the same as atomic diameter, but relates to the size of the sphere surround the molecule that leads to a scattering event [Joos 1958]. The smaller the kinetic diameter, the easier the permeant can penetrate the material. Table 2.2 lists some kinetic diameters, and Figure 2.2 gives a plot of some kinetic diameters as a function of molecular weight. Water is the permeant of importance in this report.

Table 2.1. Diffusion coefficients for water and oxygen for various polymers from the literature.

Polymer	D_{H2O} (cm^2·s^{-1}) @30°C	D_{O2} (cm^2·s^{-1}) @25°C	References
Lower Bound	7.2x10^{-10}	7.2x10^{-10}	
Ethylene Vinyl Alcohol (EVOH)		7.2x10^{-10}	[DeLassus 2000, p.282]
Polyamide (PA6) "Nylon"	7.2x10^{-10}	4.72x10^{-9}	[Hunt 1980; Kjeldsen 1993, p.128]
Polyethylene terephthalate (PET) "Dacron"		2.7x10^{-9}	[DeLassus 2000, p.282]
Vinylidene Chloride Copolymer		1.01x10^{-9}	[DeLassus 2000, p.282]
Acrylonitrile		1.0x10^{-9}	[DeLassus 2000, p.282]
Polyurethane (200 µm film)	3.2x10^{-10}@20°C		[Rezaei 2010]
Low-Density Polyethylene (LDPE)	1.9x10^{-9}	1.12x10^{-7}	[Kjeldsen 1993, p.128; DeLassus 2000, p. 282]
Polyvinylidene Chloride (PVDC) "Saran"	9.1x10^{-8}		[Seethamraju 2014]
Polycarbonate (PC) "Lexan"	6.5x10^{-8} @23°C		[Robeson 1983]
Polystyrene (PS)	2.2x10^{-8}	3.1x10^{-7}	[Feng 2015]
Polypropylene (PP)	1.8x10^{-8}	3.76x10^{-7}	[Kjeldsen 1993, p.128]
Polyethylene (PE)	1.3x10^{-8}	1.0x10^{-6}	[Evans 1978]
High-Density Polyethylene (HDPE)	2.6x10^{-7}	1.6x10^{-7}	[DeLassus 2000, p.282]
Polyvinyl Alcohol (PVA)	1.9x10^{-7}		[Seethamraju 2014]
Epoxy Resin	1.7x10^{-7}		[Krongauz 1990]
Polyvinyl Acetate (PVAc)	1.5x10^{-7}		[Seethamraju 2014]
Polyvinyl Toluene (PVT)	1.3x10^{-7} @23°C	6.8x10^{-7}	[Krongauz 1990; Feng 2015]
Polyvinyl Butyral (PVB)	4.0x10^{-6}		[Seethamraju 2014]
Poly Methyl Methacrylate (PMMA)	2.9x10^{-6}	2.49x10^{-8}	[Kjeldsen 1993, p.128]
Polyvinyl Chloride (PVC)	2.5x10^{-6}	4.00x10^{-8}	[Kjeldsen 1993, p.128]
Polytetrafluoroethylene (PTFE) "Teflon"		1.36x10^{-8}	[Kjeldsen 1993, p.128]
Upper Bound	2.5x10^{-6}	1.0x10^{-6}	

Table 2.2. Kinetic diameter of various permeant molecules

Permeant	Formula	Molecular Weight (g/mol)	Kinetic Diameter (m)	Reference
Helium	He	4.003	$2.60x10^{-10}$	[Matteucci 2006, p.6]
Water	H_2O	18.015	$2.65x10^{-10}$	[Ismail 2015, p.14]
Neon	Ne	20.180	$2.75x10^{-10}$	[NPL 2016]
Hydrogen	H_2	2.016	$2.89x10^{-10}$	[Ismail 2015, p.14]
Nitric oxide	NO	30.006	$3.17x10^{-10}$	[McKeen 2012, p.3]
Carbon dioxide	CO_2	44.010	$3.30x10^{-10}$	[Ismail 2015, p.14]
Argon	Ar	39.948	$3.40x10^{-10}$	[McKeen 2012, p.3]
Oxygen	O_2	31.999	$3.46x10^{-10}$	[Ismail 2015, p.14]
Hydrogen sulfide	H_2S	34.080	$3.60x10^{-10}$	[Matteucci 2006, p.6]
Nitrogen	N_2	28.015	$3.64x10^{-10}$	[Ismail 2015, p.14]
Carbon monoxide	CO	28.053	$3.76x10^{-10}$	[Matteucci 2006, p.6]
Methane	CH_4	16.043	$3.80x10^{-10}$	[Ismail 2015, p.14]
Ethylene	C_2H_4	28.05	$3.90x10^{-10}$	[Matteucci 2006, p.6]
Xenon	Xe	131.293	$3.96x10^{-10}$	[McKeen 2012, p.3]
Sulfur Dioxide	SO_2	64.064	$4.29x10^{-10}$	[NPL 2016]
Propane	C_3H_8	44.096	$4.30x10^{-10}$	[Matteucci 2006, p.6]
***n*-Butane**	C_4H_{10}	58.122	$4.30x10^{-10}$	[McKeen 2012, p.3]
Chlorine	Cl_2	70.906	$4.40x10^{-10}$	[NPL 2016]
Difluorodichloromethane	CF_2Cl_2	120.914	$4.40x10^{-10}$	[McKeen 2012, p.3]
Propylene	C_3H_6	42.080	$4.50x10^{-10}$	[Matteucci 2006, p.6]
Tetrafluoromethane	C_4F	67.041	$4.70x10^{-10}$	[McKeen 2012, p.3]
***i*-Butane**	C_4H_{10}	58.122	$5.00x10^{-10}$	[McKeen 2012, p.3]
Benzene	C_6H_6	78.112	$5.85x10^{-10}$	[Li 1993, p.373]

According to McKeen [McKeen 2012], permeant transfer through polymer films or membranes progresses through five consecutive steps as follows:

1. Permeant diffusion to the polymer film from the upstream atmosphere.
2. Adsorption of the permeant by the polymer film at the interface with the upstream atmosphere.
3. Diffusion of the permeant inside and through the polymer film. The diffusion step is the slowest and becomes the rate-determining step in gas permeation.
4. Desorption of the permeant at the interface of the downstream side of the film.
5. Diffusion of the permeant away from the polymer film into the downstream atmosphere.

Thus, the movement of water vapor through a layer of plastic should be viewed as water dissolving into the solid, diffusing through the material, and evaporating out the other side. McKeen also states: “Normally, exposure at 50ºC for 28 days is considered equivalent to 1 year of normal exposure.” This can be used as a rough guide for exposure.

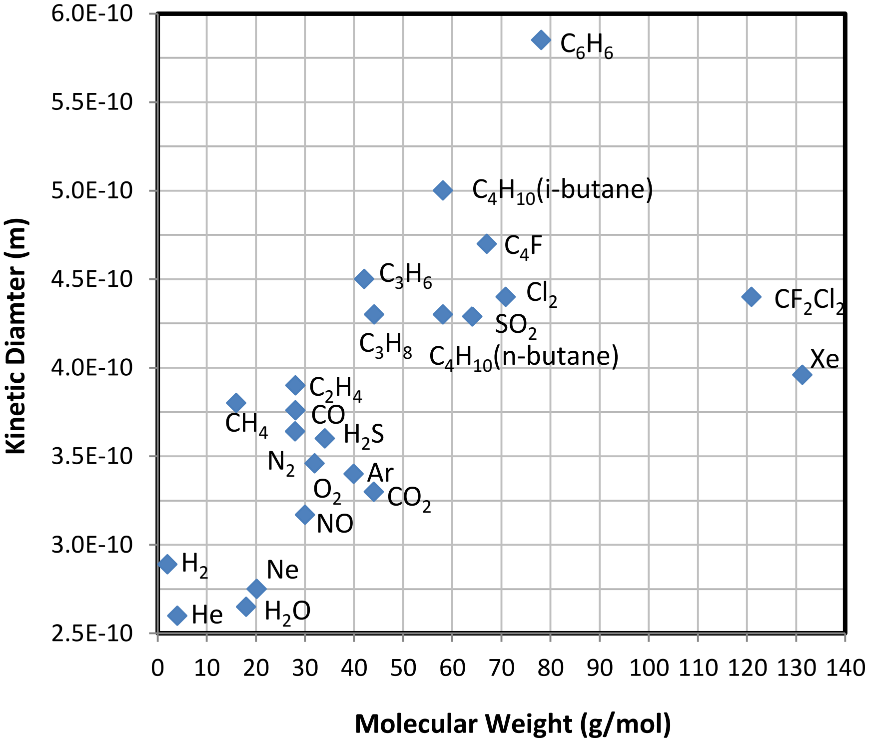

Figure 2.2. Plot of molecular weight versus kinetic diameter for various permeant gases and vapors.

3. Permeability

Permeability is a measure of a material's ability to transmit liquids, gases, and vapors (i.e., the permeant) through the material, and is given as a rate. Figure 3.1 illustrates the relative degree of permeability based on material type [Bhadha 1999]. Permeation is the rate at which a permeant passes directly through a solid. The permeability coefficient is a measure of the permeation rate and is given as the amount of the permeant (i.e., gas) passing through a unit area of polymer per unit time with a unit pressure difference across the material.

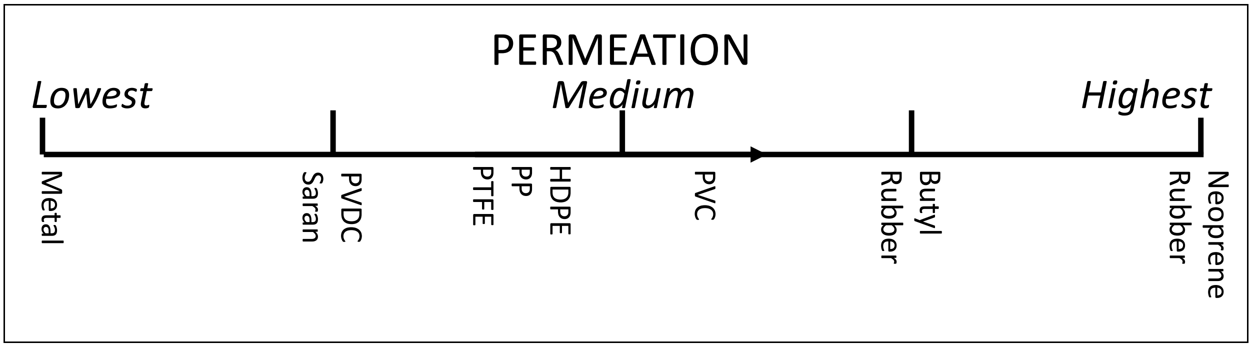

Figure 3.1. Relative degrees of permeation from [Bhadha 1999].

3.1 Gas Permeability

For gases, the amount of permeation is generally given in units of volume while for water vapor it is given as mass. Sometimes the amount is given in moles. This is the flux divided by the pressure gradient. So P for a gas, such as O_2, generally has units of

$$P_{O_2} = \frac{volume\cdot thickness}{area\cdot time\cdot pressure} = \frac{length^3\cdot length}{length^2\cdot time\cdot mass/(length\cdot time^2)} = \frac{length^3\cdot time}{mass}. \qquad \text{(Eq. 3.1)}$$

A wide variety of units are used for gas permeability due to the variation in industries measuring permeability. One industrial unit that has some usage in gas permeation is the unit Barrer named for Richard Maling Barrer [Ravishankar 1997]. Other common units of gas permeability include:

$\frac{cm^3\cdot mm}{m^2\cdot d\cdot atm}$, $\frac{cm^3\cdot cm}{cm^2\cdot s\cdot Pa}$, $\frac{cm^3\cdot cm}{cm^2\cdot s\cdot atm}$, $\frac{cm^3\cdot cm}{cm^2\cdot s\cdot (cm\ Hg)}$, $\frac{cm^3\cdot cm}{m^2\cdot 24h\cdot (mm\ Hg)}$, $\frac{cm^3\cdot cm}{m^2\cdot 24h\cdot hPa}$ $\frac{m^3\cdot mm}{m^2\cdot 24h\cdot kPa}$, $\frac{ml\cdot mm}{m^2\cdot 24h\cdot atm}$, $\frac{ml\cdot mm}{m^2\cdot d\cdot atm}$, $\frac{cm^3\cdot mil}{100in^2\cdot 24h\cdot atm}$, $\frac{m^3\cdot m}{m^2\cdot s\cdot Pa}$, and $\frac{ft^3\cdot mil}{ft^2\cdot 24h\cdot psi}$ to name a few.

The conversion between these various mixed units is given in Table 3.1.

3.2 Water Vapor Permeability

For water vapor, the amount is generally given in mass, so permeability is generally in units of

$$P_{H_2O} = \frac{mass}{area\cdot time} = \frac{length^3\cdot length}{length^2\cdot time\cdot mass/(length\cdot time^2)} = \frac{mass}{length^2\cdot time}. \qquad \text{(Eq. 3.2)}$$

Table 3.1. Conversion between volume permeability units commonly used with gases at STP.

Unit	To: $\frac{cm^3 \cdot mm}{m^2 \cdot d \cdot atm}$	From: $\frac{cm^3 \cdot mm}{m^2 \cdot d \cdot atm}$	Unit	To: $\frac{cm^3 \cdot mm}{m^2 \cdot d \cdot atm}$	From: $\frac{cm^3 \cdot mm}{m^2 \cdot d \cdot atm}$
	Multiply by	**Multiply by**		**Multiply by**	**Multiply by**
$Barrer$	65.664	0.015229	$\frac{cm^3 \cdot mm}{m^2 \cdot d \cdot (mm\ Hg)}$	760	0.0013158
$\frac{cm^3 \cdot cm}{cm^2 \cdot s \cdot (cm\ Hg)}$	6.5664×10^{11}	1.5229×10^{-12}	$\frac{cm^3 \cdot mm}{cm^2 \cdot d \cdot atm}$	10^{4}	10^{-4}
$\frac{cm^3 \cdot mm}{m^2 \cdot s \cdot Pa}$	8.7545×10^{9}	1.1142×10^{-10}	$\frac{cm^3 \cdot mm}{cm^2 \cdot s \cdot kPa}$	8.7545×10^{10}	1.14227×10^{11}
$\frac{cm^3 \cdot cm}{cm^2 \cdot s \cdot (10\ torr)}$	6.5664×10^{11}	1.5229×10^{-12}	$\frac{cm^3 \cdot N}{m^2 \cdot d \cdot bar}$	1.0133	0.98687
$\frac{cm^3 \cdot cm}{cm^2 \cdot s \cdot Pa}$	8.7545×10^{14}	1.1142×10^{-15}	$\frac{cm^3 \cdot mm}{cm^2 \cdot d \cdot Pa}$	1.0133×10^{5}	9.8687×10^{-6}
$\frac{cm^3 \cdot cm}{cm^2 \cdot s \cdot bar}$	8.7545×10^{9}	1.1142×10^{-10}	$\frac{cm^3 \cdot cm}{cm^2 \cdot s \cdot atm}$	$8.\ 6400\times10^{9}$	1.1574×10^{-10}
$\frac{cm^2}{s \cdot atm}$	8.6400×10^{9}	1.1574×10^{-10}	$\frac{cm^3 \cdot mil}{100in^2 \cdot d \cdot atm}$	39.3701	0.0254
$\frac{cm^3 \cdot mil}{100in^2 \cdot s \cdot atm}$	656.64	1.5229×10^{-3}			
$\frac{ml \cdot mil}{m^2 \cdot d \cdot atm}$	0.0254	39.37			
$\frac{in^3 \cdot mil}{100in^2 \cdot d \cdot atm}$	6.4516	0.15500×10^{-6}			
$\frac{ft^3 \cdot mil}{ft^2 \cdot d \cdot psi}$	1.1377×10^{5}	8.7897	$\frac{cm^3}{100in^2 \cdot d \cdot bar}$	15.705	0.063674
$\frac{mm^3}{m \cdot d \cdot MPa}$	0.10133	9.8687	$\frac{cm^3 \cdot mil}{100in^2 \cdot d \cdot bar}$	0.010133	98.687

As with diffusivity, permeability is temperature dependent and the relationship to temperature, for an idealized material, is given by the Arrhenius equation as:

$$P = P_0 e^{-E_A/RT}, \qquad \text{(Eq. 3.3)}$$

where E_A is the activation energy for diffusion and P_0 is the coefficient of permeation. Permeability increases roughly 5% per °C rise in temperature [SABIC 2016, p.2]. This can be used as an approximation for small temperature changes.

4. Moisture (Water) Vapor Transmission Rate

The moisture vapor transmission rate ($MVTR$), also called the water vapor transmission rate ($WVTR$), is a measure of how much water vapor mass can penetrate an area of a material in a given time period, so is given in units of mass per area per time. It comes from the more general vapor transmission rate (VTR) covering all sorts of vapor. It provides an overall view of how a thin layer of material performs as a barrier to permeants, so it is most applicable to packaging materials, where it is relevant to applications where materials are encapsulated to prevent water ingress.

The $MVTR$ can be related to the material permeability for a thin sample by [Lahtinen]:

$$MVTR = -D\frac{dC}{dx} = DS\frac{dp}{dx} = P\frac{(p_2-p_1)}{L} = P_0\frac{(p_2-p_1)}{L}e^{-E_A/RT} \qquad \text{(Eq. 4.1)}$$

where P_0 is the coefficient of permeation constant for the material, E_A is the activation energy, R is the gas constant, T the absolute temperature, p is the pressure, p_2-p_1 is the partial pressure difference of the water vapor across the material, and L is the material thickness [Lahtinen][Stevens][Arrhenius 1889a, 1889b].[1] This is the same relationship shown for J in Equation 2.1. This equation is linear in the permeability, pressure difference and material thickness, but exponential with the inverse of temperature. This explains why elevated temperature has such a dramatic impact on the rate of water absorption by PVT.

For $MVTR$, mass is usually measured in grams, area in m^2 or 100 in^2, and time in days. $MVTR$ given in per m^2 is 15.5 times larger than when measured in per 100 in^2. This mixed use of units makes analysis of the published data in the literature challenging. Table 4.1 lists the conversion between different MVTR units. The temperature at which the MVTR is measured should be given since the value is exponential in the inverse of the absolute temperature.

As an example, human skin has a $MVTR$ of ~200-400 $g \cdot m^{-2} \cdot d^{-1}$. Most plastic films have an $MVTR$ ~1-100 in these units. A $MVTR$ value of 1 x 10^{-6} $g \cdot m^{-2} \cdot d^{-1}$ has become the unofficial standard for the organic light emitting diode (OLED) industry to achieve a device lifetime of greater than 10,000 hours.

Testing at PNNL indicated that a multilayer plastic bag (Uline Dry-Shield bags) was inadequate to prevent fogging of PVT exposed to the standard Accelerated Aging Test [Kouzes 2016]. These bags, 3.6-mil thick, are formed of layers of polyethylene, aluminum, and polyester, and have a water vapor transmission rate declared by the vendors of 0.035 $g \cdot 100\text{-}in^{-2} \cdot d^{-1}$ (0.5 $g \cdot m^{-2} \cdot d^{-1}$, or 0.6 $ng \cdot cm^{-2} \cdot s^{-1}$). However, pinholes may have been a problem for this test, so a repeated test is planned.

Shannon Packaging has a material of three layers of PET surrounding two layers of aluminum foil that has an $MVTR$ declared by the vendors of 0.006 $g \cdot 100\text{-}in^{-2} \cdot d^{-1}$ (0.09 $g \cdot m^{-2} \cdot d^{-1}$, or 0.1 $ng \cdot cm^{-2} \cdot s^{-1}$).

Based on previous work, discussed later, it seems that an $MVTR$ on the order of 10^{-4} to 10^{-6} $g \cdot m^{-2} \cdot d^{-1}$ is needed for protecting PVT from fogging. A small pinhole in a material can greatly impact the $MVTR$; a seemingly small leak of 1-ppm water vapor results in a permeation rate of 0.01 $g \cdot m^{-2} \cdot d^{-1}$ [Stevens]. There is an ASTM standard for testing of $MVTR$ values (ASTM 1249).

Table 4.2 provides a list of permeability coefficients for a number of materials. The standard units used in [McKeen 2012] listed in the table for P_0 are ($cm^3 \cdot mm$)/ ($m^2 \cdot day \cdot atm$) for oxygen gas volume or ($g \cdot mm$)/($m^2 \cdot day \cdot atm$) for water gas mass. McKeen explains: "When gas volumes are used, they are usually for gas at standard temperature and pressure (STP) conditions. The current version of IUPAC's STP is a temperature of 0°C (273.15 K, 32°F) and an absolute pressure of 100kPa [14.504 pounds per

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[1] A permeability calculator is available at http://www.stevenabbott.co.uk/practical-coatings/permeability.php

square inch (psi), 0.986 atm], while NIST's version is a temperature of 20°C (293.15 K, 68°F) and an absolute pressure of 101.325 kPa (14.696 psi, 1 atm)." [McKeen 2012] The values in the table may be from measurements at different temperatures.

Table 4.1. Conversion between various gas transmission rates (left side) and water vapor transmission rights (right side).

Gas Transmission Rate			Moisture Vapor Transmission Rate		
Unit	To: $\frac{cm^3}{m^2 \cdot d}$	From: $\frac{cm^3}{m^2 \cdot d}$	**Unit**	To: $\frac{g}{m^2 \cdot d}$	From: $\frac{g}{m^2 \cdot d}$
	Multiply by	**Multiply by**		**Multiply by**	**Multiply by**
$\frac{cm^3 \cdot mil}{100in^2 \cdot 24h}$	0.393701	2.5400	$\frac{g}{100in^2 \cdot d}$	15.500	0.064516
$\frac{g}{m^2 \cdot d}$ (O_2)	1.43×10^{-3}	6.993×10^{2}	$\frac{kg}{m^2 \cdot d}$	10^{3}	10^{-3}
$\frac{g}{m^2 \cdot d}$ (H_2O)	8.03×10^{-4}	1.245×10^{-3}	$\frac{kg}{m^2 \cdot s}$	1.1574×10^{-2}	86.4
$\frac{g}{m^2 \cdot d}$ (N_2)	1.16×10^{-3}	8.620×10^{2}	$\frac{ng}{cm^2 \cdot s}$	1.1574×10^{-10}	8.64×10^{9}
$\frac{g}{m^2 \cdot d}$ (H_2)	8.92×10^{-5}	1.121×10^{4}	$\frac{lb_m}{ft^2 \cdot d}$	2.0482×10^{-4}	4882.4
			$\frac{lb_m}{100in^2 \cdot d}$	3.3601	0.29761

Table 4.2. Permeability coefficient (P_0) of oxygen and water vapor for various polymers from the literature.

Polymer	P_0 (Oxygen) $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	P_0 (Water) $g \cdot mm/m^2 \cdot d$	References
	STYRENES		
Acrylonitrile - Styrene	19.7- 102	2.0 - 6.3	
Acrylonitrile Butadiene Styrene (ABS)	19.7 - 102	2.0 - 6.3	[Kjeldsen 1993, p.127] [McKeen 2012,p.3,p.78]
Sabic Cycolac ABS	39.3	5.88	[GE 1997-2002]
BASF Terluran ABS Film	45.6 - 81	3.1	[BASF 1990]
Dow Acrylonitrile ABS Films	47 - 102	2.0 - 6.3	[Dow 1979]
Styrene-Acrylonitrile (SAN)	20.2 - 50.6	2.0 - 2.5	[McKeen 2012,p.78-79]
Dow Chemical Tyril SAN	31.5 - 39.4		[McKeen 2012,p.87]
BASF Luran 378P	20.2 - 30.4	2.0 - 2.5	[McKeen 2012,p.81]
Polystyrene (PS)	98.5-171	0.8 - 3.9	[Kjeldsen 1993, p.127]
Dow Trycite Oriented PS Film	98 - 138	1.3	[McKeen 2012,p.83-84]
BASF AG Polystyrol 168 N GPPS Film	101	1.2	[McKeen 2012,p.168]
	RUBBERS		
Rubbers	7.88 - 4330		[Kjeldsen 1993, p.127]
Polyisoprene	4320		[McKeen 2012,p.278]
Butyl rubber	7.88 - 85.4		[Kjeldsen 1993, p.127]
Methyl rubber	11.8 - 722		[Kjeldsen 1993, p.127]
Natural rubber	1530 - 1580		[Kjeldsen 1993, p.127]
Neoprene	1640 - 2630		[Kjeldsen 1993, p.127]
Silicone rubber	3940 - 4330		[Kjeldsen 1993, p.127]
Polyvinylidene fluoride/Hexafluoropropylene	95.2		[Kjeldsen 1993, p.127]
	POLYAMIDES		

Polymer	P_0 (Oxygen) $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	P_0 (Water) $g \cdot mm/m^2 \cdot d$	References
Polyamides (PA) “Nylon”	0.3 – 23.6	0.24 - 125	[McKeen 2012, p.129]
Nylon 6	0.394 – 2.50		[Kjeldsen 1993, p.127]
Nylon 6,6	2.23		[Kjeldsen 1993, p.127]
Nylon 6,9	2.3		[Kjeldsen 1993, p.127]
Nylon 6,10	1.31		[Kjeldsen 1993, p.127]
Nylon 8	3.81		[Kjeldsen 1993, p.127]
Nylon 11	7.88		[Kjeldsen 1993, p.127]
Nylon 12	23.6		[Kjeldsen 1993, p.127]
Capron Nylon		7.5-7.9	[van Weeren 2002]
DuPont Selar Amorphous Nylon	2.16 - 7.07	0.47 - 0.55	[McKeen 2012, p.126]
DuPont Selar Blends with Nylon	0.3 - 5.9		[McKeen 2012, p.126]
Honeywell Plastics Capron Nylon 6		0.24 - 5.9	[McKeen 2012, p.128]
UBE Industries Nylon 6		65 - 125	[McKeen 2012, p.134]
Polyamide Imide (PI) Resins	4.3-22.8		[McKeen 2012, p.110]
Polyamide Imide (PI) Polymers	0.1-58.7		[McKeen 2012, p.110]
Polyetherimide (PEI)	10.0-53.0	2.3-3.0	[McKeen 2012, p.115-116]
POLYETHYLENES			
Polyethylene (PE)	26.3 – 453		
High Density Polyethylene (HDPE)	26.3 – 98.5	0.1 - 0.24	[Kjeldsen 1993, p.127; McKeen 2012, p.154]
Mid Density Polyethylene (MDPE)	98.5 – 210	0.4 - 0.6	[Kjeldsen 1993, p.127; McKeen 2012, p. ??]
Low Density Polyethylene (LDPE)	98 – 453	0.39 - 0.59	[Kjeldsen 1993, p.127; McKeen 2012, p.151]
Polyethylene Naphthalate (PEN)	0.5	0.096 - 4.2	[McKeen 2012, p.100]
DuPont Mylar Films (no metal) PEN	1.13 - 1.18	0.38 - 0.57	[McKeen 2012, p.101-102]
Mitsubishi Hostaphan PEN	0.9		[McKeen 2012, p.103]
FLUOROPOLYMERS			
Fluoroplastics	1.18 – 394		
Polychlorotrifluoroethylene (PCTFE)	2.76 – 5.91		[Kjeldsen 1993, p.127]
Fluorinated ethylene propylene (FEP)	295 – 394	0.087	[van Weeren 2002]
Polyvinyl fluoride (PVF)	1.18	0.83	[van Weeren 2002]
Polytetrafluoroethylene (PTFE)	222 – 387	0.0045 - 0.30	[McKeen 2012, p.197-201]
Polyvinylidene Fluoride (PVDF)	5.52		[McKeen 2012, p.220-225]
VINYLS			
Ethylene Vinyl Alcohol (EVOH)	0.01 – 0.15	0.8 - 2.4	[McKeen 2012, p.164-165]
Polyvinylidene Chloride (PVDC)	0.00425 - 0.57	0.025 - 0.913	[McKeen 2012, p.182]
DOW Saran PVDC Films	0.00425 - 0.00625		[McKeen 2012, p.184]
Polyvinyl Chloride (PVC)	3.28-394	0.94 - 0.95@38C	[McKeen 2012, p.180]
PVC, rigid	3.28-39.4		[McKeen 2012, p.180]
PVC, plasticized	39.4-394		[McKeen 2012, p.180]
Polyvinyl toluene (PVT)			
OTHER			
Poly Propylene (PP)	35 - 377		[Kjeldsen 1993, p.127; McKeen 2012,p.157]
Llyondell Basell Adflex PP	35 - 377		[McKeen 2012, p.158]
Polyoxymethylene (POM)	5.9		[Kjeldsen 1993, p.127]
Polycarbonate (PC) ‘Lexan’	71 - 124		[McKeen 2012, p.94-97]
Dow Calibre PC	91 - 124		[McKeen 2012, p.94-97]
Bayer Makrolon PC	71 - 81		[McKeen 2012, p.94-97]
Polymethylpentene (PMP)		7.8 - 91	[McKeen 2012, p.160]
Polymethyl Methacrylate (PMMA)	5.8-6.7	1.7	[Kjeldsen 1993, p.127] [McKeen 2012,p.186-187]
Lucite Diakon PMMA	5.8	1.7	[Kjeldsen 1993, p.127]
Polyethyl Methacrylate (PEMA)			

Table 4.3 lists some permeability coefficients from [Bhadha 1999] at 25°C, where the units of P are in $cm^3 \cdot cm \cdot cm^{-2} \cdot s^{-1} \cdot cm\text{-}Hg^{-1}$, and also in $g \cdot mm \cdot m^{-2} \cdot d^{-1}$ at 1 atm. The latter values were converted using the density of water in air at this temperature. Where there is overlap of this table and Table 4.2 (for PVC), there is a difference of about a factor of two, but the results are at different temperatures and there may be variation in the exact material. The results found for PVT will be compared to these values.

Table 4.3. Some Water Vapor Permeability Coefficients [Bhadha 1999]

Polymer	**Permeability at 25°C ($Px10^{10}$)** ($cm^3 \cdot cm \cdot cm^{-2} \cdot s^{-1} \cdot cm\text{-}Hg^{-1}$)	**Permeability at 25°C** ($g \cdot mm \cdot m^{-2} \cdot d^{-1}$) @ 1 atm
Plexiglas (polymethyl methacrylate)	3200	5.25
Natural Rubber (polyisoprene)	2290	3.76
Lexan (polycarbonate)	1400	2.3
Neoprene G (polychloroprene)	910	1.49
PVT (polyvinyl toluene)	Provided later in this report	Provided later in this report
PVC (polyvinyl chloride)	275	0.451
PET (polyethylene terephthalate)	130	0.213
Butyl Rubber (polyisobutene-coisonrene)	110	0.181
LDPE (polyethylene @0.914 g/cm^3)	68	0.112
PP (polypropylene @0.907 g/cm^3)	35	0.0575
HDPE (polyethylene @0.964 g/cm^3)	9	0.0148
Teflon (polytetrafluoroethylene)	4.8	0.00788
Saran (polyvinyledene chloride)	0.5	0.000821
Kel-F81 (polytriflouro chloroethylene)	0.1	0.000164

As an example of what the values in this table imply, as will be discussed later, the mass transfer rate M is related to the permeability P, the exposed area A, the pressure difference Δp and the thickness Δx as:

$$M = PA\, \Delta p / \Delta x.$$

For a specific example, a 1-atmosphere pressure difference across a 1-mm thickness of HDPE with an area of 1 cm^2 at 25°C would give 62 ng/h of water transport. Thus, significant amounts of water can penetrate plastic over time.

5. Permeation of Water into Thick Plastic

As discussed previously, Frick's Second Law describes the concentration of a permeant in a material as a function of time and distance:

$$D\frac{d^2C}{dx^2} = \frac{dC}{dt}. \qquad \text{(Eq. 5.1)}$$

This "heat" equation can be solved by separation of variables using inhomogeneous boundary conditions, and assuming a constant diffusion constant. To understand the permeation of water into a thick plastic, the non-steady state condition of diffusion illustrated in Figure 5.1 is used. This work was based on [Crank 1975] and the solution for the concentration of sorbed permeant for this condition, described by [Karimi 2011] as a function of time, is given by this series:

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{2n+1} e^{\frac{-Dx(2n+1)^2\pi^2 t}{L^2}} \times\cos\left(\frac{(2n+1)\pi x}{L}\right). \qquad \text{(Eq. 5.2)}$$

where C_t is the concentration at a given time and position (x), and C_∞ is the concentration at equilibrium. This solution can be rewritten with the specific terms of the series as

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}(A_0 + A_1 + A_2 + A_3 + A_4 + \cdots), \qquad \text{(Eq. 5.3)}$$

where the following represent the first five terms:

$$First\ Term = A_0 = \frac{1}{1}e^{-\frac{D\pi^2 t}{L^2}} \times\cos\left(\frac{\pi x}{L}\right)$$

$$Second\ Term = A_1 = \frac{-1}{3}e^{-\frac{9D\pi^2 t}{L^2}} \times\cos\left(\frac{3\pi x}{L}\right)$$

$$Third\ Term = A_2 = \frac{1}{5}e^{-\frac{25D\pi^2 t}{L^2}} \times\cos\left(\frac{5\pi x}{L}\right)$$

$$Fourth\ Term = A_3 = \frac{-1}{7}e^{-\frac{49D\pi^2 t}{L^2}} \times\cos\left(\frac{7\pi x}{L}\right)$$

$$Fifth\ Term = A_4 = \frac{1}{9}e^{-\frac{81D\pi^2 t}{L^2}} \times\cos\left(\frac{9\pi x}{L}\right).$$

Looking at the x range *–L/2* to *+L/2,* Equation 5.3 at time $t=0$, gives:

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}\left\{\cos\left(\frac{\pi x}{L}\right) - \frac{1}{3}\cos\left(3\frac{\pi x}{L}\right) + \frac{1}{5}\cos\left(5\frac{\pi x}{L}\right) - \frac{1}{7}\cos\left(7\frac{\pi x}{L}\right) + \frac{1}{9}\cos\left(9\frac{\pi x}{L}\right) + \cdots\right\}$$

At the left boundary, where $x=-L/2$, this gives:

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}\left\{\cos\left(\frac{-1}{2}\pi\right) - \frac{1}{3}\cos\left(\frac{-3}{2}\pi\right) + \frac{1}{5}\cos\left(\frac{-5}{2}\pi\right) - \frac{1}{7}\cos\left(\frac{-7}{2}\pi\right) + \frac{1}{9}\cos\left(\frac{-9}{2}\pi\right) + \cdots\right\}$$

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}\left\{-1 + \frac{1}{3} - \frac{1}{5} + \frac{1}{7} - \frac{1}{9} + \cdots\right\} = 1,$$

and at the right boundary where $x=L/2$, this gives:

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}\left\{\cos\left(\frac{1}{2}\pi\right) - \frac{1}{3}\cos\left(\frac{3}{2}\pi\right) + \frac{1}{5}\cos\left(\frac{5}{2}\pi\right) - \frac{1}{7}\cos\left(\frac{7}{2}\pi\right) + \frac{1}{9}\cos\left(\frac{9}{2}\pi\right) + \cdots\right\}$$

$$\frac{c_t}{c_\infty} = 1 - \frac{4}{\pi}\left\{-1 + \frac{1}{3} - \frac{1}{5} + \frac{1}{7} - \frac{1}{9} + \cdots\right\} = 1.$$

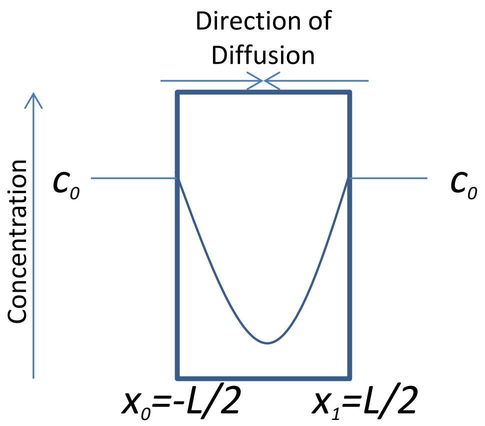

Figure 5.1. Non-steady state for diffusion from the left and right boundaries into the center of the material [Karimi 2011].

So, at time *t=0* the concentration is the same on both the left and right boundary of the material, and is the same as in the air, which is expected since nothing should have penetrated the material yet. Figure 5.2 provides a representation of the time evolution of the concentration in the material as a function of position and time, over ten equal time steps. At very early times, the concentration is zero everywhere inside the material. As time progresses, the water concentration increases, following the exponential form of Equation 5.2. At long times, the concentration becomes uniform throughout the material.

Similarly, [Crank 1975] showed that the mass of sorbed permeant as a function of time is given by this series:

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}e^{\frac{-D(2n+1)^2\pi^2 t}{L^2}} \qquad \text{(Eq. 5.4)}$$

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2}(B_0 + B_1 + B_2 + B_3 + B_4 + \cdots)$$

$$First\ Term = B_0 = \frac{1}{1}e^{-\frac{D\pi^2 t}{L^2}}$$

$$Second\ Term = B_1 = \frac{1}{9}e^{-\frac{9D\pi^2 t}{L^2}}$$

$$Third\ Term = B_2 = \frac{1}{25}e^{-\frac{25D\pi^2 t}{L^2}}$$

$$Fourth\ Term = B_3 = \frac{1}{49}e^{-\frac{49D\pi^2 t}{L^2}}$$

$$Fifth\ Term = B_4 = \frac{1}{81}e^{-\frac{81D\pi^2 t}{L^2}}.$$

The series sum goes to zero when a larger number of terms are included in the summation. This is expected because at time zero there should be no penetrant permeant into the material. For a thickness of *L*, this mass ratio can be rewritten for early times of absorption as

$$\frac{M_t}{M_\infty} = \frac{4}{L}\left(\frac{D}{\pi}\right)^{1/2} t^{1/2}. \qquad \text{[Crank 1975, pg. 244] (Eq. 5.5)}$$

For later stages of diffusion, this relation becomes

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} e^{-4\pi^2 Dt/L^2} \qquad \text{(Eq. 5.6)}$$

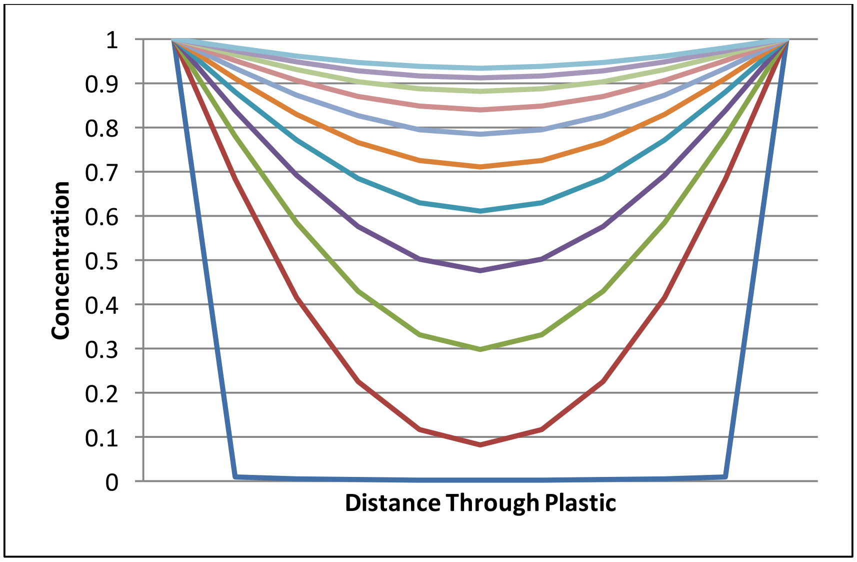

Figure 5.2. Concentration as a function of time.

From Equation 5.2, a characteristic half-saturation time can be defined when $\frac{M_t}{M_\infty} = 0.5$. From this, the characteristic half-saturation time τ for the permeation into the material would be:

$$\tau = \frac{0.049\, L^2}{D} \qquad \text{(Eq. 5.7)}$$

6. MVTR for PVT

The MVTR value is specific to a sample of a given composition, thickness, temperature, and pressure difference. From measurements at different temperatures, a trend can be fit to provide the temperature dependence constant of the relationship for a given material, E_a. It can also provide the value of the permeability constant, P_0.

6.1 MVTR Measurement

A sample of PVT ~23 mil (0.58 mm) thick provided by Saint Gobain, shown in Figure 6.1, was sent to MOCON (Brooklyn Park, MN) to have the MVTR measured under the ASTM F-1249 WVTR standard following the procedure written in QMS 504-004. Four samples of PVT were cut from the sheet for measurement at three temperatures (30°C, 40°C, and 50°C), all at 100% relative humidity. The results of these measurements are listed in Table 6.1 and plotted in Figure 6.2. The units of MVTR used here are $g \cdot m^{-2} \cdot d^{-1}$. Only one thickness sample was tested due to the cost.

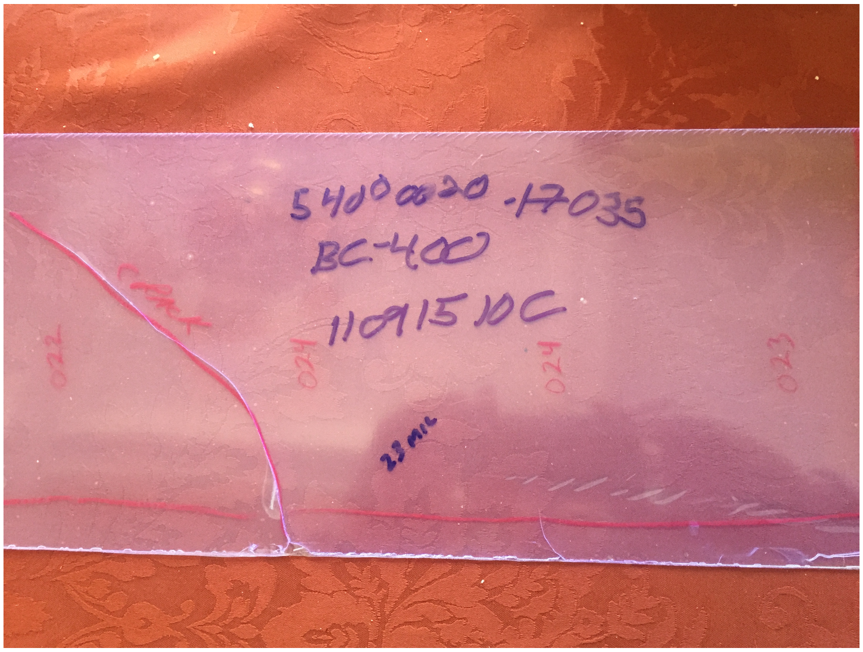

Figure 6.1. Thin sample of PVT provided by Saint Gobain.

Table 6.1. WVTR results from MOCON in $g \cdot m^{-2} \cdot d^{-1}$.

Temperature	Sample 1	Sample 2	Sample 3	Sample 4	Average	Std. Dev.
30°C	2.49	2.70	2.51	2.89	2.65	0.19
40°C	4.00	4.57	4.25	4.76	4.40	0.34
50°C	6.41	7.64	7.05	7.67	7.19	0.59

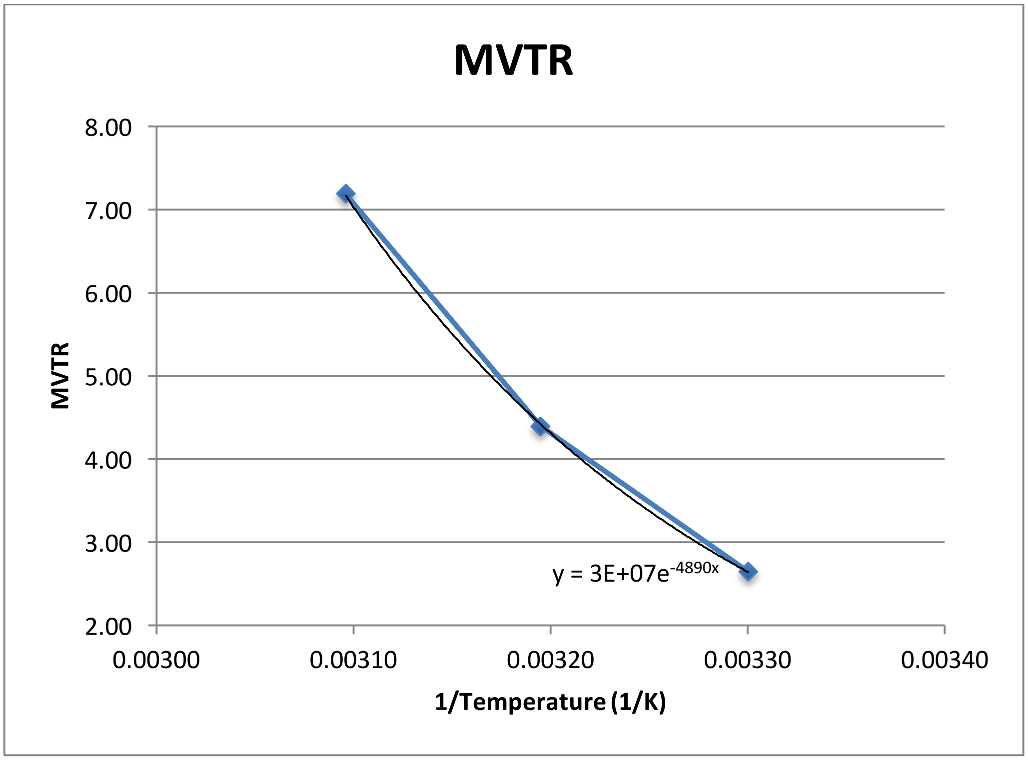

Figure 6.2. WVTR results, with trendline.

6.2 Constants Derived From MVTR Measurement

A trendline fit to the average of the data gives the equation:

$$\text{MVTR} = 2.69 \times 10^{7}\ e^{-4890/T}\ [\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}]$$

The predicted MVTR values at other temperatures using this fit function are listed in Table 6.2. This demonstrates the impact of the exponential rise in MVTR, showing that the MVTR value at 55°C is six times larger than the value at 20°C.

Table 6.2. MVTR versus temperature predictions based on MOCON results.

Temperature (°C)	**Temperature (K)**	**1/Temperature (1/K)**	**Measured MVTR ($g\cdot m^{-2}\cdot d^{-1}$)**	**Predicted MVTR ($g\cdot m^{-2}\cdot d^{-1}$)**
0	273	0.00366	-	0.45
10	283	0.00353	-	0.84
20	293	0.00341	-	1.52
30	303	0.00330	2.65	2.64
40	313	0.00319	4.40	4.42
50	323	0.00310	7.19	7.16
55	328	0.00305	-	9.02
60	333	0.00300	-	11.29
65	338	0.00296	-	14.03

As described in Section 4, the equation for MVTR is:

$$MVTR = P\frac{(p_2-p_1)}{L} = P_0\frac{(p_2-p_1)}{L}e^{-E_A/_{RT}} \quad \text{(Eq. 4.1)}$$

In Equation 4.1, the exponent is $-E_A/RT$, where the gas constant $R = 8.31\ J \cdot K^{-1} mol^{-1}$. The result of the measurement of MVTR gives a value of **$4.1x10^4\ J \cdot mol^{-1}$ for E_A**.

MVTR is often listed as the value at 20°C, which for PVT is 1.5(1) $g \cdot m^{-2} \cdot d^{-1}$ based on the extrapolated value listed above.

Using the thickness L = 0.58 mm, and a pressure difference of 1 atm ($1x10^5$ Pa [N/m^2]), P_0 is found to be $P_0 = 1.6x10^4\ g \cdot m^{-2} \cdot d^{-1} \cdot m \cdot atm^{-1} = 1.6x10^7\ g \cdot m^{-2} \cdot d^{-1} \cdot mm \cdot atm^{-1}$ for an infinite temperature. Computed at a temperature of **25°C, $P_0 = 1.2\ g \cdot m^{-2} \cdot d^{-1} \cdot mm \cdot atm^{-1}$**. This value of P_0 is between PVC and neoprene as listed in Table 4.3.

6.3 Saturation Time

As an example of the application of Equation 5.7 for the half-saturation time, if we consider a 5 cm thick piece of PVT with $D = 1.3x10^{-7}\ cm^2 \cdot s^{-1}$ [Krongauz 1990], **then the half-saturation time comes out to ~109 days at 23°C** (the temperature used by Krongauz). This time of about one third of a year for half-saturation indicates the nature of the long periods of time it takes for equilibrium to be established inside of a piece of plastic when exposed to water vapor. This time becomes shorter at higher temperatures due to the exponential increase in diffusion with temperature. As stated earlier, permeation increases with temperature, so a 25°C increase in temperature to 55°C would increase the permeation rate by about a factor of 3.4 based on the MOCON results (Table 6.2). Under such conditions, the half-saturation time would drop to **~32 days at 55°C**. This indicates the Accelerated Aging Test [Kouzes 2016] period of 30 days exposure at 55°C is perhaps too short to fully saturate samples of PVT that are 2” thick since that is only approximately the time to reach half-saturation.

It has been suggested by S. Payne (Lawrence Livermore National Laboratory)[2] that the value of D used above may be about five times larger than the value published by Krongauz. Assuming this, the saturation half-saturation time would become ~22 days at 30°C, and ~6 days at 55°C.

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[2] Private communication November 28, 2016

7. Humidity and Equilibrium

Absolute humidity is the amount of water vapor in the air, which is temperature dependent. Absolute humidity is measured in g·m^{-3}. Relative humidity (RH) is the percent of the maximum absolute humidity that is possible at a given temperature. Thus, 100% RH is the maximum absolute humidity at a given temperature. Table 7.1 lists the maximum absolute humidity (100% RH) of air at temperatures from 20ºC to 65ºC in terms of grams of water per cubic meter of air as listed on five different web sites. There are inconsistencies in these values, especially among the last three columns. It is suggested that the second column (B) from Vaisala.com be used as the values for computations.

Table 7.1. Water density in air from various online sources.

Temp. (ºC)	**A Water in air @ 100% RH (g·m^{-3})**	**B Water in air @ 100% RH (g·m^{-3})**	**C Water in air @ 100% RH (g·m^{-3})**	**D Water in air @ 100% RH (g·m^{-3})**	**E Water in air @ 100% RH (g·m^{-3})**
20	17.3	17.3	17.4	17.3	14.6
22	19.4	19.5	19.5	19.4	16.6
23	20.5	20.6	20.7	20.6	17.6
30	26.5	30.4	30.5	30.5	27.3
40	51.0	51.2	51.3	51.0	50.9
50	82.7	83.0	83.3	80.7	94.9
55	104.0	104.3	104.7	99.5	129.6
60	132.2	130.0	130.5	121.4	177.0
65	160.3	160.7	161.4	146.4	241.8

A: https://www.rotronic.com/en/humidity_measurement-feuchtemessung-mesure_de_l_humidite/humidity-calculator-feuchterechner-mr
B: http://go.vaisala.com/humiditycalculator/5.0/
C: http://www.humcal.com/index.php
D: http://hyperphysics.phy-astr.gsu.edu/hbase/Kinetic/relhum.html
E: http://www.lenntech.com/calculators/humidity/relative-humidity.htm

Table 7.2 lists the properties of small, medium-small, medium and large PVT samples used in previous and current testing. The density of PVT is taken as 1.05 g·cm^{-3}. The Small samples are being used in the present near-term and root-cause studies, while the Medium-Small and Medium samples were used in previous studies of PVT fogging [Fritz 2014]. The Large samples represent full-sized PVT panels used in deployed RPMs.

Table 7.2. PVT Samples Used in Testing.

Sample	**Dimensions (cm)**	**Dimensions (in.)**	**Volume (cm^3)**	**Mass (g)**
Small	20.32 x 20.32 x 5.715	8 x 8 x 2.25	2360	2478
Medium-Small	35.56 x 35.56 x 3.81	14 x 14 x 1.5	4818	5059
Medium	35.56 x 35.56 x 5.08	14 x 14 x 2	6424	6745
Large	35.56 x 178 x 5.715	14 x 70 x 2.25	36 174	37 983

Based on the concept that PVT acts like an open matrix through which water vapor can pass, the following hypothesis is posed:

Hypothesis: At equilibrium, the concentration of water (mass per unit volume) in the PVT will be the same as the concentration in the air around the plastic.

If this hypothesis is correct, then there is likely no chemical bonding of the water to the plastic taking place. If it is found that this hypothesis is incorrect, it could have two interpretations. If the density of water vapor is below that of air when the plastic saturated, then it is likely that the volume of "empty" space inside the plastic is limiting the amount of water. If the density of water vapor is higher than that in air, it is likely that there is a chemical bonding effect drawing water into the plastic.

Table 7.3 list the properties of humidity of air at temperatures from 20ºC to 65ºC in terms of grams of water per cubic meter of air for 100% RH, as above. The next three columns of the table list the mass of water (in g) that would be in each of the plastic sample if they were saturated with water at the same density as the air. This assumption may or may not be valid, but indicates the potential water that could saturate the PVT sample if in equilibrium with the air. The last column gives the ratio of water mass to PVT mass at equilibrium in parts per million (ppm), which is equivalent to mg/kg. These values can be compared to the approximate values found previously in fogging studies [Kouzes 2015] of about 1 g per 4 kg of PVT (~250 ppm). This comparison could imply that PVT can absorb water at a higher density than is in the surrounding air, but may just be due to the poor precision of the earlier estimate. This is related to the lack of knowledge about whether water forms any chemical bonds in PVT, or not. These questions will be invested through future measurements.

Table 7.3. Water density in air and in plastic under equilibrium assumption.

Temp. (ºC)	**Water in air @ 100% RH ($g\cdot m^{-3}$)**	**Water in Small Sample @ 100% RH (g)**	**Water in Medium Sample @ 100% RH (g)**	**Water in Large Sample @ 100% RH (g)**	**Water per PVT Mass (ppm)**
20	17.3	0.04	0.11	0.63	16
30	30.4	0.07	0.19	1.10	29
40	51.2	0.12	0.33	1.84	49
50	83.0	0.20	0.53	2.99	79
55	104.3	0.25	0.67	3.76	99
60	130.0	0.31	0.83	4.69	123
65	160.7	0.38	1.03	5.80	153

8. Diffusion in Thin Plastic

As discussed in Section 5, the diffusion coefficient *D*, also known as diffusivity, is the proportionality constant for mass flow through a layer of material that follows Frick's first law of diffusion. In the steady state, the concentration in a thin layer of material is a linear distribution across a thin sample (see Figure 2.1), giving $\frac{d^2C}{dx^2} = 0$. For the steady state condition [Crank 1975, p. 44]:

$$J = MVTR = -D\frac{dC}{dx} = D\frac{(C_1 - C_2)}{L}$$

Measurements of WVTR are made using thin samples of material, with high humidity on one side (C_1) and low humidity on the other side (C_2). The solution to Frick's second law is thus different than that for thick samples. The thin sample extends from x=0 to x=L, and the boundary conditions have $C=C_1$ at x=0 and $C=C_2$ at x=L. This is one method used to measure MVTR, such as by MOCON (see Section 6).

This equation can be reformatted to find the value of *D* as:

$$D = MVTR\ \frac{L}{(C_1 - C_2)} \qquad \text{(Eq. 1)}$$

The value of *D* is temperature dependent, and temperature values for measurements should be given with the value of *D*. However, if the temperature dependence of *D* and C is the same, then this relationship will give the value D_0 (see Equation 2.5). **Table 8.1** lists the value of *D* for polyvinyl toluene from various derivations at various temperatures, as discussed below.

The only published value for *D* for PVT (at 23°C) listed in the table is by [Krongauz 1990] using a new technique of a radiotracer penetrating the material. Steve Payne (LLNL) has stated that a preliminary estimate for *D* from PVT sample saturation and drying times, gives a much larger PVT diffusion coefficient.

The MVTR for PVT samples was measured by MOCON at 30°C, 40°C, and 50°C, and this allows extrapolation to other temperatures (listed in Table 6.2). The values of *D* derived from the MOCON measured values are shown in bold in **Table 8.1**, while the extrapolated values are not bolded. The humidity densities listed in Table 7.1, column B, were used in the calculation. These values derived for *D* are 45 times larger than the value from [Krongauz 1990], and somewhat smaller than the estimate by LLNL.

Table 8.1. Diffusion coefficient for PVT.

Temp. (°C)	**D From [Krongauz 1990] ($cm^2 \cdot s^{-1}$)**	**PNNL D From MOCON Data ($cm^2 \cdot s^{-1}$)**
20		5.9×10^{-6}
22		5.9×10^{-6}
23	0.13×10^{-6}	5.9×10^{-6}
30		**5.8×10^{-6}**
40		**5.8×10^{-6}**
50		**5.8×10^{-6}**
55		5.8×10^{-6}
60		5.8×10^{-6}
65		5.9×10^{-6}

The reason for these discrepancies may be due to differences in the preparation of the PVT material, surface versus volume effects for thin and thick samples, or errors in the experimental methods used. Further work will be required to determine the source of this problem and the correct value for *D* for PVT.

The data in the table derived from the extrapolation of the MOCON MVTR measurements gives a constant value of D_0 of $5.8\text{x}10^{-6}$ $\text{cm}^2\cdot\text{s}^{-1}$ as the temperature varies. This implies that the temperature dependence of *D* and C must be the same.

9. Mitigation Approaches

Mitigation of water-induced damage in PVT is needed to prevent permanent damage to the plastic scintillators when it gets very cold [Cameron 2014]. Two mitigation approaches have been proposed for preventing damage to plastic: heaters to keep the plastic from getting cold enough to produce damage, and encapsulation of the plastic to prevent water intrusion that leads to damage during cold cycles.

Encapsulation must be sufficient to prevent water vapor intrusion over years of exposure to hot and humid conditions while remaining thin enough to not interfere with the passage of low energy gamma radiation (typically down to 25 keV [Stromswold 2003]). This requires that the encapsulating material, either aluminum or plastic, at least on the front face, be no more than ~250 micrometers (10 mil) thick. The encapsulation must have a MVTR small enough (approximately $<10^{-6}$ $g \cdot m^{-2} \cdot d^{-1}$) to keep water out of the plastic for 20 years while being less than ~250 micrometers thick.

It is known that exposure of PVT to 30 days of high temperature and humidity (90% RH and 55°C) will produce severe fogging in all PVT samples when cooled to -30°C [Cameron 2014; Kouzes 2015]. Even cooling to just room temperature produces temporary fogging. This thermal cycle is referred to as the Accelerated Aging Test (AAT) [Kouzes 2016].

Encapsulation of plastic requires that it be completely sealed against water vapor intrusion, including any electronics interface, at the MVTR level listed above. There are candidate materials being evaluated. An encapsulated PVT assembly would be subjected to the AAT to verify its resistance to water vapor intrusion.

There are at least three approaches to encapsulation, referred to here as metal, membrane and chemical encapsulation. Metal encapsulation involves sealing the plastic inside aluminum foil and/or thin metal sheets, while keeping the front face covering of the plastic thin enough to allow the low energy radiation detection requirement to be met.

Membrane encapsulation involves using commercial multilayered plastic and metal membranes, such as those used to seal solar cells from the environment, that have low MVTR values. A previous study showed that membranes with MVTR of ~0.5 $g \cdot m^{-2} \cdot d^{-1}$ did not prevent fogging [Kouzes 2015]. Membranes with much smaller MVTR values ($<10^{-4}$ $g \cdot m^{-2} \cdot d^{-1}$) exist.

Chemical encapsulation involves chemical sealants applied to the PVT. Such sealants would have to retain the reflective properties now used with PVT (usually aluminum foil wrapping, but TiO_2 is also used). There are currently no known chemical encapsulants with the desired properties.

A separate report on encapsulation is in preparation [Kouzes 2016b].

10.Conclusions

The permeation of water into plastic scintillators can have important effects on their performance [Cameron 2014]. This report has investigated various theoretical and experimental aspects of water permeation into materials. A number of conclusions can be reached from the information provided in this report:

- Permeability increases about 5% per °C rise in temperature. [SABIC 2016, p.2].
- Hypothesis: At equilibrium, the concentration of water (mass per unit volume) in the PVT will be the same as the concentration in the air around the plastic.
- The heat equation describes the concentration of water in plastic as a function of time and temperature.
- The solution for the water concentration in a sheet of plastic is an infinites sum of cosine functions of the distance into the plastic sheet, and an exponential in the inverse of the temperature.
- The measured value of MVTR for PVT for a 0.58 mm (23 mil) thick sample at 20°C was found to be 1.7(1) $g \cdot m^{-2} \cdot d^{-1}$ by measurements at MOCON.
- A 5 cm thick piece of PVT with $D = 1.3x10^{-7}\ cm^2 \cdot s^{-1}$ at 30°C, will have a characteristic saturation time of ~109 days.
- Normally, exposure at 50ºC for 28 days is considered equivalent to 1 year of normal exposure [McKeen 2012].
- Computed at a temperature of 25°C, PVT has a value of $P_0 = 1.2\ g \cdot m^{-2} \cdot d^{-1} \cdot mm \cdot atm^{-1}$. This value of P_0 is between that of PVC and neoprene.
- Encapsulation to keep plastic scintillator dry must have a MVTR small enough (approximately $<10^{-6}\ g \cdot m^{-2} \cdot d^{-1}$) to keep water out of the plastic while having a thickness less than 250 micrometers (10 mil) thick.

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A. Appendix – Properties of Plastics

References for this table: [Kouzes 2012; REXON 2016; Saint Gobain 2009; Saint Gobain 2016]

Table A.1. Properties of PVT.

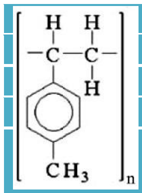
IDENTIFICATION	
 **Chemical Name**	POLY(VINYLTOLUENE)
Molecular Formula	$[CH_2CH(C_6H_4CH_3)]_n$; $(C_9H_{10})_n$
CAS Number	9017-21-4
CB Number	CB1168060
Synonyms	POLY(VINYLTOLUENE); VINYL TOLUENE RESIN; vinyltoluene homopolymer; poly(vinyltoluene),mixedisomers; ethenylmethyl-benzenhomopolymer; Benzene,ethenylmethyl-,homopolymer; POLYVINYLTOLUENE MIXED ISOMERS AVERAGE; POLYVINYLTOLUENE, MIXED ISOMERS, UNIFORM; poly(vinyltoluene)mixedisomersaveragemwca.80,000(gpc)
DENSITY	
Molecular Weight	118.1757
Density	1.032 g/cm³, 1.05 g/cm³ @ 25°C
Electron Density	3.39×10^{22} electrons/cm³
Hydrogen Density	5.28×10^{22} atoms/cm³
Carbon Density	4.78×10^{22} atoms/cm³
Hardness	0 mho
CHEMICAL PROPERTIES	
Combustibility	Combustible.
Reactivity	Incompatible with strong oxidizing agents, ammonia.
Vapor Pressure	Negligible. May be used in high vacuum
Safety	S24: Avoid contact with skin, S25: Avoid contact with eye
THERMAL PROPERTIES	
Coefficient of Linear Expansion (<67°C)	[Rexon 2016] ~9×10^{-5} °C^{-1}, [Kouzes 2012] $78(5)\times10^{-6}$ K^{-1}
Melting Point	188°C
Softening Point	70°C
Boiling Point	169.8°C @ 760 mmHg
Stability	Stable
OPTICAL PROPERTIES	
Attenuation length (e^{-1})	~43 cm
Wavelength of maximum emission	423 nm
Refractive index at emission max (423 nm)	1.581
Refractive index at sodium D line (589.3 nm)	1.59 @20°C
SCINTILATION PROPERTIES	
Typical resolution % FWHM 137Cs	180
Alpha/Beta Ratio (RP-102A)	0.072
Rise time	0.9 ns
Primary Decay time (e^{-1})	2.4 ns
Luminescent efficiency	~15% of NaI(Tl)
Phosphorescence/Afterglow (after 6 ms)	0.01%
Light yield	10,000 photons/MeV
Light Output vs. Temperature	Independent of temperature from -60°C to +20°C. At 60°C is 95% that @ 20°C

Table A.2. List of common polymers, their IUPAC names, abbreviations, and trade/product names.

Common Name	IUPAC Name	Abbreviation	Trade names
STYRENES			
Acrylonitrile	Prop-2-enenitrile	AN	
Acrylonitrile Butadiene Styrene		ABS	
Acrylonitrile Styrene Acrylate		ASA	
Polyacrylonitrile		PAN	
Polystyrene	Poly(1-phenylethene)	PS	Thermocol, Styrofoam, Polystyrol, Trycite
Styrene-Acrylonitrile	Prop-2-enenitrile; styrene	SAN	
POLYAMIDES			
Aramides / Aromatic Polyamides			Kevlar, Nomex
Polyamide	poly(hexano-6-lactam)	PA6, PA66	Nylon, Capron, Zytel, Technyl, Rilsan, Rapidol
Polyimide	2,4-diisocyanato-1-methylbenzene	PI	Kapton, UPILEX, Kaptrex
Polyphthalamides		PA6T	Trogamid
Polyetherimide		PEI	
FLUOROPOLYMERS			
Fluorinated ethylene propylene	poly(tetrafluoroethene)	FEP	Neoflon, Dyneon
Polychlorotrifluoroethylene	1-chloro-1,2,2-trifluoroethene	PCTFE	Kel-F, Neoflon
Polytetrafluoroethylene	poly(tetrafluoroethene)	PTFE	Teflon
Polyvinyl fluoride	poly(1-fluoroethylene)	PVF	Tedlar
Polyvinylidene Fluoride	poly(1,1-difluoroethylene)	PVDF	Kynar, Hylar, Solef, Syge
POLYETHYLENES			
Cross-linked polyethylene	polyethene or poly(methylene)	PEX	
High Density Polyethylene	polyethene or poly(methylene)	HDPE	
Low Density Polyethylene	polyethene or poly(methylene)	LDPE	
Mid Density Polyethylene	polyethene or poly(methylene)	MDPE	
Polyethylene	polyethene or poly(methylene)	PE	
Polyethylene Naphthalate	poly(ethylene-2,6-naphthalene dicarboxylate)	PEN	
Polyethylene Terephthalate	poly(ethyl benzene-1,4-dicarboxylate)	PET	Dacron, Terylene, Lavsan
Biaxially-oriented Polyethylene Terephthalate	poly(ethyl benzene-1,4-dicarboxylate)	BoPET	Hostaphan, Mylar, Melinex
VINYLS			
Chlorinated polyvinyl chloride	poly(chloroethylene)	CPVC, PVC-C	
Ethylene Vinyl Alcohol	ethenol	EVOH	
Polyvinyl Acetate	poly (1-acetyloxiethene)	PVAc	
Polyvinyl Alcohol	ethenol	PVA, PVOH, PVAI	Kuraray Poval, Mowiol, Celvol, Polyviol, Elvanol
Polyvinyl Butyral	poly(vinyl butyral)	PVB	
Polyvinyl Chloride	poly(1-chloroethene)	PVC	
Polyvinyl Toluene		PVT	
Polyvinylidene Chloride	Poly(1,1-dichloroethene)	PVDC	Saran, Ixan, Diofan
Vinylidene Chloride Copolymer		VDC	
OTHER			
Epoxy Resins	poly(epoxides)		
Polypropylene	poly(propene)	PP	
Polycarbonate	4-[2-(4-hydroxyphenyl)propan-2-yl]phenol	PC	Lexan, Makrolon, Hammerglass, Skylite, Sunlite, Makroclear
Polyester			
Polyethyl Methacrylate		PEMA	
Polymethyl Methacrylate	poly(methyl 2-methylpropenoate)	PMMA	Acrylic, Plexiglas, Acrylite, Lucite, and Perspex
Polymethylpentene		PMP	
Paraformaldehyde	poly(oxymethylene)	PFA,POM	Delrin
Polyurethane	butane-1,4-diol; 1-isocyanato-4-[(4-isocyanatocyclohexyl)methyl]cyclohexane; oxepan-2-one	PU, PUR	

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