# PERMEATION OF BTEX COMPOUNDS THROUGH POLYETHYLENE PIPE

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การส่องด้วยกล้องจุลทรรศน์ และอุปกรณ์ไปป์บอทเทิลถูกนำมาใช้ใน สมดุลย์การดูดซับ การศึกษาการแทรกซึมของตัวทำละลายอินทรีย์ผ่านท่อพีวิซีและท่อโพลิเอทธิลีน ท่อพีวีซีจาก 5 แหล่ง การผลิตถกนำมาใช้ในการศึกษา จากผลการทคลองพบว่าท่อพีวีซีจาก 5 แหล่งการผลิตให้ผลการ ทดสอบสมคุลการดุดซับสารไตรคลอโรเอทธิลีนและโทลอีนที่เหมือนกัน โดยท่อพีวีซีดุดซับสารทั้งสอง ชนิดได้อย่างรวดเร็ว และการดูดซับเข้าสู่สภาวะสมดย์ภายใน 7 ถึง 11 วัน แก๊สโซลีนสามชนิดถูก นำมาใช้ทุดสอบ พบว่าท่อพีวีซีดูดซับแก๊สโซลีนได้น้อยกว่า 1.5 เปอร์เซ็นต์ของน้ำหนักที่ดูดซับได้ หลังจาก 100 วันการทคลอง และเป็นที่น่าสังเกตว่า ท่อพีวีซีดูคซับพรีเมียมแก๊ส โซลีนได้มากกว่าเรกูลาร์ แก๊สโซลีนและแก๊สโซฮอล์ ซึ่งกาดว่าเป็นผลมาจากความเข้มของสารประกอบบีเท็กซ์ (เบนซีน โทลอีน เอทธิลเบนซีน และไซลีน) ในพรีเมียมแก๊สโซลีนที่สุงกว่านั่นเอง ผลจากการส่องกล้องจุลทรรศน์ยืนยัน การแทรกซึมของพรีเมียมแก๊ส โซลีนผ่านท่อพีวีซี ซึ่งพบแถบแนวการเคลื่อนที่ของพรีเมียมแก๊ส โซลีน เป็นระยะห่างประมาณ 0.3 มม. ณ 120 วันการทดลองแต่ทว่าไม่ปรากฏแถบแนวการเคลื่อนที่ของเรกูลาร์ และแก๊สโซฮอล์ในท่อพีวีซี จากผลการทคลองแนะนำได้ว่าเปอร์เซ็นต์ของน้ำหนักที่ดูคซับได้อาจจะเป็น สัคส่วนแปรผันตรงกับความเข้มข้นของสารประกอบบีเท็กซ์ในแก๊สโซลีน และการมีอย่ของคลอโรเอ ทธิลีนหรือโทลูอีนในแก๊สโซลีนคาดว่าจะช่วยเพิ่มการแทรกซึมผ่านของแก๊สโซลีนได้ การแทรกผ่าน ้ของสารประกอบบีเท็กซ์ในแก๊สโซลีนผ่านท่อโพลิเอทธิลีน เกิดขึ้นหลังจาก 7 วัน ที่ท่อสัมผัสแก๊สโซ ้อื่น และการแทรกผ่านของสารประกอบบีเท็กซ์ในสารละลายอิ่มตัวด้วยแก๊สโซลีนผ่านท่อโพลิเอทธิลีน เกิดขึ้นหลังจาก 10 วัน ที่ท่อสัมผัสสารละลายอิ่มตัวด้วยแก๊สโซแก๊สโซลีน ความเข้มข้นของ สารประกอบบีเท็กซ์ในน้ำที่อยู่ภายในท่อโพลิเอทธิลีนสูงกว่าระดับมาตรฐานที่ยอมรับได้ และพบว่า ้สัมประสิทธิ์การแทรกผ่านของสารประกอบบีเท็กซ์ขึ้นอยู่กับความเข้มข้นของสารประกอบบีเท็กซ์ที่ผิว ภายนอกของท่อ

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Equilibrium sorption test, microscopic visualization test, and pipe-bottle permeation test were conducted to study the permeation of various organic solvents through polyvinyl chloride (PVC) and polyethylene (PE) pipes. Five PVC pipes from different manufacturers were used and all five pipes gave similar sorption results of trichloroethylene and toluene using the equilibrium sorption test. Sorption was rapid and reached equilibrium within 7-11 days. When different types of gasoline was used, PVC pipes sorbed less than 1.5% of gasoline after 100 days indicating that PVC can resist permeation of gasoline. It was noted that PVC pipes sorbed more premium gasoline than regular- and 10% ethanol gasoline about 0.85 % gain in weight after 90 days. The visualization test confirmed the permeation of the premium gasoline where a moving front of 0.3 mm was observed at 120 days while there was no moving front detected in regularand 10% ethanol gasoline. The results suggested that percent gain in weight of gasoline may be directly proportional to concentration of BTEX (benzene, toluene, ethylbenzene, xylene) in gasoline. In the presence of a solvent such as TCE or toluene, permeation of gasoline through PVC pipes may be enhanced. For PE pipe exposed to pure gasoline and aqueous solution saturated with gasoline, breakthrough of BTEX compounds occurred after 7 and 10 days of exposure. Concentration of individual BTEX compounds in pipe water at stationary state of permeation was over their MCL for both PE pipe exposed to gasoline and aqueous solution saturated with gasoline. Diffusion coefficients were strongly dependent on the concentration of individual BTEX compounds at the outer surface of the pipe wall.

Field of study	Environmental Management	Student's signature
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# NOMENCLATURES

J	=	flux of the diffused contaminant (g/ $m^2 \cdot s^{-1}$ )
D	=	diffusion coefficient (m <sup>2</sup> /s)
С	=	concentration of the diffused contaminant (g/m <sup>3</sup> )
х	=	length in the direction of movement (m)
Q(t)	=	quantity of contaminant permeate through polymer as a function of
		time $(g/m^2)$
S	=	Henry's law constant (= constant) (dimensionless)
Co	=	outer concentration of the contaminant (mg/m <sup>3</sup> )
1	=	thickness of the polymer (m)
t	=	time (s)
$\partial Q/\partial t$	=	permeation rate at steady state
Р	=	permeability coefficient (m <sup>2</sup> /s)
$T_L$	=	Time-lag of Barrer
a	=	activity (dimensionless)
$C_{\rm w}$	=	concentration in water (mg/ L)
$C_{w,m}$	=	maximal solubility in water (mg/L)
$C_v$	=	concentration in vapor phase (mg/ L)
$C_{v,m}$	=	maximal solubility in vapor phase (mg/ L)
$\mathbf{K}_{\mathbf{p}}$	=	soil-water partition coefficient (dimensionless)
K <sub>oc</sub>	=	organic carbon partition coefficient (dimensionless)
$\mathbf{f}_{oc}$	=	fraction of organic carbon in the soil (dimensionless)
$C_{soil}$	=	concentration of chemical in the organic carbon component of soil
		(µg absorbed/ kg organic carbon)
C <sub>water</sub>		the concentration of chemical in water ( $\mu g / kg$ )
X <sub>I</sub>	1	Flory-Huggins interaction parameter (dimensionless)
$\mathbf{V}_1$	=	volume fraction solvent in the swollen polymer (mL)
$V_2$	=	polymer volume fraction (mL)
W%	=	percent weight gain
$T_{g}$	=	glass transition temperature (°C)
PB	=	polybutylene

PE	=	polyethylene
PVC	=	polyvinyl chloride
AC	=	asbestos cement
ABS	=	acrylonitrile-butadiene-styrene
SBR	=	styrene-butadiene-rubber
EDPM	[ =	ethylene-propylene-diene-momomer
BTEX	=	benzene, toluene, ethylbenzene, and xylene
MTBE	E =	methyl <i>tert</i> -butyl ether
TCE	=	trichloroethylene
PCE	=	perchloroethylene or tetrachloroethylene
PEG 4	= 00	polyethylene glycol molecular weight 400
$C_6H_6$	=	benzene
$C_7H_8$	=	toluene
$C_8H_{10}$	=	ethylbenzene
$C_8H_{10}$	=	xylene
CNS	=	central nervous system
MCL	=	maximum contaminant level (mg/L)

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## **CHAPTER I**

### **INTRODUCTION**

### **1.1 MOTIVATION**

A water distribution system in a developed area may comprise of hundreds of kilometers of buried pipes. As a result of pollutants from a leaking underground storage tank or spills from industries, the buried pipes may be close or in contact with the pollutants or contaminated soils. Plastic pipes have been widely used in water distribution since they can resist corrosion when in contact externally with soil and internally from the corrosivity of the water it is conveying. However plastic pipes are made of polymers that may be permeated by certain types of chemicals from contaminated soils (Holsen, 1991a; Goodfellow, 2002; Park, 1991). Incidents of contamination of drinking water due to the permeation of the contaminants through plastic pipes or through the gaskets have been reported. The permeation incidents are typically revealed when customers complain of taste and odor of chemicals in drinking water.

Polybutylene (PB), polyvinyl chloride (PVC) and polyethylene (PE) are some of most common plastic pipes that are concerned with the occurrences of hydrocarbon permeations. In applications where gaskets are used, permeation through the gasket materials have been suspected as the route of permeation. It may be assumed that because of the high flow rate and small surface area of the gasket material in the water main, diffusion of contaminants would be diluted resulting in non detection of the contaminants.

Benzene, toluene, ethyl benzene, and o-, m-, p- xylene (BTEX) are the most common contaminants related to most of permeation incidents with certain number of permeation incidents concerning chlorinated solvents such as trichloroethylene (TCE) and tetrachloroethylene (PCE) (Holsen et al., 1991a). Contamination of drinking water with BTEX compounds is a serious problem since benzene is a known carcinogen, and BTEX compounds have exhibited acute and long toxic effects. Since chlorinated solvents are persistent in the environment and may be carcinogens, there is also much interest in these compounds. Generally, BTEX are present in petroleum products such as gasoline, diesel fuel, and heating oil. One of the most common encountered sources of BTEX contamination is gasoline from leaking underground storage tanks of petroleum stations. In the case of chlorinated solvents, they are present in the contaminated soils of military and industrial sites and at dry cleaners. BTEX and chlorinated solvents can soften PVC pipe resulting in a change in the polymer structure of PVC from glassy polymer of strong cross-liked bonds to rubberized polymer that are more flexible and susceptible to permeation. Most of the previous investigations were conducted using pure solvents with results indicating that individual BTEX compounds will not be soften PVC if the activity of the compound is less than 0.25 and for individual chlorinated solvent at activities of less than 0.1 (Berens, 1985). Most studies have shown that pure individual BTEX compounds with an activity of 1.0 will swell PVC rapidly (Berens, 1985). In the previous studies, the diffusion rates through the amorphous regions of unswollen PE are higher than those through unswollen PVC (Berens, 1985; Vonk, 1986).

Information on the impact of petroleum-based hydrocarbons consisting of a mixture of aromatic compounds and chlorinated hydrocarbons commonly found in the environment on various types of plastic pipes have been inadequately investigated and incomplete with various data gaps. The major purpose of this study is to investigate the permeation of BTEX compounds through polyethylene pipe. The results from the experiment will be applied in the prediction of permeation of these compounds in the real fields.

#### **1.2 OBJECTIVES**

This research goal is to study the permeation of BTEX compounds in gasoline through PVC and PE pipe. The main objectives of this study are:

- To investigate the organic chemical resistance of different manufactures of PVC and PE pipe by conducting equilibrium sorption test experiments.
- 2. To examine the extent of permeation of organic chemicals within PVC pipe by conducting microscopic visualization test experiments.
  - 3. To investigate the permeation of BTEX compounds in gasoline through PE pipe by conducting permeation test experiments.

### **1.3 HYPOTHESES**

Equilibrium sorption test experiments and permeation test experiments can be used to investigate the permeation of petroleum hydrocarbon compounds through plastic pipes and to identify the resistance of plastic pipes to the permeation under the simulated condition. The following hypotheses are proposed:

1. PVC pipe from different manufacturers will give different chemical resistances.

2. PE pipe will be an effective barrier against permeation of BTEX compounds at the low concentrations typically found in contaminated groundwater or soils.

#### **1.4 SCOPES OF WORK**

The scopes of this study are:

1. Equilibrium sorption test experiments will be conducted to investigate the Flory–Huggins interaction parameter,  $X_I$  which can be used as a basis for identifying the chemical resistance of PVC and PE pipes.

2. Visualization diffusion experiments will be conducted to visualize the transportation of chemicals into PVC pipes.

3. Permeation test experiments will be carried out in such a way to determine breakthrough time of petroleum hydrocarbons through PE pipes, the permeation rate at steady state (dQ/dt), permeability coefficient, P and diffusion coefficient, D. The experiment will be directed under the following conditions: PE pipes exposed to a pure gasoline, aqueous solution saturated with gasoline.

4. Five different manufacturers of PVC pipe and one PE pipe will be used in the equilibrium sorption test experiments. One type or manufacturer of PE pipe will be used in permeation test experiments.

## **1.5 ADVANTAGES OF THIS WORK**

This work is expected to provide:

• Information to increase the water industry's understanding about permeation of BTEX compounds through PE pipes under conditions of soil contamination that are typically encountered in the field.

- The basis knowledge to predict the permeation of other contaminants through plastic pipes.
- Information that can assist us to prevent permeation of contaminants.

# 1.6 ORGANIZATION OF THESIS

The first chapter of the thesis provides the motivation and objectives for this study. The second chapter is a literature review highlighting the work that is already done and the data gaps. Chapter 3 covers the methodology of this study and chapter 4 provides the results and discussions obtained from the experiments. The final chapter is a conclusion and recommendation from this study.



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#### **CHAPTER II**

### **BACKGROUND AND LITERATURE REVIEW**

#### **2.1 INTRODUCTION**

Over 40 years, plastic pipes have been widely used in potable water distribution systems since they offer several advantages over other piping materials such as steel pipes. These advantages include excellent chemical resistance, light weight, low cost, and easy installation. A survey conducted by the Plastic Pipes and Fittings Association showed that polyvinyl chloride (PVC) is the most widely used pipe material and polyethylene (PE) is the second (Polyethylene Water Service, 2004).

Although plastic pipes provide excellent resistance to corrosion when in contact externally with subsurface soils and internally from the corrosivity of the water it is conveying, plastic pipes may be permeated by certain types of organic contaminants in the presence of contaminated soils and groundwater. Over 100 incidents concerning contaminated drinking water due to permeation of contaminants through plastic pipes have been reported in the U.S. The materials involved permeation incidents in U.S. water system are presented in Figure 2.1 (Holsen et al., 1991a). Polybutylene (PB), PE, and PVC pipes were some of most common pipe materials involved in the permeation incidents.



**Figure 2.1** Pipe materials involved in permeation incidents in U.S. water system (Holsen et al., 1991a).

(ABS = Acrylonitrile-butadiene-styrene, AC = Asbestos Cement

PB = Polybutylene, PE = Polyethylene, PVC = Polyvinyl chloride)

Holsen and co-workers (1991a) found that PB was more susceptible to permeation than PE and PVC respectively. The occurrences of permeation incident were revealed by customers' complaints of odor and taste of contaminants in their potable water (Goodfellow, 2002 and Holsen, 1991a).

Figure 2.2 (Holsen, et al., 1991a) shows the contaminants involved in permeation incidents in U.S. water system. Petroleum products such as gasoline, diesel fuel, and engine oil were the major contaminants involved in the permeation incidents while a small numbers of the permeation involved chlorinated solvents such as trichloroethylene (TCE) and perchloroethylene (PCE).



**Figure 2.2** Contaminants involved permeation incidents in U.S. water system (Holsen et al., 1991a).

In United Kingdom, eleven incidents of drinking water contamination as a result of permeation of organic chemicals through plastic pipes were reported to CIRS (Chemical Incident Response Service), UK between 1998 - 1999. The types of chemicals and sources of contamination involved in these incidents are presented in Table 2.1 (Goodfellow, et al., 2002).

permeation (Goodfellow et al., 2002)

Date of incident	Chemical involved	Cause of contamination
May 1998	Trichloroethylene 1,1,1 trichloroethane	Spill of solvent onto land above water pipes within an industrial estate
July 1998	Petrol	Migration of spill from nearby petrol filing station
July 1998	Domestic heating oil	Longstanding leaks from domestic heating oil pipe which were located near plastic water pipes
October 1998	Diesel PAHs	Diesel spill on a building site near a PVC pipe leading to pipe damage and ingress of contamination
November 1998	Xylene Methylbenzene	Spill in a garden leading to permeation of plastic water pipes
April 1999	Xylene Alkylenbenzene Naphthalenes	Unknown source of ground contamination leading to permeation of plastic water pipes
July 1999	Petrols Benzene	Petrol spill permeated plastic water pipes Kerozene spill permeated plastic water pipes
September 1999	Kerozene	Kerozene leak from heating oil pipes permeating plastic water pipes
November 1999	Benzene and toluene	Leak from petrol station leading to permeation and contamination of water supplied to a block of flats
December 1999	Polyaromatic hydrocarbons Dissolved hydrocarbons	Leak from heating oil pipe in the same trench as a pre-existing plastic pipe.

Gaskets are materials used in pipe connections - to join both iron and plastic pipes. More than 90% of gaskets used in water distribution system are styrenebutadiene-rubber (SBR). Other types of gasket materials include neoprene (chlorinated rubber), ethylene-propylene-diene-momomer (EPDM), nitrile rubbers and fluorinated rubbers. In many cases where gaskets are used, permeation through the gasket materials have been suspected as the route of permeation. It may be assumed that because of the high flow rates and small surface area of the gasket material in contact with the water, diffusion of contaminants would be diluted resulting in non detection of the contaminants.

### 2.2 PETROLEUM BASED HYDROCARBONS

Many cases of permeation incidents involve petroleum hydrocarbons (Holsen, 1991a; Goodfellow, 2002,). The group of contaminants that is of concern and is regulated consists of benzene, toluene, ethylbenzene, and three isomers of xylene (o-, p-, m-). They are commonly refered to as BTEX. They are presented in petroleum products such as gasoline, diesel fuel, and heating oil (Christensen and Elton, 1996). The presence of BTEX compounds in drinking water are considered to be a serious problem since benezene is a known carcinogen and the other compounds are known to be hazardous to human health. Previous investigations have indicated that PVC softens and swells when exposed or in contact with individual BTEX compounds (Berens, 1985; Vonk, 1986; Parker, 1986; Goodfellow, 2002). Swollen PVC is the result of a change in the PVC polymer structure from glassy polymer of strong cross-liked bonds to rubberized polymer which is more flexible and susceptible to permeation. Most of the previous investigations were conducted using pure solvents with PVC films. These researchers found that individual BTEX compounds will not soften PVC if the activity of the compound is less than 0.25 while pure individual BTEX compounds with an activity of 1.0 soften and swell PVC rapidly (Berens, 1985 and Vonk, 1986)

### 2.2.1 Gasoline

One of the most common sources of BTEX contamination is gasoline from leaking underground storage tanks of petroleum stations (Holsen et al, 1991a). The major components of gasoline are alkanes, monocycloalkanes, dicycloalkanes, akylbenzenes, indanes, tetralins, naphthalenes and some oxygenated alcohol additives. The specific composition varies depending on the source of the petroleum as well as the production method (e.g., distillation or fractionation, thermal and catalytic cracking, re-forming, isomerization) (Seagren et al., 2002). Generally, 18% of standard gasoline blend is made up of BTEX. The percent weight composition of BTEX in gasoline is presented in Table 2.2.

 Table 2.2
 BTEX compounds in gasoline

Compound	Percent weight (%)		
Benzene	1.73		
Toluene	9.51		
Ethylbenzene	1.61		
m-, p-xylene	5.95		
o-xylene	2.33		

### 2.2.2 Fate and transport

Upon release in the subsurface environment, gasoline will partition accordingly – some will volatilize, dissolve into the groundwater and sorbed onto soils as presented in Figure 2.3. However, BTEX compounds are not as strongly sorbed to the soil particles than aliphatic components and are more likely to contaminate groundwater. Similarly, comparison of water solubilities of different compounds in gasoline indicates that BTEX compounds are highly soluble and are likely to be dissolved in groundwater. Table 2.3 shows the saturated concentrations of BTEX compounds in water. The fate and transport of BTEX compounds are influenced by their physical-chemical properties. A summary of the physicalchemical of BTEX compounds are presented in Table 2.4.



**Figure 2.3** Distribution of gasoline in the subsurface environment (Christensen and Elton, 1996)

Compound	Aqueous conc. (mg/L)
Benzene	42.6
Toluene	69.4
Ethylbenzene	3.2

m-, p-xylene

o-xylene

 Table 2.3 Saturated concentrations of BTEX compounds (Cline et al., 1991)

**Table 2.4** Physical-chemical properties of BTEX compounds (Christensen and Elton, 1996)

11.4

5.6

	Benzene	Toluene	Ethyl	m-	0-	p-
			benzene	Xylene	Xylene	Xylene
Chemical formula	C <sub>6</sub> H <sub>6</sub>	C <sub>7</sub> H <sub>8</sub>	C <sub>8</sub> H <sub>10</sub>			
Molecular weight	78	92	106	106	106	106
Water solubility [mg/L]	1 <mark>7</mark> 00	515	152	-	175	198
Vapor pressure (at 20 °C) [mm Hg]	95.2	28.4	9.5	-	6.6	-
Specific density (at 20 °C)	0.8787	0.8669	0.8670	0.8642	0.8802	0.8610
Octanol-water partition coeff. (at $20^{\circ}$ C) [log K <sub>ow</sub> ]	2.13	2.69	3.15	3.20	2.77	3.15
Henry's law constant (at 25 °C) [kPa*m <sup>3</sup> /mole]	0.55	0.67	0.80	0.70	0.50	0.80
Soil mobility	Very	High	Moder-	Moder-	Moder-	Moder-
	high		ate	ate	ate	ate
Maximum contaminant level (MCL) [mg/L]	0.005	ыны	0.7	10	10	10

## 2.2.3 Health effects

Exposure of drinking water contaminated with BTEX compounds can result in potential health effects. The acute and chronic effects of individual BTEX compound are presented as follow:

Benzene (U.S. E.P.A, 2004; Technical Factsheet on: Benzene)

Acute effect: Benzene has the potential to cause central nervous system (CNS) effects and death from short-term exposure at level above MCL.

Chronic effect: Benzene has the potential to cause chromosomal damage from long- term exposure at levels above the MCL.

Benzene has the potential to cause cancer from a lifetime exposure at levels above the MCL.

Toluene (U.S. E.P.A, 2004; Technical Factsheet on: Toluene)

Acute effect: Toluene has the potential to cause low oral toxicity to central nervous system, fatigue, nausea, weakness, confusion from short-term exposure at level above the MCL.

Chronic effect: Toluene has potential to cause spasms, tremors, imbalance, impairment of speech, hearing, vision, memory, coordination; liver and kidney damage from long-term exposure at levels above the MCL.

Ethylbenzene (U.S. E.P.A, 2004; Technical Factsheet on: Ethylbenzene)

Acute effect: Ethylbenzene has potential to cause drowsiness, fatigue, headache, mild eye and respiratory irritation from short-term exposures at levels above the MCL.

Chronic effect: Ethylbenzene has potential to cause damage to the liver, kidneys, central nervous system and eyes in people from long-term exposure at levels above the MCL.

Xylenes (U.S. E.P.A, 2004; Technical Factsheet on: Xylene)

Acute effect: Xylene has potential to cause disturbances in the central nervous system, such as changes in cognitive abilities, balance, and coordination from short-term exposures at levels above the MCL.

Chronic effect: Xylene has potential to cause damage to the central nervous system, liver and kidneys from long-term exposure at levels above the MCL.

There is inadequate evidence to indicate that toluene, ethylbenzene and xylenes have the potential to cause cancer from lifetime exposure in drinking water.

#### **2.3 POLYMERS**

#### 2.3.1 Polyvinyl Chloride (PVC)

Polyvinyl chloride (PVC) is prepared by the polymerization of vinyl chloride monomers combining with other polymer and compounding ingredients (Haward, 1973). The structure of PVC is presented in Figure 2.4.



Figure 2.4 Structure of polyvinyl chloride (PVC)

PVC consists of long polymer chains which are packed and arranged in the configuration of amorphous structure. The glass transition temperature, Tg of PVC is 65 °C (Polymers, 2004). At room temperature the structure of PVC acts as glassy polymer whose chains are a strong cross-liked bond resulting in very low flexibility. Generally, PVC is resistant to many alcohols, fats, oils, aromatic free petrol, and almost all common corroding agents including inorganic acids, alkalis and salts. However, PVC may not be used with esters, ketones, ethers and aromatic or chlorinated hydrocarbons since it can absorb these substances leading to swelling and a decrease in tensile strength.

#### 2.3.2 Polyethylene (PE)

PE is prepared by the polymerization of ethylene monomers (Haward, 1973). The structure of PE is presented in Figure 2.5.



Figure 2.5 Structure of polyethylene (PE)

PE is made of long polymer chains which are packed and arranged in the configuration of semi-crystalline structure. Glass transition temperature,  $T_g$  of PE is - 125 °C (Polymers, 2004). At room temperature, the structure of PE behaves like a rubber-polymer whose chains have a high mobility and flexibility (Vonk et al., 1984). PE is resistant to inorganic salts solutions, weak acids, strong organic acids, strong alkaline solutions, aliphatic hydrocarbons. PE cannot resist attacks of esters, ketones, ethers, aromatic hydrocarbons, mineral oil, light naphtha, fuel mixture, unsaturated chlorinated hydrocarbons, and turpentine. The glass transition temperature and the structure of several polymers are illustrated in Table 2.5.

Polymer	T <sub>g</sub> (°C)	Structure
Polyvinyl chloride (PVC)	65	amorphous
Polystyrene (PS)	100	amorphous
Polyethylene, low density	-125	semi crystalline
Polyethylene, high density	-125	semi crystalline
Polypropylene	-10	semi crystalline
Nylon 66	50	semi crystalline

 Table 2.5 Glass transition temperatures, Tg for several polymers (Polymers, 2004)

The differences in polymer structures of PVC and PE affect the diffusion of chemicals. For PVC, contaminant diffuses through the free small volume between the strong cross-linked bonds of polymer chains while the diffusion of PE occurs though the highly mobile amorphous areas. Previous investigations indicate that PE pipes are more susceptible to permeation than PVC pipes (Vonk, 1986). However, permeation of PE pipes might not occur when PE pipes are exposed to polar organic compounds, such as pesticides, complex aromatic compounds and methanol even though they absorbed onto the outer surface of the pipe wall (Goodfellow et al., 2002). Tiganis (2001) found that the migration of pesticide constituents and their associated solvents, through PE pipe did not occur. This work suggested that the concentration of solvents was not enough to cause permeation through PE pipe.

#### 2.4 PHYSICOCHEMICAL THEORY OF PERMEATION

#### 2.4.1 Permeation process

Permeation can be defined as the process in which the contaminant moves or diffuses through the polymer. Permeation of contaminant through polymers occurs through two physical processes - partitioning at the surface of the polymers and diffusion within the polymer (Vonk, 1986; Vonk, 1984). The mechanism of permeation occurs by a three-step process as described by Vonk (1984). Firstly, contaminants partition or dissolve between the pore water or soil gas and the outer surface of polymers. Solubility of contaminant in polymer is dependent on the cohesive energy (the energy that holds liquid or solid molecules together) of the contaminant and the polymer. If the contaminant and polymer have the similar cohesive energies, the likelihood that contaminant will interact with polymer is greater. Next, the contaminants diffuse through the plastic pipe via pore structure of polymers. Finally, contaminants partition between the inner surface of polymers and water inside the pipe. Both partitioning steps may be assumed to follow Henry's law, wherein the concentration of contaminant in the receiving phase is equal to the concentration in transmitting phase multiplied by a constant (i.e., Henry's law constant) which is a characteristic of contaminant and the two phases. The three stage process of the permeation is shown in Figure 2.6.



Figure 2.6 Three-stage process of permeation

The mathematics of diffusion has been thoroughly described by Crank (Crank and Park, 1968). The contaminants move through the polymer from the influence of a concentration gradient within the thickness of the polymer. The diffusion process can be described by Fick's first and second laws based on the assumption that there is no interaction between the contaminants and the polymers. Fick's first law is given by:

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where

 $J = \text{flux of the diffused contaminant } (g/\text{ m}^2 \cdot \text{ s}^{-1})$   $C = \text{concentration of the diffused contaminant } (g/\text{m}^3)$   $D = \text{diffusion coefficient } (\text{m}^2/\text{s})$  x = length in the direction of movement (m)

The diffusion coefficient, D is a parameter expressing the transfer rate of a substance by random molecular motion. Mathematically, it is defined as the specific transfer rate (the transfer rate per unit cross-sectional area) under a unit driving concentration gradient. Fick's second law is given by:

$$\frac{\partial C}{\partial x} = D \frac{\partial^2 C}{\partial x^2} \qquad (2.2)$$

The expression of Henry's and Fick's laws were combined by Barrer (Crank, 1975) to derive an expression to quantify contaminants permeating a polymer as a function of time, Q(t) (g/m<sup>2</sup>/sec),. The assumptions made include the concentrations of the contaminants exposed to the outer surface of the polymer wall remain constant, the initial concentration of a diffused contaminant in the polymer is zero and the concentration of the contaminant at the other end of the polymer is kept at zero. The derived expression is given by:

$$Q_{t} = l \cdot S \cdot C_{o} \cdot \left\{ \frac{D \cdot t}{l^{2}} - \frac{1}{6} - \frac{2}{\pi^{2}} \cdot \sum_{n=1}^{\infty} (1)^{n} \exp(-D \cdot n^{2} \cdot \pi^{2} \cdot t) \right\} \dots \dots (2.3)$$

where

S = the dimensionless Henry's law constant (= constant)

 $C_o$  = the outer concentration of the contaminant (mg/m<sup>3</sup>)

D = diffusion coefficient (m<sup>2</sup>/s)

- l = the thickness of the polymer (m)
- t = time (seconds)

The exponential term can be negligible for infinite time and the total mass permeated per unit area will approach linearity as shown in Figure 2.7.

The rate of permeation at steady state  $(g/m^2/sec)$  is determined from the slope of the line in Figure 2.7 and is expressed as:

$$\frac{\partial Q_t}{\partial t} = C_0 \cdot \frac{P}{l} \quad \dots \tag{2.5}$$

where P is the permeability coefficient  $(m^2/s)$  and is given by



Figure 2.7 Permeation of an organic chemical through a polymer

The non linear increase of Q(t) in the initial part of the curve is attributed to the diffusion process in non-stationary state. When the diffusion process reaches stationary state, the Q(t) will approach linearity. The dotted line in Figure 2.7 describes the non-stationary state of equation 2.5. The intersect of the line with the xaxis (time) denoted by  $T_L$  is known as the time-lag of Barrer.  $T_L$  is given by:

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It appears from equation 2.5 and equation 2.7 that the rate of change of the total quantity of the contaminant with time (dQ(t)/dt) is indirectly proportional to the thickness of the polymer. The time lag,  $T_L$  which indicates the time required to observe a discernible permeation is directly proportional to the square of the thickness of the polymer. Doubling the thickness of the polymer will quadruple the time-lag. Therefore, using a thicker water pipe as compared to acceptable nominal thickness water pipe in areas of contamination may reduce the risk of the occurrences of permeation incident.

The diffusion coefficient, D is strongly dependent on the outer concentration of the contaminant, Co (check this fact – diffusion coefficient is a constant and is only dependent on the material not on the concentration – there is already the concentration gradient term). The diffusion coefficient is a function of polymer chains, molecular size of the contaminant, and the shape of the contaminant (Vonk, 1986). The longer the chain length of the contaminant, the slower is the velocity of diffused contaminant through the polymer due to increasing entanglement of the large molecule (Crank et al., 1968). Berens (1982) found that diffusion coefficient of molecules having spherical shape in PVC decreased exponentially with molecular diameter. Diffusion coefficient of elongated molecules was smaller than that of spherical molecules. Table 2.6 provides the example of diffusion coefficient, D (m<sup>2</sup>/sec) of toluene and trichloroethylene for polyethylene pipe (PE) and polyvinyl chloride pipe (PVC).

จพัญลงกร	D (m <sup>2</sup> /day)		
Polymer	Toluene	TCE	
Polyethylene (PE) LDPE	$3.5 \times 10^{-8}$	$5.1 \times 10^{-8}$	
HDPE	-	$1.9 \times 10^{-8}$	
Polyvinyl Chloride	$7.7 \times 10^{-24}$	-	

Table 2.6 Diffusion coefficients of Toluene and Trichloroethylene

Permeation of contaminants through PE pipes can be described by the two laws of Fick even in the presence of high concentrations of contaminants in the surrounding environment (Vonk, 1986). In the case of PVC pipes, permeation of organic compounds through PVC pipe under the condition of low concentration of the contaminants has been shown to follow Fick's laws. However the permeation cannot be described by Fick's law under the condition of high concentration of contaminants. Under high concentration of contaminants, the contaminants interact at the outside surface of the pipe material and soften the polymer and converting the polymer structure from a glassy polymer of high rigid structure to a rubbery polymer of flexible structure and accelerating permeation. This case is known as Case II diffusion. The "moving front", i.e., the interface of the softened polymer and the nonsoftened polymer progresses into the polymer material as shown in Figure 2.8 (Vonk et al., 1984).



Figure 2.8 Microscopic visualization of moving Front

For Case II diffusion, the discernible permeation cannot be detected until the softened polymer reaches into the inside wall of polymer. After this point, the permeation will appear suddenly and the rate of permeation increases rapidly at a constant rate as presented in Figure 2.9.



Figure 2.9 Case II diffusion in pipe

### 2.4.2 Activity and permeation

The permeation of contaminants through plastic pipes is based on their activity rather than their concentration since the transport of contaminants occurs from one phase to another phase until the equilibrium is achieved. The transport of contaminants is related to their solubility (Berens, 1985 and Vonk, 1986). Activity is a dimensionless quantity; and can be calculated in the terms of the ratio of the concentration of the compound in aqueous or vapor phase to its maximal concentration in aqueous or vapor phase as expressed in equation 2.8

(2.8)  $a = C_w / C_{w, m}$  or  $a = C_v / C_{v, m}$  (2.8)

where

a = activity
C<sub>w</sub> = concentration in water (mg/L)
C<sub>w, m</sub> = maximal solubility in water (mg/L)
C<sub>v</sub> = concentration in vapor phase (mg/L)
C<sub>v, m</sub> = maximal solubility in vapor phase (mg/L)

Chlorinated hydrocarbon, aromatic hydrocarbon, and some anilines can strongly soften or swollen PVC. At an activity of less than 0.1 the permeation of these compounds through PVC pipes follows Fickian diffusion (Vonk 1986). For organic compounds which soften PVC to lesser extent such as BTEX, Fickian diffusion can be used to describe the mechanism of permeation for an activity of less than 0.25. Under these conditions (be more specific – activity of < 0.25), PVC pipe can be used as an effective barrier against the permeation of these compounds (Vonk 1986).

## 2.4.3 Flory-Huggins Interaction Parameter

Case II diffusion is used to describe the diffusion of a contaminant in PVC. Contaminants having high activity (>0.5) can soften and swell PVC rapidly. Consequently, PVC is converted from glassy polymer to rubbery polymer resulting in an increase in the permeability of PVC. The ability of a contaminant to rubberize PVC can be indicated by the Flory-Huggins Interaction Parameter ( $X_I$ ) (Jenkins et al., 1985). A lower value of  $X_I$  indicates a relatively high swelling tendency by the contaminant causing Case II diffusion in PVC. The Flory-Huggins equation (Berens, 1985) is used to describe the equilibrium solubility of organic solvents in polymers as a function of solvents activity as given in equation 2.11.

$$\ln(a) = \ln(V_1) + V_2 + X_1 (V_2)^2 \dots (2.11)$$

where

а	= activity of solvent - ratio of the partial pressure P
	of solvent to its saturated vapor pressure
$V_1$	= volume fraction of solvent in swollen polymer
$V_2$	= polymer volume fraction $(V_2 = 1 - V_1)$
X <sub>I</sub>	= Flory–Huggins interaction parameter which
	characterizes the affinity between the solvent
	and the polymer

Volume fraction solvent in the swollen polymer,  $V_1$  can be calculated by using the relation between the densities of the solvent ( $d_1$ ) and the polymer ( $d_2$ ), and the percent weight gain (W%) as expressed in equation 2.12.

$$W\% = \frac{100V_1d_1}{(1-V_1)d_2}....(2.12)$$

The interaction parameter for a standard mixture of organics, or pure of compounds can be served as identifying and certifying the resistance of PVC pipe to contaminants. Berens (1985) found that at room temperature PVC would only be softened by pure liquid solvent at an  $X_I$  value equal to less than 1. For a strong swelling agent with  $X_I$  equal to zero, PVC would only be softened at an activity greater than 0.5. The less aggressive solvents ( $X_I$  values is in the range of 0.5-1.0), require activity around 0.75 or higher to cause softening or swelling. The aromatic hydrocarbon, chlorinated solvents are strong swelling agent for PVC, having  $X_I$  in the

than 1. The  $X_I$  of organic solvents determined is illustrated in Table 2.7.

Compound	XI	Compound	XI
Methylene chloride	< 0.53	Tetrachloroethylene	1.17
Carbon tetrachloride	0.64	1,2-dichloropropane	0.68
1,1-dichloroethane	< 0.68	Benzene	0.83
1,2-dichloroethane	< 0.55	Toluene	0.80
1,1,1-trichloroethane	0.85	Xylene	0.88
1,1,2-trichloroethane	< 0.56	Acetone	0.61
Trichloroethylene	0.88	Vinylchloride	0.98

Table 2.7 X<sub>I</sub> for PVC- organic liquid system (Berens, 1985)

## 2.4.4 Impact of soil type on permeation

The rate permeation of contaminants through plastic pipes is affected by soil types and their textures. Soils particles can sorb contaminants resulting in decreasing concentrations and mobility of the contaminants. Previous studies have found that the rate of contaminants permeation through plastic pipe was controlled by the concentrations of contaminants in the soil pore space (reference). Plastic pipes in soil with high organic carbon content would be permeated more slowly than plastic pipes in soils of low organic carbon content (Holsen et al., 1991b).

The amount of contaminant sorbed by soil can be estimated by equation 2.9 (Michael et al., 2001).

$$X = K_p C.....(2.9)$$

where  $X = \text{concentration of contaminant in soil } (\mu g / kg)$ C = concentration of contaminant in water ( $\mu g / kg$ )

The soil-water partition coefficient,  $K_p$  indicates the tendency of the chemical to be sorbed by soil. If partitioning into organic matter is the dominating mechanism, the partition coefficient  $K_p$  can be normalized with respect to the organic carbon fraction present in the soil, the organic carbon partition coefficient (dimensionless),  $K_{oc}$  is given by (Michael et al., 2001):

 $K_p = K_{oc} \cdot f_{oc}$ .....(2.10)

where  $f_{oc}$  = fraction of organic carbon in the soil (dimensionless)

#### 2.4.5 Impact of pipe additives

In most pipe materials, additives are added to polymer granulates to modify the properties of polymer. These additives include: antistatic agents are to increase the surface conductivity by attracting moisture from the air, stabilizers to prevent deterioration during processing; and heat stabilizers to prevent the formation of HCl during processing. Plasticizers are low molecular weight molecule that are added to reduce the glass transition temperature (Tg) and improve the formability and plastic properties. Plasticizers are particularly important for PVC with T<sub>g</sub> above room temperature. The strength and rigidity of polymer can be improved by adding by reinforcement materials such as glass and/or carbon fibers into polymer matrix. There is no information on the effect of additives on the permeation process because most of the previous investigations were conducted using pure polymer powders or sheets pressed or made from the pure powder. It was found that equilibrium sorption and sorption rate of acetone, toluene and TCE in PVC pipes and sheets pressed from a commercial pipe powder compound (unspecified additives) were similar to the results for pure PVC (Berens, 1985).

#### **2.5 PREVIOUS INVESTIGATIONS**

The effects of organic compounds on plastic pipes have been studied since 1985. Holsen et al. (1991a) studied contamination of potable water by permeation of hydrocarbons through plastic pipe. In this study, a set of sampling instructions for reporting and documenting permeation were sent to about 1,200 water utilities in California in July 1986. Seven permeation incidents were reported between July 1986 and December 1987. They found that nearly all permeation incidents were related to gasoline (five out seven incidents) from gross soil contamination in the area surrounding the plastic pipe. Polybutylene (PB) was found to be the major pipe material with a high frequency of permeation, followed by pipes made of polyethylene (PE), and polyvinyl chloride (PVC). The permeation incidents were located at industrial sites and residential neighborhoods with gas stations.
Berens (1985) studied the permeation of organic solvents by varying the concentration of solvents in contact with PVC powders, films, and sheets by conducting gravimetric sorption experiments. He found that for molecules which are spherical, the diffusion coefficients, D in PVC increase exponentially with the molecular diameters. For elongated molecules, the diffusion coefficient, D is larger than that compared with the diffusion coefficient, D of the spherical molecule. Beren concluded that at an organic chemical activity of less than 0.25, sorption of these compound followed Fick's laws. Organic chemical activity is defined as ratio of the concentration of the contaminant in aqueous or vapor phase to its maximal concentration in aqueous or vapor phase. Beren estimated that toluene with activity between 0 and 0.25 took centuries to reach steady-state permeation whereas toluene with a high activity (> 0.25) has lag time of only 48 hours indicating that PVC will not be a suitable barrier against permeation if exposed to toluene.

Vonks (1986) studied the permeation of several organic compounds through PE pipe. The laboratory experiment were carried out by exposing high density polyethylene (HDPE), low density polyethylene (LDPE) and PVC pipe to contaminated soils in both simulated unsaturated soil conditions and to vapor phase. In the case of HDPE and LDPE pipes, the results showed that all organic compounds took 60 - 400 days to reach to a constant weight or concentration. Lipophilic organic compounds such as aromatic compounds and chlorinated hydrocarbons have greater permeability than polar organic compounds such as ketones, alcohols and phenol. For PVC pipe, Vonks concluded that permeation cannot be expected to occur for alcohol, aliphatic hydrocarbons, and organic acid and that permeation of benzene and alkylated benzene with activities > 0.25 and chlorinated hydrocarbons, ketones, nitrobenzene and anilines with activities > 0.1 followed Fick's laws.

Park et al. (1991) conducted pipe-bottle experiments to study the permeation of organic chemicals at varying activities through polyethylene pipe (PB) and gasket material. Their results indicated that the addition of a readily permeable organic chemical to a mixture of relatively nonpermeable organic chemical would enhance the rate of permeation. Additionally, organic chemicals were found to be more permeable in gasket materials than in PB pipe. But only a small number of permeation incidents involving gasket material have been reported. This is true because gaskets are used in

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the water main which has high flow rate which may diluted the chemical resulting in the permeation being undetected.

Cassaday et al. (1983) conducting limited experiments exposing gaskets of unspecified types to toluene, hexane and 1,1,1-trichloroethane found that gaskets were more permeable that any pipe material.

Holsen et al. (1991b) buried polybutylene pipes in both unsaturated and saturated soils contaminated with organic chemicals in order to investigate the effect of soils on permeation. They found that the rate of permeation is strongly influenced by organic carbon content. Soil with high organic carbon content could reduce the rate of permeation of organic chemicals through PB pipes and could increase the time needed to permeate a pipe. Additionally, the permeability of organic compound could be limited by putting native soils having a high organic content as a backfilled surrounding the pipe.

Parker et al. (1994) examined the oftening of rigid PVC by an aqueous solution of strong PVC swelling agents. They found that methylene chloride with an activity of about 0.6 resulted in the rigid PVC becoming rubbery and a solution containing three PVC swelling agents (methylene chloride, 1,1,2- trichloroethane, and 1,2- dichloroethane) soften PVC rapidly (within 2 days). They further studied the softening of rigid PVC by using a combined solution containing 18 PVC swelling agents, each solute with an activity about 0.05. They found that the combined solution soften the PVC rapidly (Parker et al., 1995).

#### 2.6 SUMMARY

Although many cases of permeation of single organic compound through plastic pipes such as PB, PVC, and PE pipes have been investigated in previous works, there are inadequate investigation and incomplete with various data gaps for permeation of organic compounds in the presence of multiple solvents which are typically encountered in the real contaminated areas. In the further study, the conditions of plastic pipes buried in contaminated soil or ground water with petroleum based hydrocarbons that have BTEX compounds as the basis components should be simulated and the permeation of these compounds through plastic pipes should be investigated. The results from this investigation will fulfill data gaps for permeation behavior of BTEX compounds in mixing form and will provide water utility's understanding about types of piping material that can prevent permeation incidents.



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#### **CHAPTER III**

#### METHODOLOGY

#### **3.1 MATERIALS**

#### 3.1.1 Pipe materials

Nine PVC pipes of different manufactures and one PE pipe were purchased from local stores in city of Ames, Iowa, USA and named as PVC A to PVC I and PE A. In this study, five PVC pipes and the PE pipe were selected for the equilibrium sorption test: PVC B, PVC C, PVC D, PVC F, PVC H, and PE A. The PE pipe was used for permeation test. The five PVC pipes were of Schedule 40 with diameters of 1 or 1.25 inches. The density, wall thickness, and specifications of the pipes used are listed in Table 3.1.

Table 3.1 Specifications of pipes

Pipe ID	Density (g/mL)	Wall thickness (mm)	Specifications
PVC B	1.4577	3.65	1.25" diameter Manufacturer - J-M Pipe Marking on Pipe; PVC 1120 SCH 40 370 PSI @ 73°F NSF-PW ASTMD-1785
PVC C	1.4504	3.55	1" diameter Manufacturer - Charlotte Pipe Marking on Pipe; S/W 04010 PVC 1120 SCH. 40 PR 450 PSI@ 23°C ASTM D-1785
PVC D	1.4461	3.60	1" diameter Manufacturer - J-M Pipe Marking on Pipe; PVC 1120 SCH 40 450 PSI @ 73°F NSF PW ASTMD-1785 JM90
PVC F	1.4591	4.00	1 <sup>1</sup> /4" diameter Manufacturer - Bristolpipe Marking on Pipe; PVC 1120 SCH 40 370 PSI @ 73°F ASTM-D-1785
PVC H	1.4604	3.72	1 <sup>1</sup> / <sub>4</sub> " diameter Manufacturer - Silver-Line® Marking on Pipe; PVC-1120 SCH. 40 PR. 370 PSI @ 73°F [ASTM D-1785 NSF-pw]
PE -A	0.9451	3.30	1" diameter Manufacturer - Endot Marking on Pipe; 160PSI@73F ENDO-POLY SIDR-9 IPS NSF PW PE3408 ASTM-D2239-81

#### 3.1.2 Chemicals

Chemicals used in the tests were commercial grade benzene, toluene, ethylbenzene, mixed xylene, trichloroethylene (TCE), polyethylene glycol 400 (PEG 400), and methyl *tert*-butyl ether (MTBE). The chemicals were purchased from Fisher Scientific International Inc, USA. Three types of gasoline, including regular gasoline, premium gasoline, and 10% ETOH gasoline were also used in this experiment. All three types of gasoline were purchased from Central Store of Iowa State University, USA and kept in safety containers.

Chemical used in microscopic visualization test were epoxy hardener, epoxy resin purchased from LECO Corporation, USA and diamond compound purchased from WENDT Dunnington, USA.

Saturated solutions of gasoline were prepared by adding gasoline to distilled water in an approximate ratio of 50 mL to 1 L in a glass container. The solution was constantly stirred to maximize solubilization and saturation of the water with gasoline. The concentrations of individual BTEX compounds in regular-, premium-, 10% ethanol gasoline and aqueous solution saturated with gasoline used in this study are presented in Table 3.2

Solvent	Concentration (mg/L)					
Solvent	Benzene	Toluene	Ethyl- benzene	m-xylene	o + p- xylene	
Regular gasoline	17,310	43,710	8,450	21,690	20,470	
Premium gasoline	20,110	70,980	11,070	28,260	27,540	
10% Ethanol gasoline	15,650	40,860	7,810	20,320	19,160	
Aqueous solution saturated with gasoline	57.02	53.49	2.70	6.56	7.35	

Table 3.2 Concentration of BTEX compounds in gasoline

#### **3.2 EXPERIMENTAL PROCEDURES**

To study the permeation of BTEX compounds in gasoline through PVC and PE pipe, the resistance of the pipe to attack by solvents, the movement of solvents within pipe, and the quantity of solvents permeated through the pipe were investigated using the equilibrium sorption test, microscopic visualization test and pipe-bottle permeation test. The experiments were conducted in three parts as follow:

Part 1: Solvents resistance of PVC/PE pipes were investigated using equilibrium sorption test. The parameters determined or estimated from this study included:

- Percent weight gain (W%)
- Flory-Huggins interaction parameter (X<sub>I</sub>)
- Diffusion coefficient,  $D(m^2/day)$

Part 2: The extent of permeation of solvents within PVC pipes were investigated using microscopic visualization test.

Part 3: Permeation of solvents through PE pipe was investigated using pipebottle permeation test. The parameters determined or estimated from this experiment included:

- Time-lag intercept,  $T_L$  (day)
- Polymer-solvent partition coefficient (dimensionless)
- Permeability coefficient,  $P(m^2/day)$
- Diffusion coefficient,  $D(m^2/day)$

#### **3.2.1** Equilibrium sorption test

For the equilibrium sorption test, specimens of pipe were cut to a length of 1 cm with a miter saw. The specimens were washed with detergent, rinsed with tap water and distilled water, placed on a paper towel to air dry, and weighed using an analytical balance. The volume was determined by a water displacement method using an overflow can aluminum purchased from Fisher Scientific International Inc, Vernon Hills, IL, USA. Specimens were immersed in 50 mL of the test solvent in a 120 mL glass jar with a Teflon-lined lid and the glass jars were kept at the  $23 \pm 1^{\circ}$ C.

At various times, the specimens were removed from the solvents, placed on paper towels, wiped dry, and allowed to air dry for thirty seconds before weighing. In all steps, the specimens were handled with stainless steel forceps. Pipe specimens were considered to have reached equilibrium absorption when three consecutive weighings differed by no more than 20 mg.

#### **3.2.1.1 Data analysis**

Flory-Huggins interaction parameter for the polymer-solvent sorption test was determined by using the weight gain of the absorbed solvent achieved at the equilibrium absorption. Flory-Huggins equation has been used to describe the equilibrium solubility of a solvent in polymer as a function of a solvent activity. This equation is given as:

$$\ln(a) = \ln(V_1) + V_2 + X_I (V_2)^2 \quad \dots \quad (3.1)$$

where

*a* = activity of solvent, (pure solvent activity = 1.00)  $V_1$  = volume fraction solvent in the swollen polymer  $V_2$  = polymer volume fraction and  $V_2$  = 1 –  $V_1$  $X_1$  = the Flory-Huggins interaction

Percent weight gain, W% of solvent in PVC/ PE pipe was calculated by using equation 3.2

$$W\% = \frac{100V_1d_1}{(1-V_1)d_2} \dots (3.2)$$

where

W% = percent weight gain
d<sub>1</sub> = density of solvent (g/mL)
d<sub>2</sub> = density of polymer (g/mL)

Diffusion coefficient, D ( $m^2/day$ ) was determined by using equation 3.3 described by Crank (Crank, 1968)

where

 $t_{0.5}$  = time required for the sorption process to attain half of its maximum percent weight gain (day) l = thickness of polymer sample (m)

#### **3.2.2** Microscopic visualization test

The procedure for examining the extent of permeation within the polymer by microscopic visualization was followed materials analysis and research laboratory method (Town Engineering, Iowa State University);

- The pipes to be tested were cut across the longitudinal axis of the pipe, so as to form ring-like specimens that were 1 cm in length. In the case of the sorption test, the specimens were cut and then exposed to the organic solvent. In the case of the pipe-bottle experiments, the pipes were exposed to the organic solvent and then the specimens were cut.
- 2. Sections of the exposed specimens were cut parallel the longitudinal axis of the pipes, so as to form test specimens from approximately 1.5 cm of the circumference of the pipes.
- 3. The test specimens were mounted in a molds, previously coated with a thin layer of grease, so as to afford a view of both the inner and outer edges of the pipes. The mold was filled with epoxy (Leco Epoxy Resin (LC) part number 811-563) and allowed to cure for 24 hours. Mounting the test specimens in this manner made possible a microscopic view of the cross sections of the pipes, and a visualization of the effects of the organic solvents on the inner and outer surfaces of the pipes.
- 4. The cured epoxy mountings were removed from the molds for successive polishing of the viewing surfaces with 180 grit-, 400 grit-, 800 grit-, 1200 grit silicon carbide papers (Carbimet<sup>TM</sup>; Buehler, Ltd.) using an Buehler "Ecomet II" Polisher/Grinder, followed by a final polishing with a 2 μm diamond polishing compound.
- 5. The polished test specimen mountings were examined using reflected light microscopy (OLYMPUS BHM microscope). The regions of the test

specimens that had been swollen, or rubberized, by the organic solvents appeared darker than the un-swollen portions. The boundary between the swollen and un-swollen regions, the moving front, was easily visible. The thickness of the swollen region was measured by using the calibrated reticule of the eyepiece.

#### 3.2.3 Pipe-Bottle Permeation Test 3.2.3.1 Pipe-Bottle apparatus

The permeation test was conducted using a pipe-bottle permeation apparatus. The Pipe-bottle apparatus consisted of 1 L glass bottle with a Teflon line screw cap. Two holes of approximately 1.16 inches were made through the bottle such that a one-inch diameter PE pipe with a length of 3.9 inches could be mounted horizontally through the bottle as shown in Figure 3.1. The connections and gaps between the pipe and bottle were tightly wrapped with Teflon gasket tape and sealed with PolyBond 33 Adhesive (epooxy) purchased from Nbond Adhesives, Int'l (Littleton, Colorado). One end of the pipe was sealed with a 1/2-in Teflon plug and the other end of the pipe was sealed with a Stainless steel sampling port and a stainless steel screwed cap for filling and removing water from the pipe for analysis.



Figure 3.1 Schematic of Pipe-Bottle apparatus

Pipe surface area exposed to the contaminants and the volume of water inside the pipes are presented in Table 3.3.

Pipe-bottle	No.1	No.2
Surface area (cm <sup>2</sup> )	81.89	81.89
Volume of water (mL)	44.8	45.0

 Table 3.3 Exposed pipe surface areas and total volume of water inside pipe

#### **3.2.3.2 Experimental procedure**

To study permeation of solvents through pipe, aqueous solution saturated with gasoline was prepared as follow: 50 mL of gasoline was added to 1 L of distilled water in glass bottle with a Telfon cap. Solutions were mixed using a magnetic stirrer for 48 hours at  $23 \pm 1$  °C. The solutions were analyzed the concentrations of individual BTEX compounds before conducting the permeation experiment. In this experiment, the conditions to study permeation of BTEX compounds through PE pipe were maintained to follow the assumption of permeation in which the concentrations of the contaminants exposed to the outer surface of the polymer wall remain constant, the initial concentration of a diffused contaminant in the polymer is zero and the concentration of the contaminant at the other end of the polymer is kept at zero (Vonk and Veenendaal, 1984).

The pipe segment was filled with DI water pipe via the sampling port and the sampling port capped tightly with a stainless steel cap. This was followed by filling the bottle with the test solution and the pipe-bottle apparatus cap tightly with a Teflon lined screw cap. The pipe-bottle apparatus was placed in a fume hood and the temperature was maintained at  $23 \pm 1$ °C. The stainless steel sampling port was wrapped with Teflon tape to minimize potential contamination of sampling ports by volatilized gasoline. Water samples were taken at regular intervals and analyzed for the presence of BTEX. The pipe water was obtained as follows: the sampling port was cleaned with distilled water; the sampling cap was removed and a 20 mL syringe was used to remove water sample from the pipe; and placed in a 40 mL screw cap sample vial with Teflon lined septum. The pipe was rinsed and flushed with approximately 45 mL of distilled water four times; and finally the pipe was filled with

fresh distilled water. Fresh gasoline saturated solutions were replaced in the bottle every week to maintain the constant concentration of BTEX compounds in the test solution.

#### **3.2.3.3 Data analysis**

The time-lag intercept,  $T_L$  was determined by extrapolating the data line to the x-axis for plot of total quantity of individual BTEX compounds permeated through one square meter of pipe wall,  $Q_t$  (mg/m<sup>2</sup>) versus time, t (day) of exposure.

The diffusion coefficient, D ( $m^2/day$ ) and permeability coefficient, P ( $m^2/day$ ) were estimated by using equation 3.4 and 3.5, respectively.

where

 $D = diffusion coefficient (m^2/s)$ 

 $T_L$  = Breakthrough time (s)

l = the thickness of the polymer (m)

$$P = S \cdot D \dots (3.5)$$

where

S = polymer-solvent partition coefficient (dimensionless) which was determined from the slope of a plot of total quantity of individual BTEX compounds permeated through one square meter of pipe wall,  $Q_t$  (mg/m<sup>2</sup>) versus time, t (day) of exposure and was expressed in equation 3.6.

where

 $C_o$  = the outer concentration of contaminant (mg/ m<sup>3</sup>)

#### **3.3 ANALYTICAL MEASUREMENT**

The concentrations of individual BTEX compounds (benzene, toluene, ethylbenzene and o-, p-, m- xylene) and MTBE were determined using an automated purge & trap concentrator (Tekmar LSC2/ALS) and gas chromatography (GC) with a packed column (6 ft  $\times$  2 mm; 1% SP1000 on 60/80 mesh Carbopack B), and a photoionization detector. The GC conditions were: injector and detector temperatures at 175 °C and 250 °C, respectively; column temperature programming at 50 °C for 3 minutes then to 220 °C at 8 °C per minute and hold at 220 °C for 15.8 minutes. The elution times for benzene, toluene, ethylbenzene, m-xylene, o+p-xylene and MTBE peak were 16.28, 23.00, 26.06, 30.12, 30.99, and 12.00 minutes, respectively. Helium was used as the carrier gas. The sample volume used was 5 mL for the purge and trap concentrator. The concentrations of individual BTEX compounds and MTBE were calculated from the corresponding peak area.

#### **3.4 EXPERIMENTAL CHART**

The experimental activity chart is as shown below:



Figure 3.2 Experimental flow chart

#### **CHAPTER IV**

#### **RESULTS AND DISCUSSION**

#### **4.1 EQUILIBRIUM SORPTION TEST**

 $X_I$  can be served as the basis for identifying the resistance of polymers to attack by solvents and the relationship between appropriate solvent and polymer properties will be expected to bring to prediction of permeation of solvents through polymers. The result from this study was presented as follows:

# 4.1.1 Equilibrium sorption test of PVC pipe in Trichloroethylene (TCE) and Toluene

Figure 4.1 and 4.2 present the results of the equilibrium sorption test on five PVC pipes with pure trichloroethylene (TCE) and toluene. The percent weight gain with time was almost linear for the first half of the sorption process. After 4 days of exposure to TCE and after 11 days of exposure to toluene, the weight of the pipe specimens (averaged of two samples) reached a maximum weight gain.



Figure 4.1 Sorption of TCE by PVC pipes at 23°C



Figure 4.2 Sorption of Toluene by PVC pipes at 23°C

TCE and toluene were sorbed rapidly by PVC pipes and the pipes became soft and swollen after exposure to the two solvents. All five PVC pipes gave the similar sorption results with a weight gain of approximately 71.94 % for TCE and 50.45% for toluene. It can be inferred that sorption of the solvents was not impacted by the different compositions of additives in PVC pipes produced from the different sources.

The percent weight gain, W%, Flory-Huggins interaction parameter,  $X_I$ , and diffusion coefficient, D for TCE and toluene from the sorption tests are presented in Table 4.1 and 4.2 for TCE and toluene, respectively.

 Table 4.1 Summary of parameters for sorption of Trichloroethylene (TCE) by PVC

 pipe

		_	0./
Pipe ID	W% ( <u>+</u> S.D.)*	XI	$D (cm^2/hr)$
	<b>PANI</b> 9 PAN	<b>001110</b>	3 M O I 161 O
PVC B	$70.84\pm0.62$	0.86	$1.83  imes 10^{-4}$
PVC C	$71.33 \pm 0.48$	0.86	$1.68  imes 10^{-4}$
PVC D	$73.44\pm0.54$	0.86	$1.89  imes 10^{-4}$
PVC F	$72.23\pm0.63$	0.86	$2.13  imes 10^{-4}$
PVC H	$71.86\pm0.09$	0.86	$1.85  imes 10^{-4}$

\*S.D. - standard deviation

Pipe ID	W% ( <u>+</u> S.D.)*	$X_{I}$	$D (cm^2/hr)$
PVC B	$49.71\pm0.15$	0.82	$6.77  imes 10^{-5}$
PVC C	$50.80\pm0.15$	0.81	$6.23  imes 10^{-5}$
PVC D	$50.45 \pm 0.19$	0.81	$7.51 imes10^{-5}$
PVC F	$50.17 \pm 0.17$	0.81	$7.57  imes 10^{-5}$
PVC H	$51.13 \pm 0.02$	0.81	$6.87 imes10^{-5}$

Table 4.2 Summary of parameters for sorption of Toluene by PVC pipe

\* S.D. – standard deviation

The  $X_I$  values of 0.86 and 0.81 for PVC-TCE and PVC-toluene, respectively along with D values of approximately 2 x 10<sup>-4</sup> cm<sup>2</sup>/hr for TCE and 7 x 10<sup>-5</sup> cm<sup>2</sup>/hr for toluene indicate that TCE and toluene can dissolve very well in PVC pipe resulting in rapid diffusion of these solvents into the pipe. D values for TCE was larger than that for toluene indicating that TCE interact and diffused faster into PVC pipes than toluene.

# 4.1.2 Equilibrium sorption test of PVC pipe in gasoline and Methyl *tert*- butyl ether (MTBE)

In this experiment, PVC pipes were exposed to pure MTBE, regular gasoline, premium gasoline, 10% ethanol gasoline, and combined solution of 15% MTBE in premium gasoline. Figure 4.3 shows that PVC pipes sorbed very low amounts (less than 1.5% in 100 days) of the four different gasoline and MTBE. The percent weight gain, W% (average of two samples) for the five PVC pipes with different gasoline are presented in Figure 4.3.



**Figure 4.3** Sorption of four types of gasoline and MTBE at 23°C by: (a) PVC B, (b) PVC C, (c) PVC D, (d) PVC F, and (e) PVC H

Even after 90 days, sorption of gasoline or MTBE has not reached equilibrium. But since there was a limitation of time in completing this work, data for 90 days is reported here. As such, the Florry-Huggins interaction parameter,  $X_I$  and diffusion coefficient, D could not be estimated. The percent weight gain, W% for sorption of these solvent are presented in Table 4.3.

 Table 4.3 Percent weight gain, W% for sorption of gasoline and MTBE by PVC

 pipes

	W% ( <u>+</u> S.D.)* (at 90 days)					
Pipe ID	Regular gasoline	Premium gasoline	10% Ethanol gasoline	MTBE	15% MTBE in Premium Gasoline	
PVC B	$0.43 \pm 0.06$	$1.05 \pm 0.11$	$0.54 \pm 0.09$	$0.74\pm0.00$	$0.52\pm0.15$	
PVC C	$0.28 \pm 0.15$	$0.77\pm0.05$	$0.73 \pm 0.16$	$0.69\pm0.00$	$0.31\pm0.03$	
PVC D	$0.21 \pm 0.11$	$1.18\pm0.04$	$0.45\pm0.13$	$0.79\pm0.01$	$0.59\pm0.07$	
PVC F	$0.19 \pm 0.05$	$0.84\pm0.08$	$0.50\pm0.12$	$0.64\pm0.01$	$0.80\pm0.25$	
PVC H	$0.28\pm0.09$	$0.82\pm0.03$	$0.64 \pm 0.15$	$0.79\pm0.02$	$1.09\pm0.12$	

\* S.D. - standard deviation

The amount of gasoline and MTBE sorbed in comparison to TCE and toluene is very low with a small difference of W% between types of PVC pipes and gasoline. Comparison of W% between types of gasoline used indicated that PVC B, C, and D sorbed more premium gasoline than the other gasolines and MTBE. Unlike PVC B, C, and D, PVC F and H sorbed more combined solution of 15% MTBE in premium gasoline than the other gasoline and MTBE used.

In previous investigations, individual BTEX compounds were found to soften and swell PVC materials rapidly (Berens, 1985 and Vonk, 1986). Premium gasoline is composed of high concentration of BTEX compounds than regular-and 10% ethanol gasoline (see Table 3.2 for the composition of the various gasolines). Concentration of BTEX compounds in regular gasoline is almost similar to that in 10% ethanol gasoline. Based on the W% of sorption and concentration of BTEX compound in gasoline, it can be assumed that the amount of gasoline sorbed in PVC pipes is directly proportional to the concentration of BTEX compounds in gasoline.

## **4.1.3** Equilibrium sorption test of PVC pipe in combined solution of gasoline and Trichloroethylene (TCE)

In this study, the effect of TCE present in gasoline on sorption by PVC pipes was investigated. At many hazardous waste sites and industrial spill sites, multiple solvents may be present including a combination of gasoline and TCE. Five pipes were exposed to combined solutions of TCE in premium gasoline. The percentage of TCE in premium gasoline were; 1%, 5%, 10%, and 15%. PVC B was also exposed to diluted TCE solution (1%, 5%, 10% and 15%) in polyethylene glycol (PEG) as control experiments. Figure 4.4 shows the percent weight gain (average of two samples) for PVC pipes exposed to combined solutions of TCE and premium gasoline.





**Figure 4.4** Sorption of combined solution of TCE and gasoline at 23°C by (a) PVC B, (b) PVC C, (c) PVC D, (d) PVC F, and (e) PVC H.

After 90 days, the amount sorbed did not reach equilibrium. As such, the Florry-Huggins interaction parameter,  $X_I$  and diffusion coefficient, D could not be estimated. The percent weight gain, W% (average of two samples) is presented in Table 4.4.

**Table 4.4** Percent weight gain, W% for sorption of combined solution ofTrichloroethylene (TCE) and gasoline by PVC pipes

		W% (	<u>+</u> S.D.)* (at 90	days)	
Pipe ID	Premium	1% TCE in	5% TCE in	10% TCE in	15% TCE in
	gasoline	gasoline	gasoline	gasoline	gasoline
PVC B	$1.05\pm0.11$	$0.71\pm0.03$	$1.99\pm0.08$	$1.68\pm0.06$	$2.97\pm0.05$
PVC C	$0.77\pm0.05$	$0.72\pm0.06$	$1.99\pm0.05$	$1.89\pm0.09$	$3.23\pm0.02$
PVC D	$1.18\pm0.04$	$0.73\pm0.01$	$1.25\pm0.06$	$1.87\pm0.19$	$3.27\pm0.22$
PVC F	$0.84\pm0.08$	$0.87\pm0.03$	$1.42\pm0.11$	$1.36\pm0.05$	$2.51\pm0.17$
PVC H	$0.82\pm0.03$	$0.93\pm0.05$	$2.27\pm0.15$	$2.88 \pm 0.11$	$2.81\pm0.23$
		W% (	<u>+</u> S.D.)* (at eq	uilibrium absor	ption)
Pipe ID		1%TCE in	5%TCE in	10% TCE in	15% TCE in
		PEG	PEG	PEG	PEG
PVC B		$0.04 \pm 0.00$	$0.24\pm0.03$	$0.74\pm0.05$	$1.73\pm0.04$
*CD at	ndard daviatio	2			

\* S.D. – standard deviation

From Table 4.4, it can be seen that the weight gain, W% (for 90 days) was directly proportional to the percentage of TCE present in premium gasoline. For the combined solutions of 5%TCE and 10%TCE in gasoline, W% values obtained for PVC C were similar. For PVC D and H, W% seemed to increase proportionately to the percent of TCE present. The W% for PVC B and PVC F were lower in 10% TCE in gasoline than 5% TCE. This is contrary to the results found for the other PVC pipes. It is possible that the rate of uptake may be different for different percentage of TCE and it is also possible that there may be an error in the measurement of the weight of the specimen. For specimens in premium gasoline and combined solution of 1%TCE in gasoline the W% were similar (based on S.D.).

A comparison of W% between the combined solution of TCE in gasoline and the control (TCE in PEG) showed that sorption of TCE in gasoline was much larger than in the control. From these result, it can be inferred that the presence of TCE, a readily permeable organic chemical, in gasoline, a low permeable organic solvent, would result in an increase in the amount of solvents (TCE and gasoline) sorbed or permeated into PVC pipe. Using the 15% TCE in gasoline as an example, it can be seen that when the percent weights gained for premium gasoline alone were added to percent weights gained for 15% TCE in PEG, the total percent weight gained for all the pipes except for PVC F was lower than the percent weights gained for 15% TCE in gasoline may have assisted in the extra weight gained or permeation.

# 4.1.4 Equilibrium sorption test of PE pipe in gasoline and Methyl *tert*- butyl ether (MTBE)

In this experiment, PE pipes were exposed to pure MTBE, regular gasoline, premium gasoline, and 10% ethanol gasoline. Figure 4.5 presents the percent weight gain for PE pipes exposed to these organic solvents. From the figure, sorption was rapid within the first 7 days for the three gasolines and reached equilibrium mass gained by the 7<sup>th</sup> day, For MTBE, initial sorption of MTBE was non linear for the first 11 days followed by linear sorption to the 34<sup>th</sup> day where equilibrium sorption was observed.



Figure 4.5 Sorption of MTBE, and gasoline by PE at 23°C

Comparison of the percent weight gain, W% indicates that PE pipe sorbed more gasoline than MTBE. The percent weight gain (W%), Flory-Huggins interaction parameter  $(X_I)$ , and diffusion coefficient (D) for sorption of the three gasoline and MTBE were estimated and presented in Table 4.5.

Table 4.5 Summary of parameters for the sorption of gasoline and MTBE by PE pipe

Organic solvents	W% ( <u>+</u> S.D.)*	XI	D (cm <sup>2</sup> /hr)
Regular gasoline	$7.26 \pm 0.04$	1.83	$2.86  imes 10^{-4}$
Premium gasoline	$-7.32 \pm 0.01$	1.82	$2.44  imes 10^{-4}$
10% ethanol gasoline	$6.89\pm0.07$	1.87	$2.74 imes10^{-4}$
MTBE	$4.93\pm0.05$	2.24	$0.53  imes 10^{-4}$

\* S.D. – standard deviation

 $X_I$  of PE-gasoline and PE-MTBE were greater than 1 indicating that gasoline and MTBE did not dissolve well in PE pipe. However the diffusion coefficient, D was in the order of 10<sup>-4</sup> cm<sup>2</sup>/hr which was an orders of magnitude larger than the diffusion coefficient for PVC-TCE sorption experiments. These results imply that PE pipe is a rubbery polymer which is susceptible to permeation by organic solvents.

## **4.1.5** Equilibrium sorption test of PE pipe in Trichloroethylene (TCE), Benzene, Toluene, Ethylbenzene, and mixed-Xylene

In this experiment, PE pipes were exposed to pure TCE, benzene, toluene, ethylbenzene, and xylene. Figure 4.6 gives the percent weight gain for PE pipes exposed to these organic solvents. The figure shows rapid sorption of the solvents reaching equilibrium sorption after 4 days of exposure.



**Figure 4.6** Percent weight gain (W%) versus time for sorption of TCE, benzene, toluene, ethylbenzene, and mixed-xylene by PE pipes at 23°C

Comparison of the percent weight gain, W% indicates that PE pipe sorbed more TCE than the individual BTEX compounds. Percent weight gain (W%), Flory-Huggins interaction parameter ( $X_I$ ), and diffusion coefficient (D) for sorption of TCE and individual BTEX compounds were estimated and presented in Table 4.6.

Organic solvents	W% ( <u>+</u> S.D.)*	X <sub>I</sub>	D (cm <sup>2</sup> /hr)
TCF	$16.72 \pm 0.00$	1 75	$2.02 \times 10^{-4}$
TCL D	$10.72 \pm 0.00$	1.75	$2.02 \times 10^{-4}$
Benzene	$7.61 \pm 0.01$	1.94	$1.34 \times 10^{-1}$
Toluene	$8.00 \pm 0.04$	1.89	$1.24  imes 10^{-4}$
Ethylbenzene	$7.42\pm0.00$	1.95	$1.09  imes 10^{-4}$
mixed-Xylene	$8.28 \pm 0.02$	1.87	$1.17  imes 10^{-4}$

**Table 4.6** Summary of parameters for sorption of Trichloroethylene (TCE) and

 individual BTEX compound by PE pipes

\* S.D. - standard deviation

 $X_I$  of PE-individual BTEX compounds and PE-TCE were greater than 1 indicating that individual BTEX compounds and TCE did not dissolve well in PE pipe. The diffusion coefficients, D of these organic solvents in PE pipe were in the order of  $10^{-4}$  cm<sup>2</sup>/hr. These results imply that PE pipe is a rubbery polymer which is susceptible to permeation by organic solvents.

#### **4.2 MICROSCOPIC VISUALIZATION TEST**

The extent of permeation within the polymer can be determined by conducting microscopic visualization test. In this test, the pipe specimens exposed to organic solvents were cut radially and examined by microscope to determine the moving front of the permeation of the solvent. This is indicated by a boundary or interface between the rubberized zone where the polymer structure was relaxed by solvent and the unrubberized zone where the polymer structure was rigid and of low mobility. The moving front of the solvent within polymer can be served as a basis to predict the required time for solvents to permeate through the pipe. In this test, PVC B and PVC D were selected as representative PVC materials to examine the permeation of solvents using the microscopic visualization test since five different manufacturers of PVC pipe tested gave almost similar results from equilibrium sorption tests. The results from this work could be correlated with the results from equilibrium sorption test to evaluate the resistance of PVC pipes to permeation of solvents.

# **4.2.1** Microscopic visualization of permeation of Trichloroethylene (TCE) and Toluene within PVC pipe

Figure 4.7 and Figure 4.8 show the moving fronts formed by the penetration of trichloroethylene (TCE) and toluene when the specimens were exposed for 2 days and 3 days, respectively. Since the inside wall and outside surface of the pipe were exposed to the solvents, two fronts can be seen in the Figures. Movement of both fronts over time was measured accordingly. Figures 4.9 (a) and (b) present the results of the inside and outside moving fronts in PVC D exposed to pure TCE and toluene.



**Figure 4.7** Picture of moving-front of TCE penetrating within PVC D after 2 days.



**Figure 4.8** Picture of moving-front of toluene penetrating within PVC D after 3 days.



**Figure 4.9** Penetration of TCE (a) and toluene (b) into PVC D based on microscopic visualization tests (outside – penetration from outside surface to inside wall; inside – penetration from inside wall to outside surface)

From Figure 4.9, penetration of TCE and toluene from outside wall into the pipe appeared to be faster than penetration from inside wall into pipe. PVC pipe was swollen rapidly and was completely swollen after 2 day of exposure to TCE and after 5 days of exposure to toluene. It could be seen that TCE and toluene penetrated into PVC pipe rapidly and penetration of TCE did so at a faster rate than toluene.

Diffusion coefficient, D values of TCE and toluene were estimate from the results of the moving front. It indicated that D value of TCE and toluene penetration from outside to inside wall were approximately  $1.50 \times 10^{-3}$  and  $4.59 \times 10^{-4}$  cm<sup>2</sup>/hr and D value of TCE and toluene penetration from inside to outside wall were approximately  $1.01 \times 10^{-3}$  and  $2.04 \times 10^{-4}$  cm<sup>2</sup>/hr. Comparison D values of TCE and toluene obtained from equilibrium sorption (D<sub>TCE</sub> =  $1.89 \times 10^{-4}$ , D<sub>Toluene</sub> =  $7.51 \times 10^{-5}$  cm<sup>2</sup>/hr) with D values obtained from microscopic visualization test found that microscopic visualization test provider D values of TCE and toluene larger than equilibrium sorption test. However D values obtained from these two methods were similar in which diffusion of TCE did at faster rate than for toluene. So, both of equilibrium sorption test and microscopic visualization test could be used to determine the resistance of PVC pipe to attack by solvent and to investigate permeation of solvent through PVC pipe.

Based on the results from equilibrium sorption test and microscopic visualization test, TCE and toluene could dissolve very well in PVC pipe and they were rapidly permeable organic solvents. This information suggests that PVC might not be an effective barrier to against permeation of pure solvents such as TCE and toluene.

# 4.2.2 Microscopic visualization of permeation of gasoline and Methyl *tert*- butyl ether (MTBE) within PVC pipe

Figures 4.10 shows the moving front for PVC pipe exposed to premium gasoline. Similarly for the other solvents, the presence of the moving fronts were also examined. Table 4.7 summarizes the results of the examination of the moving fronts in PVC B exposed to premium gasoline and MTBE for 120 days and to regular gasoline, 10% ethanol, and combined solution of 15% MTBE in premium gasoline for 90 days.



**Figure 4.10** Picture of moving front of premium gasoline penetrating within PVC D after 120 days (a)  $\times$  25 magnification, (b)  $\times$  160 magnification

Solvent	Time of	Distance (mm)		
Solvent	exposure (Days)	Outside	Inside	
Regular gasoline	90	Non detected	Non detected	
Premium gasoline	120	0.3	Non detected	
10% Ethanol gasoline	90	Non detected	Non detected	
MTBE	120	0.2	Non detected	
Combined solution of 15%	90	Non detected	Non detected	
MTBE in premium gasoline				

Table 4.7 Distance of moving front for gasoline and MTBE in PVC pipe

The different times of exposure of the pipe to solvents resulted in observation of moving-front at different distances. From Table 4.7, the results show that there was no penetration of regular gasoline, 10% ethanol and combined solution of 15% MTBE in premium gasoline at 90 days into PVC pipe as indicated by the lack of appearance of the moving front in the pipe. There was an appearance of the moving front in the pipe. There was an appearance of the moving front in the pipe exposed to premium gasoline and MTBE for 120 days. The premium gasoline and MTBE had penetrated from the outside wall into pipe to a distance of about 0.3 and 0.2 mm, respectively. However, moving front from the penetration from inside wall into pipe was not observed.

By estimation, premium gasoline and MTBE would take 4.1 years and 6.1 years, respectively to permeate through PVC pipe.

Based on the results from equilibrium sorption test and microscopic visualization test, PVC pipe did not sorb gasoline and MTBE well and that gasoline and MTBE were low permeable organic solvent for PVC pipe. This suggest that exposure of PVC pipe to gasoline and MTBE might not result in permeation of gasoline through the pipe or if permeation does occur it would take a long time to permeate through the pipe.

## **4.2.3** Microscopic visualization of permeation of combined solution of Trichloroethylene (TCE) and gasoline within PVC pipe

Figure 4.11 shows the moving front of different TCE concentrations in gasoline in PVC after 120 days of exposure while Table 4.8 summarizes the results of examination of moving front in PVC B which has been exposed to combined solution of TCE in premium gasoline for 120 days.

**Table 4.8** Distance the moving front of combined solution of Trichloroethylene (TCE)

 and gasoline in PVC B

Solvent	Time of	Distan	ice (mm)
Solvent	exposure (Days)	Out side	Inside
1% TCE in premium gasoline	120	0.15	Non detected
5% TCE in premium gasoline	120	0.40	0.04
10% TCE in premium gasoline	120	0.37	0.04
15% TCE in premium gasoline	120	0.53	0.10

The pipes which have been exposed to combined solution of 15% TCE in premium gasoline provided a large the distance the moving front than pipes which have been exposed to combined solution of 10%, 5%, 1% TCE in premium gasoline. Moreover, the distance the moving front in the pipes which have been exposed to combined solution of 10% and 5% TCE in premium gasoline were almost similar.

Comparison the distance of moving fronts in the pipes which have been exposed to pure premium gasoline and the distance of moving fronts in the pipes which have been exposed to combined solution of 1% TCE in premium gasoline found that there was no difference of the distance of the moving front. The picture of moving front which has been formed by penetration of combined solution of TCE in premium gasoline was illustrated in figure 4.10.



**Figure 4.11** Picture of moving front of combined solution of 1%TCE (a)-, 5%TCE (b)-, 10%TCE (c)-, and 15%TCE (d) in gasoline penetrating from outside wall into pipe

These results agreed with the results from equilibrium sorption test of combined solution of TCE in premium gasoline by PVC pipe in which sorption of combined solution of 15% TCE in premium gasoline provided he highest percent weight gain, W% and sorption of combined solution of 15% TCE and 10% TCE in

premium gasoline had a similar W%. Furthermore, sorption of pure premium gasoline and combined solution of 1% TCE in premium gasoline provided the similar W%.

By estimation, combined solution of 1%, 5%, 10%, and 15% TCE in premium gasoline would take 8.1, 3.0, 3.3 and 2.3 years, respectively to permeate through PVC pipe. Almost combined solution excepted for combined solution of 1% TCE in gasoline took less time to permeate though the pipe than premium gasoline which would take for 4.1 years.

It could be indicated from this study that addition of TCE in premium gasoline would promote the permeation rate of gasoline through the pipe. TCE having high solvent power could change the polymer structure of PVC from glassy polymer to rubbery polymer which was flexible to permeation. Consequently, the numbers of gasoline entering into PVC pipe was increasing. However, low concentration of TCE such a combined solution of 1% TCE in premium gasoline could not promote for changing the polymer structure of PVC pipe.

Based on the results from equilibrium sorption test and microscopic visualization test, exposing of PVC pipe to gasoline contained high concentration of TCE might then result in permeation through the pipe.

#### **4.3 PERMEATION TEST**

In this experiment, permeation of BTEX compounds through PE pipe was investigated. The PE pipe was exposed to gasoline and to an aqueous solution saturated with gasoline. The result of this experiment was presented as follows:

#### 4.3.1 Permeation of BTEX compounds in gasoline through PE pipe

Results of permeation of BTEX through PE pipe are presented in Figure 4.12. The results showed a sharp increase in the BTEX compounds in the pipe water after 7 days and the water in the pipe had a strong petroleum hydrocarbon odor. Figure 4.19 presents permeation of individual BTEX compounds.



**Figure 4.12** BTEX compounds permeation through PE pipe wall, Q ( $\mu$ g/cm<sup>2</sup>) versus time (day) of exposure at 23°C



**Figure 4.13** Individual BTEX compound permeation through PE pipe wall, Q  $(\mu g/cm^2)$  versus the time (day) of exposure at 23 °C

From Figure 4.13, the breakthrough of benzene, toluene, ethylbenzene, and xylene occurred after approximately 7 days but concentration of benzene and toluene in pipe water were higher than concentration of ethylbenzene, m-xylene, and o+p-xylene in pipe water pretty much. It might be assumed that the amounts of individual BTEX compounds entering to PE pipe were affected on its size of molecule. Figure 4.14 presents the estimated time-lag,  $T_L$  of individual BTEX compounds.



(b)







**Figure 4.14** Time lag,  $T_L$  (hour) for permeation of (a) Benzene, (b) Toluene, (c) Ethylbenzene, (d) m-Xylene, and (e) o+p-Xylene through PE pipe wall.

Time lag,  $T_L$  (hour), polymer-solvent partition coefficient, S, diffusion coefficient, D (m<sup>2</sup>/hr), and permeability coefficient, P (m<sup>2</sup>/hr) of individual BTEX compound were determined and presented in Table 4.9.

Compounds	T <sub>L</sub> (hr)	$D (cm^2/hr)$	S	$P(cm^2/hr)$
		-	250	0
Benzene	366.50	$4.95 \times 10^{-5}$	$2.07 \times 10^{-4}$	$1.03 \times 10^{-8}$
Toluene	363.14	$5.00\times10^{\text{-5}}$	$5.15 \times 10^{-5}$	$2.58\times10^{-9}$
Ethylbenzene	359.83	$5.04 \times 10^{-5}$	$1.31 \times 10^{-5}$	$6.62\times 10^{10}$
m-Xylene	361.06	$5.03  imes 10^{-5}$	$1.34 \times 10^{-5}$	$6.75\times10^{10}$
o+p-Xylene	362.85	$5.00 \times 10^{-5}$	$1.52 \times 10^{-5}$	$7.61 \times 10^{-10}$

Table 4.9 Summary parameters of permeation of BTEX compounds

Comparison of D values from Table 4.9 with the D values obtained from equilibrium sorption of individual BTEX compounds by PE pipe in which D of benzene, toluene, ethylbenzene, and mixed-xylene were  $1.34 \times 10^{-4}$ ,  $1.24 \times 10^{-4}$ ,  $1.09 \times 10^{-4}$ , and  $1.17 \times 10^{-4}$  (cm<sup>2</sup>/hr), respectively. The results showed that D values obtained from equilibrium sorption and permeation test were different magnitudes in which D values obtained from equilibrium sorption were larger than D values

obtained from permeation test. Since concentrations of individual BTEX compounds in pure solvent used in equilibrium sorption test were higher than concentration of individual BTEX compounds consisted in gasoline used in permeation test, it might be assumed that D value of individual BTEX compounds would depend on their concentration at the outer surface of the pipe wall.

Comparison of polymer-solvent partition coefficient, S values of individual BTEX compounds from Table 4.9, results showed that there was more partition of benzene between gasoline and outer surface of PE wall than partition of toluene, ethylbenzene, and xylene between gasoline and outer surface of PE wall. Moreover, permeation of benzene provided the highest rate of permeation at stationary state.

The maximum contaminant levels (MCsL) for benzene, toluene, ethylbenzene, xylene are 0.005, 1.00, 0.70, and 10 mg/L respectively. From the results, the concentration of benzene, toluene, ethylbenzene, and xylene at stationary state was 82.39, 73.03, and 10.91 mg/L, which were over their MCL. It imply that people may have high risk will be expose to high concentration level of BTEX compounds via drinking water and consuming water and resulting in health effects for acute effect and chronic effect.

Based on these results, it indicates that PE pipe can not be buried in areas that it may directly contact with gasoline such as laying under the gasoline underground storage tank since it can not definitely resist to permeation of BTEX consisted in gasoline.

# 4.2.2 Permeation of BTEX compounds in aqueous solution saturated with gasoline through PE pipe

Results of permeation of BTEX through PE pipe are presented in Figure 4.15. The results showed a sharp increase in the BTEX compounds in the pipe water after 10 days and the water in the pipe had a strong petroleum hydrocarbon odor. Figure 4.16 presents permeation of individual BTEX compounds.


Figure 4.15 BTEX compounds permeation through PE pipe wall,  $Q_t (\mu g/cm^2)$  versus time (day) of exposure at 23°C



Figure 4.16 Individual BTEX compound permeation through PE pipe wall,  $Q_t$  ( $\mu$ g/cm<sup>2</sup>) versus the time (day) of exposure at 23 °C

From Figure 4.16, the breakthrough of benzene, toluene, ethybenzene, mxylene, o+p-xylene occurred after approximately 10 days but concentration of benzene and toluene in pipe water were higher than concentration of ethylbenzene, mxylene, and o+p-xylene in pipe water pretty much. It might be assumed that the amounts of individual BTEX compounds entering to PE pipe were affected on its size of molecule. Figure 4.17 presents the estimated time-lag,  $T_L$  of individual BTEX compounds.





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**Figure 4.17** Time lag,  $T_L$  (hour) for permeation of (a) Benzene, (b) Toluene, (c) Ethylbenzene, (d) m-Xylene, and (e) o+p-Xylene through PE pipe wall.

Time lag,  $T_L$  (hour), polymer-solvent partition coefficient, S, diffusion coefficient, D (m<sup>2</sup>/hr), and permeability coefficient, P (m<sup>2</sup>/hr) of individual BTEX compound were determined and presented in Table 4.10.

Table 4.10 Summary parameters of permeation of BTEX compounds

Compounds	T <sub>L</sub> (hr)	$D (cm^2/hr)$	S	$P(cm^2/hr)$
Benzene	427.05	$4.25 \times 10^{-5}$	$7.03 \times 10^{-2}$	$2.99 \times 10^{-6}$
Toluene	423.17	$4.29  imes 10^{-5}$	$7.93  imes 10^{-2}$	$3.40\times10^{\text{-}6}$
Ethylbenzene	399.98	$4.54  imes 10^{-5}$	$6.09 \times 10^{-2}$	$2.76\times 10^{\text{-}6}$
m-Xylene	399.49	$4.54 \times 10^{-5}$	$6.24 \times 10^{-2}$	$2.83  imes 10^{-6}$
o+p-Xylene	402.55	$4.51 \times 10^{-5}$	$6.11 \times 10^{-2}$	$2.76  imes 10^{-6}$
Ethylbenzene m-Xylene o+p-Xylene	423.17 399.98 399.49 402.55	$4.29 \times 10^{-5}$ $4.54 \times 10^{-5}$ $4.54 \times 10^{-5}$ $4.51 \times 10^{-5}$	$7.93 \times 10^{-2}$ $6.09 \times 10^{-2}$ $6.24 \times 10^{-2}$ $6.11 \times 10^{-2}$	$3.40 \times 10^{-6}$ $2.76 \times 10^{-6}$ $2.83 \times 10^{-6}$ $2.76 \times 10^{-6}$

Comparison of D values obtained from permeation of BTEX compounds in gasoline experiment with the D values from permeation of BTEX compounds in aqueous solution saturated with gasoline experiment, the results showed that D values of BTEX compounds in gasoline were larger than D values of BTEX in aqueous solution saturated with gasoline. Concentrations of BTEX compounds in gasoline were higher than that in aqueous solution saturated with gasoline, these results might support the assumption that diffusion coefficient, D of individual BTEX compounds is dependent on their concentration at outer surface of pipe wall.

S values obtained from permeation of BTEX compounds in gasoline experiment were smaller than S values obtain from permeation of BTEX compounds in aqueous solution saturated with gasoline. These results indicated that there was more partitioning of BTEX compound between water and outer surface of PE pipe than partitioning of BTEX in gasoline and outer surface of PE pipe.

The maximum contaminant levels (MCsL) for benzene, toluene, ethylbenzene, m-xylene, and o+p-xylene are 0.005, 1.00, 0.70, and 10 mg/L respectively. From the results, the concentration of benzene, toluene, ethylbenzene, and xylene, at stationary state of permeation was 68.19, 72.82, 10.4 mg/L, which were over their MCL excepted. It imply that people may have high risk to be exposed to high concentration level of BTEX compounds via drinking water and consuming water and resulting in health effects for acute effect and chronic effect.

Based on these results, it indicates that PE pipe can not be buried in areas that it may be directly contact with high concentration of BTEX compounds in gasoline such as laying in the zone where groundwater saturated with gasoline since it can not definitely resist to permeation of BTEX compounds.

## **CHAPTER V**

### CONCLUSIONS AND RECOMMENDATIONS

### **5.1 CONCLUSIONS**

Contamination of drinking water and consuming water due to permeation of organic chemicals through plastic pipes is a serious problem since contaminated drinking water has a potential to cause adverse health effects in human. This research was conducted to further investigate the permeation of more commonly found compounds in the environment such as gasoline and to address some of the data gaps on permeation of organic solvents using commonly used and different techniques. Results showed that some organic solvents with an ability to soften power can permeate through PVC pipe rapidly. However, PVC is an effective material against permeation of organic compounds such as toluene and benzene of high softening ability, however, have low impact on permeation. A possible reason is that the low molar fraction of these compounds in gasoline (in the range of less than 5%) reduced the softening ability of these compounds in gasoline. For PE pipe, the experimental results showed that polyethylene polymer is highly permeable to gasoline and chlorinated solvents. Major conclusions of this research are as follow:

- Five PVC pipes from different manufacturers provided similar results for both equilibrium sorption test and microscopic visualization test. This implies that either the additives used by the manufacturers were similar or different compositions of additives used by different manufacturers of PVC pipes might not impact the permeation behavior. The additives used are proprietary information and are not available.
- PVC pipes were rapidly permeated by the strong swelling agents such as trichloroethylene (TCE) and toluene. The Florry-Huggins index of less than 1 (TCE = 0.86 and toluene = 0.81) generally indicate that the solvent is a strong swelling agent.

- 3. The experimental results showed that PVC pipes are resistant to permeation of regular gasoline and methyl *tert*-butyl ether (MTBE). However, exposure of PVC pipe to gasoline containing higher concentrations of BTEX compounds such as premium gasoline may result in the possibility of permeation. Moreover, addition of a strong swelling chemical such as TCE will promote permeation.
- 4. Permeation of BTEX compounds in gasoline and aqueous solution saturated with gasoline through PE pipe generally occurred after 7 days of exposure. The concentration of individual BTEX compounds in pipe water at stationary state of permeation was beyond the maximum contaminant level (MCL).
- 5. The Diffusion coefficients, D (cm<sup>2</sup>/hr) for individual BTEX compounds through the PE pipe were strongly dependent on their concentration at outside surface of pipe wall. Additionally, the amount of BTEX compounds permeate through PE pipe was affected by their molecule size and their polymer-solvent partition coefficient.

# **5.2 RECOMMENDATIONS**

There is a need to improve the industry's understanding in the selection of the appropriate pipe materials for conveying potable in the places where there is a history of petroleum based hydrocarbon contamination especially with gasoline contamination. Additionally, proper methodologies to investigate the permeation of organic chemicals must be established. The following are suggested based on this study:

1. PVC pipe is not a suitable material to convey potable water in the contaminated areas where there are high concentrations of organic chemicals having high solvent power ( $X_I < 1.0$ ). However, PVC pipes may be buried in areas contaminated with low permeable organic chemicals, for example, gasoline and MTBE since it offers a good resistance against permeation.

- 2. PE pipe should not be buried in areas contaminated with organic chemicals such as TCE, MTBE and gasoline even at low concentrations of these compounds since the polymer structure is subjected to permeation by these chemicals.
- 3. To study the permeation of organic chemicals through PE pipe, equilibrium sorption test is an effective way to investigate since the results of the equilibrium sorption test is similar to the permeation test. Furthermore, this method offers advantages such as shorter experimental time and lower cost.
- 4. For the further study, permeation test should be conducted to investigate permeation of BTEX in gasoline through PVC pipe. The results from this experiment can be combined with results from equilibrium sorption test and microscopic visualization test to ensure that PVC can resist permeation of BTEX in gasoline.

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APPENDICE

# **APPENDIX** A

Day		Percent weight gain (%)				
2	PVC B	PVC C	PVC D	PVC F	PVC H	
0	0.00	0.00	0.00	0.00	0.00	
2	$47.62\pm0.54$	$46.53 \pm 0.12$	$52.40 \pm 1.40$	$47.09\pm0.17$	$47.14\pm0.02$	
4	$70.14\pm0.28$	$69.18\pm0.19$	$73.48 \pm 0.10$	$70.75\pm0.20$	$71.18\pm0.71$	
7	$71.52\pm0.53$	$71.68\pm0.06$	$72.88 \pm 0.10$	$72.88 \pm 0.05$	$73.23\pm0.13$	
9	$70.67 \pm 0.13$	$71.53\pm0.00$	$73.95\pm0.29$	$72.19\pm0.34$	$71.96\pm0.07$	
11	$70.32\pm0.17$	$70.79\pm0.42$	$71.61 \pm 1.93$	$71.61\pm0.48$	$71.84 \pm 0.12$	
14	$69.31 \pm 0.23$	$70.09\pm0.01$	$70.65 \pm 2.01$	$71.01 \pm 0.43$	$71.79\pm0.02$	
16	$68.93 \pm 0.21$	$69.75 \pm 0.01$	$70.80 \pm 2.14$	$70.80\pm0.63$	$71.65\pm0.01$	

Table A-1 Percent weight gain of five PVC pipes in Trichloroethylene at 23°C

Table A-2 Percent weight gain of five PVC pipes in Toluene at 23°C

Day	Percent weight gain (%)				
,	PVC B	PVC C	PVC D	PVC F	PVC H
0	0.00	0.00	0.00	0.00	0.00
2	$16.13 \pm 0.95$	$15.78\pm0.01$	$15.13\pm2.25$	$15.17\pm0.15$	$16.14\pm0.00$
4	$24.78 \pm 1.64$	$24.86\pm0.10$	$27.00\pm0.13$	$23.87\pm0.26$	$25.12\pm0.02$
7	$37.91 \pm 0.10$	$37.28 \pm 0.03$	$42.30\pm0.08$	$35.44 \pm 0.17$	$37.14\pm0.02$
9	$47.54\pm0.01$	$48.71 \pm 1.83$	$49.85\pm0.10$	$45.74\pm0.52$	$49.49 \pm 0.04$
11	$49.88\pm0.16$	$50.64\pm0.00$	$50.65\pm0.02$	$50.06\pm0.15$	$51.12\pm0.04$
14	$49.66\pm0.00$	$50.93 \pm 0.13$	$50.43 \pm 0.03$	$50.37\pm0.04$	$51.15\pm0.00$
16	$49.59\pm0.03$	$50.82 \pm 0.03$	$50.27\pm0.09$	$50.09\pm0.12$	$51.11\pm0.13$

Day		Percent weight gain (%)					
2	PVC B	PVC C	PVC D	PVC F	PVC H		
0	0.00	0.00	0.00	0.00	0.00		
3	$0.00\pm0.00$	$0.01\pm0.00$	$0.00\pm0.00$	$0.00\pm0.00$	$0.00\pm0.01$		
9	$0.00\pm0.03$	$0.03\pm0.01$	$0.02\pm0.00$	$0.00\pm0.00$	$0.00\pm0.00$		
15	$0.02\pm0.00$	$0.04 \pm 0.00$	$0.03\pm0.00$	$0.01\pm0.00$	$0.01\pm0.00$		
34	$0.04\pm0.00$	$0.04 \pm 0.01$	$0.05 \pm 0.00$	$0.04\pm0.02$	$0.02\pm0.00$		
45	$0.10\pm0.02$	$0.07\pm0.09$	$0.06 \pm 0.01$	$0.06\pm0.01$	$0.07\pm0.02$		
66	$0.16\pm0.03$	$0.14 \pm 0.07$	$0.10 \pm 0.01$	$0.09\pm0.00$	$0.10\pm0.01$		
82	$0.34\pm0.04$	$0.25 \pm 0.09$	$0.16\pm0.08$	$0.14\pm0.04$	$0.24\pm0.08$		
90	$0.43 \pm 0.06$	$0.28 \pm 0.15$	$0.21\pm0.10$	$0.19\pm0.05$	$0.28\pm0.12$		

Table A-3 Percent weight gain of five PVC pipes in Regular gasoline at 23°C

Table A-4 Percent weight gain of five PVC pipes in Premium gasoline at 23°C

Day	Percent weight gain (%)						
5	PVC B	PVC C	PVC D	PVC F	PVC H		
0	0.00	0.00	0.00	0.00	0.00		
7	$0.13 \pm 0.00$	$0.10\pm0.00$	$0.12\pm0.01$	$0.10\pm0.01$	$0.10\pm\ 0.01$		
18	$0.21\pm0.02$	$0.13\pm0.02$	$0.21\pm0.02$	$0.14\pm0.00$	$0.18\pm0.03$		
23	$0.25 \pm 0.01$	$0.14\pm0.01$	$0.24\pm0.01$	$0.16\pm0.01$	$0.21\pm0.01$		
34	$0.30\pm0.03$	$0.16\pm0.00$	$0.33\pm0.02$	$0.19\pm0.01$	$0.27\pm0.04$		
55	$0.51 \pm 0.02$	$0.36\pm0.02$	$0.61\pm0.02$	$0.37\pm0.03$	$0.48\pm0.03$		
70	$0.68 \pm 0.04$	$0.48\pm0.02$	$0.77 \pm 0.02$	$0.64 \pm 0.01$	$0.65\pm0.04$		
75	$0.77\pm0.10$	$0.55 \pm 0.02$	$0.87\pm0.04$	$0.74\pm0.00$	$0.73\pm0.04$		
82	$0.80\pm0.12$	$0.59\pm0.07$	$0.92\pm0.03$	$0.78\pm0.03$	$0.76\pm0.03$		
90	$1.05\pm0.11$	$0.77\pm0.05$	$1.18\pm0.04$	$0.84\pm0.08$	$0.82\pm0.03$		

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Day	Percent weight gain (%)					
2	PVC B	PVC C	PVC D	PVC F	PVC H	
0	0.00	0.00	0.00	0.00	0.00	
3	$0.00\pm0.00$	$0.00\pm0.00$	$0.00\pm0.00$	$0.01 \pm 0.00$	$0.00\pm0.00$	
9	$0.04{\pm}0.02$	$0.05\pm0.01$	$0.03\pm0.02$	$0.08\pm0.00$	$0.08\pm0.00$	
15	$0.09\pm0.02$	$0.06 \pm 0.00$	$0.08\pm0.03$	$0.14\pm0.02$	$0.16\pm0.03$	
34	$0.17\pm0.00$	$0.22 \pm 0.01$	$0.15 \pm 0.04$	$0.19\pm0.02$	$0.23\pm0.06$	
45	$0.28\pm0.01$	$0.32\pm0.03$	$0.20 \pm 0.04$	$0.30\pm0.03$	$0.32\pm0.05$	
66	$0.33\pm0.06$	$0.42 \pm 0.08$	$0.27\pm0.07$	$0.44\pm0.03$	$0.44\pm0.08$	
82	$0.47\pm0.10$	$0.65 \pm 0.11$	$0.37 \pm 0.10$	$0.49\pm0.08$	$0.49\pm0.09$	
90	$0.54 \pm 0.09$	$0.73 \pm 0.16$	$0.45 \pm 0.13$	$0.59\pm0.12$	$0.64\pm0.15$	

Table A-5 Percent weight gain of five PVC pipes in 10% ETOH gasoline at 23°C

Table A-6 Percent weight gain of five PVC pipes in MTBE at 23°C

Day		Percent weight gain (%)						
5	PVC B	PVC C	PVC D	PVC F	PVC H			
0	0.00	0.00	0.00	0.00	0.00			
7	$0.24 \pm 0.00$	$0.10\pm0.00$	$0.21\pm0.00$	$0.14\pm0.00$	$0.15\pm0.01$			
18	$0.30\pm0.03$	$0.17\pm0.02$	$0.29 \pm 0.01$	$0.22\pm0.01$	$0.25\pm0.01$			
24	$0.39 \pm 0.04$	$0.29\pm0.01$	$0.36\pm0.01$	$0.32\pm0.01$	$0.35\pm0.00$			
39	$0.51 \pm 0.01$	$0.33\pm0.00$	$0.43\pm0.02$	$0.39 \pm 0.01$	$0.42\pm0.04$			
59	$0.57\pm0.01$	$0.52\pm0.00$	$0.64\pm0.00$	$0.56\pm0.02$	$0.61\pm0.02$			
70	$0.60 \pm 0.01$	$0.59\pm0.01$	$0.70\pm0.00$	$0.60\pm0.00$	$0.69\pm0.02$			
75	$0.65\pm0.00$	$0.61\pm0.01$	$0.73\pm0.02$	$0.61 \pm 0.01$	$0.72\pm0.02$			
84	$0.67\pm0.00$	$0.64\pm0.01$	$0.76\pm0.02$	$0.63\pm0.01$	$0.76\pm0.02$			
90	$0.74\pm0.00$	$0.69\pm0.00$	$0.79\pm0.01$	$0.64\pm0.01$	$0.79\pm0.02$			



Day	Percent weight gain (%)					
2	PVC B	PVC C	PVC D	PVC F	PVC H	
0	0.00	0.00	0.00	0.00	0.00	
3	$0.02\pm0.00$	$0.00\pm0.00$	$0.00\pm0.00$	$0.01\pm0.00$	$0.01\pm0.00$	
9	$0.09 \pm 0.02$	$0.02\pm0.01$	$0.05\pm0.02$	$0.04\pm0.00$	$0.06\pm0.00$	
15	$0.15\pm0.02$	$0.06 \pm 0.00$	$0.10 \pm 0.03$	$0.11\pm0.02$	$0.12\pm0.03$	
34	$0.24\pm0.00$	$0.07\pm0.01$	$0.17 \pm 0.04$	$0.24\pm0.02$	$0.29\pm0.06$	
45	$0.29\pm0.01$	$0.15\pm0.03$	$0.27\pm0.04$	$0.35\pm0.08$	$0.42\pm0.05$	
66	$0.37 \pm 0.06$	$0.19\pm0.08$	$0.42\pm0.07$	$0.47\pm0.13$	$0.54\pm0.08$	
82	$0.45\pm0.10$	$0.26 \pm 0.05$	$0.51 \pm 0.05$	$0.68\pm0.18$	$0.92\pm0.09$	
90	$0.52 \pm 0.15$	$0.31 \pm 0.03$	$0.59 \pm 0.07$	$0.80 \pm 0.25$	$1.09\pm0.12$	

**Table A-7** Percent weight gain of five PVC pipes in 15% MTBE + 85% Premiumgasoline at 23°C

**Table A-8** Percent weight gain of five PVC pipes in 1%TCE + 99%Premium gasoline at 23°C

Day		Chemical studies					
5	PVC B	PVC C	PVC D	PVC F	PVC H		
0	0.00	0.00	0.00	0.00	0.00		
3	$0.06 \pm 0.00$	$0.04\pm0.00$	$0.02\pm0.00$	$0.01 \pm 0.00$	$0.01\pm0.00$		
7	$0.17\pm0.02$	$0.14\pm0.01$	$0.14\pm0.02$	$0.08\pm0.00$	$0.08\pm0.00$		
11	$0.22 \pm 0.02$	$0.16\pm0.00$	$0.18\pm0.03$	$0.14 \pm 0.02$	$0.16\pm0.03$		
18	$0.27\pm0.00$	$0.18\pm0.01$	$0.26\pm0.04$	$0.19 \pm 0.02$	$0.23\pm0.06$		
23	$0.31\pm0.01$	$0.22\pm0.03$	$0.32\pm0.04$	$0.30\pm0.03$	$0.32\pm0.05$		
34	$0.39\pm0.00$	$0.32\pm0.03$	$0.40\pm0.04$	$0.44 \pm 0.03$	$0.44\pm0.08$		
40	$0.44 \pm 0.01$	$0.37\pm0.01$	$0.49\pm0.04$	$0.49\pm0.02$	$0.49\pm0.05$		
53	$0.48\pm0.00$	$0.49\pm0.04$	$0.54\pm0.00$	$0.59\pm0.00$	$0.64 \pm 0.03$		
82	$0.66\pm0.02$	$0.65\pm0.04$	$0.68\pm0.02$	$0.80\pm0.05$	$0.83\pm0.02$		
90 <sup>9</sup>	$0.71\pm0.03$	$0.72\pm0.06$	$0.73\pm0.01$	$0.87\pm0.03$	$0.93\pm0.05$		

Day		С	hemical studies	8	
	PVC B	PVC C	PVC D	PVC F	PVC H
0	0.00	0.00	0.00	0.00	0.00
3	$0.10 \pm 0.00$	$0.04\pm0.01$	$0.06\pm0.01$	$0.08\pm0.02$	$0.06\pm0.01$
7	$0.23\pm0.05$	$0.20\pm0.00$	$0.16\pm0.01$	$0.16\pm0.02$	$0.21\pm0.02$
11	$0.34\pm0.02$	$0.33\pm0.00$	$0.25 \pm 0.01$	$0.28\pm0.02$	$0.30\pm0.01$
18	$0.47\pm0.02$	$0.50\pm0.05$	$0.32 \pm 0.00$	$0.37\pm0.03$	$0.40\pm0.04$
23	$0.62\pm0.05$	$0.66\pm0.02$	$0.42\pm0.00$	$0.48\pm0.02$	$0.52\pm0.03$
34	$0.85 \pm 0.04$	$0.92\pm0.03$	$0.57 \pm 0.03$	$0.65\pm0.01$	$0.71\pm0.03$
40	$0.95\pm0.01$	$1.03\pm0.04$	$0.62\pm0.05$	$0.71\pm0.05$	$0.78\pm0.03$
53	$1.25 \pm 0.04$	$1.34\pm0.06$	$0.83\pm0.04$	$0.90\pm0.04$	$1.04\pm0.03$
82	$1.79 \pm 0.04$	$1.81\pm0.05$	$1.12 \pm 0.04$	$1.25 \pm 0.12$	$1.99\pm0.06$
90	$1.99 \pm 0.08$	$1.99 \pm 0.05$	$1.25 \pm 0.06$	$1.42 \pm 0.11$	$2.27\pm0.15$

**Table A-9** Percent weight gain of five PVC pipes in 5%TCE + 95%Premium gasoline at 23°C

**Table A-10** Percent weight gain of five PVC pipes in 10%TCE + 90%Premiumgasoline at 23°C

Day		C	hemical studies	S	
5	PVC B	PVC C	PVC D	PVC F	PVC H
0	0.00	0.00	0.00	0.00	0.00
3	$0.21\pm0.02$	$0.12\pm0.02$	$0.10\pm0.00$	$0.11 \pm 0.02$	$0.14\pm0.01$
7	$0.30\pm0.01$	$0.27\pm0.04$	$0.29\pm0.03$	$0.25\pm0.05$	$0.33\pm0.05$
11	$0.42\pm0.01$	$0.41 \pm 0.04$	$0.42 \pm 0.04$	$0.37\pm0.02$	$0.50\pm0.03$
18	$0.52\pm0.00$	$0.56\pm0.04$	$0.54\pm0.01$	$0.45\pm0.03$	$0.71\pm0.04$
23	$0.67\pm0.02$	$0.69\pm0.09$	$0.67\pm0.01$	$0.58\pm0.03$	$0.92\pm0.03$
34	$0.84\pm0.03$	$0.91\pm0.03$	$0.88\pm0.06$	$0.73\pm0.03$	$1.28\pm0.04$
40	$0.94\pm0.04$	$1.02\pm0.06$	$0.97\pm0.08$	$0.79\pm0.03$	$1.45\pm0.05$
53	$1.15\pm0.04$	$1.28\pm0.09$	$1.17\pm0.12$	$0.96\pm0.06$	$1.83\pm0.06$
82	$1.52\pm0.06$	$1.70\pm0.07$	$1.68\pm0.26$	$1.24\pm0.05$	$2.62\pm0.08$
90	$1.68\pm0.06$	$1.89 \pm 0.09$	$1.87 \pm 0.19$	$1.36 \pm 0.05$	$2.88 \pm 0.11$

Day		С	hemical studies	8	
	PVC B	PVC C	PVC D	PVC F	PVC H
0	0.00	0.00	0.00	0.00	0.00
3	$0.31\pm0.02$	$0.28\pm0.04$	$0.31\pm0.01$	$0.24\pm0.02$	$0.25\pm0.04$
7	$0.52\pm0.01$	$0.54 \pm 0.05$	$0.53\pm0.04$	$0.50\pm0.02$	$0.49\pm0.01$
11	$0.72\pm0.07$	$0.77 \pm 0.07$	$0.78\pm0.04$	$0.69\pm0.01$	$0.66\pm0.03$
18	$0.94\pm0.03$	$1.05\pm0.02$	$1.03 \pm 0.03$	$0.92 \pm \ 0.03$	$0.87\pm0.03$
23	$1.15\pm0.02$	$1.24\pm0.07$	$1.26 \pm 0.01$	$1.07\pm0.02$	$1.06\pm0.05$
34	$1.48\pm0.01$	$1.59\pm0.07$	$1.60\pm0.08$	$1.32\pm0.05$	$1.34\pm0.08$
40	$1.63 \pm 0.03$	$1.75\pm0.09$	$1.77 \pm 0.11$	$1.45\pm0.07$	$1.49\pm0.11$
53	$1.96 \pm 0.04$	$2.16\pm0.10$	$2.15\pm0.23$	$1.75 \pm 0.09$	$1.85\pm0.16$
82	$2.71 \pm 0.01$	$2.93\pm0.14$	$82.9\pm0.14$	$2.29\pm0.16$	$2.53\pm0.20$
90	$2.97\pm0.05$	$3.23\pm0.02$	$3.27 \pm 0.22$	$2.51\pm0.17$	$2.81\pm0.23$

**Table A-11** Percent weight gain of five PVC pipes in 15%TCE + 85%Premiumgasoline at 23°C

 Table A-12 Percent weight gain of PE A pipe in three different gasolines and MTBE

 at 23°C

Day	Percent weight gain (%)				Percent weight gain (%)		
5	Reg-gas	Pre-gas	10%ETOH	MTBE			
0	0.00	0.00	0.00	0.00			
2	$3.42 \pm 0.01$	$3.02\pm0.00$	$3.20\pm0.06$	$0.75\pm0.01$			
4	$4.61\pm0.01$	$4.71\pm0.09$	$4.26\pm0.05$	$1.20\pm0.03$			
7	$6.74\pm0.01$	$6.73\pm0.02$	$6.27\pm0.06$	$1.71\pm0.02$			
9	$7.06\pm0.02$	$7.16\pm0.02$	$6.65\pm0.05$	$2.07\pm0.02$			
11	$7.12\pm0.02$	$7.34\pm0.00$	$6.76 \pm 0.03$	$2.19\pm0.04$			
14	$7.21 \pm 0.04$	$7.32\pm0.02$	$6.85\pm0.05$	$2.65\pm0.01$			
23	$7.31\pm0.04$	1 9 9 99 9	$6.92\pm0.08$	$3.75\pm0.01$			
34				$4.93\pm0.03$			
40				$4.92\pm0.07$			

Day	Percent weight gain (%)				
5	TCE	Benzene	Toluene	Ethylben-	Xylenes
0	0.00	0.00	0.00	0.00	0.00
2	$15.17\pm0.11$	$4.58\pm0.03$	$4.50\pm0.08$	$3.65\pm0.14$	$4.36\pm0.02$
4	$16.16\pm0.03$	$6.83\pm0.03$	$7.06\pm0.08$	$5.64\pm0.17$	$6.73\pm0.06$
7	$16.71\pm0.02$	$7.51 \pm 0.01$	$8.03 \pm 0.00$	$7.11\pm0.03$	$8.05\pm0.05$
9	$16.73\pm0.01$	$7.53\pm0.01$	$8.06 \pm 0.01$	$7.28 \pm 0.02$	$8.10\pm0.06$
11	$16.72\pm0.00$	$7.56\pm0.01$	$8.09\pm0.00$	$7.37\pm0.00$	$8.24\pm0.02$
14		$7.60\pm0.01$	$8.00\pm0.05$	$7.40\pm0.02$	$8.26\pm0.02$
15		$7.61 \pm 0.01$	$8.00 \pm 0.04$	$7.42 \pm 0.00$	$8.28\pm0.02$

**Table A-13** Percent weight gain of PE A pipe in individual BTEX compounds and TCE and MTBE at 23°C

**Table A-14** Flory-Huggins interaction parameter (XI) and percent weight gain (W%)of Toluene-PVC B and TCE-PVC B

No:	#1	#2
Vtce	2.9699	2.8564
Vpvc	4.2337	4.0645
Gpvc	6.1716	5.925
V1	0.4123	0.4127
V2	0.5877	0.5873
Average V1	0.4125	
STD DVN	0.0003	
RSD	0.0007	
%W	70.45	70.58
Average %W	70.51	15
STD DVN	0.0899	0.7
RSD	0.0013	101000
X1	0.86	0.86
Average X1	0.86	
STD DVN	0.0004	
RSD	0.0004	

Pipe No:	#1	#2
Vtoluene	3.3738	3.1353
Vpvc	4.0385	3.7486
Gpvc	5.8870	5.4644
V1	0.4552	0.4555
V2	0.5448	0.5445
Average V1	0.438	
STD DVN	0.0002	
RSD	0.0005	
%W	49.68	49.74
Average W%	49.71	
STD DVN	0.0417	
RSD	0.0008	
X1	0.82	0.82
Average X1	0.82	
STD DVN	0.0002	
RSD	0.0003	

Table A-15 Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%) of TCE-PVC B

 Table A-16 Flory-Huggins interaction parameter (XI) and percent weight gain (W%)
 of Toluene-PVC C

No:	#1	#2
Vtoluene	2.8775	2.7905
Vpvc	3.3843	3.2850
Gpvc	4.9085	4.7646
V1	0.4595	0.4593
V2	0.5405	0.5407
Average V1	0.461	າຍາລຍ
STD DVN	0.0002	10 100
RSD	0.0004	
%W	50.82	50.77
Average %W	50.80	
STD DVN	0.0347	
RSD	0.0007	
X1	0.81	0.81
Average X1	0.81	
STD DVN	0.0002	
RSD	0.0002	

No:	#1	#2
Vtce	2.2331	2.2539
Vpvc	3.1530	3.1898
Gpvc	4.5731	4.6265
V1	0.4146	0.4140
V2	0.5854	0.5860
Average V1	0.414	0.414
STD DVN	0.0000	
RSD	0.0000	
%W	71.49	71.32
Average %W	71.41	
STD DVN	0.1188	
RSD	0.0017	
X1	0.86	0.86
Average X1	0.86	
STD DVN	0.0005	
RSD	0.0006	

Table A-17 Flory-Huggins interaction parameter (X<sub>I</sub>) and percent weight gain (W%) of TCE-PVC C

**Table A-18** Flory-Huggins interaction parameter (X<sub>I</sub>) and percent weight gain (W%)

 of Toluene-PVC D

No:	#1	#2
Vtoluene	2.7904	2.7029
Vpvc	3.3150	3.2124
Gpvc	4.7939	4.6456
V1	0.4570	0.4569
V2	0.5430	0.5431
Average V1	0.4570	
STD DVN	0.0001	1211212
RSD	0.0002	10 1010
%W	50.46	50.44
Average %W	50.45	
STD DVN	0.0152	
RSD	0.0003	
X1	0.81	0.81
Average X1	0.81	
STD DVN	0.0001	
RSD	0.0001	

No:	#1	#2
Vtce	1.9604	2.3918
Vpvc	2.8129	3.2980
Gpvc	4.0678	4.7694
V1	0.4107	0.4204
V2	0.5893	0.5796
Average V1	0.416	
STD DVN	0.0068	
RSD	0.0164	
%W	70.55	73.42
Average %W	71.99	
STD DVN	2.0256	
RSD	0.0281	
X1	0.87	0.85
Average X1	0.86	
STD DVN	0.0080	
RSD	0.0093	

Table A-19 Flory-Huggins interaction parameter (X<sub>I</sub>) and percent weight gain (W%) of TCE-PVC D

**Table A-20** Flory-Huggins interaction parameter (XI) and percent weight gain (W%)of Toluene-PVC F

No:	#1	#2
Vtoluene	4.3337	4.4883
Vpvc	5.1321	5.3144
Gpvc	7.4882	7.7542
V1	0.4578	0.4579
V2	0.5422	0.5421
Average V1	0.4578	ายาวย
STD DVN	0.0000	
RSD	0.0001	
%W	50.17	50.18
Average %W	50.17	
STD DVN	0.0000	
RSD	0.0000	
X1	0.81	0.81
Average X1	0.81	
STD DVN	0.0000	
RSD	0.0000	

No:	#1	#2
Vtce	2.9952	3.1237
Vpvc	4.2526	4.3899
Gpvc	6.2049	6.4053
V1	0.4133	0.4157
V2	0.5867	0.5843
Average V1	0.4145	
STD DVN	0.0018	
RSD	0.0042	
%W	70.67	71.39
Average %W	71.03	
STD DVN	0.5123	
RSD	0.0072	
X1	0.86	0.86
Average X1	0.86	
STD DVN	0.0021	
RSD	0.0024	

Table A-21 Flory-Huggins interaction parameter (X<sub>I</sub>) and percent weight gain (W%) of TCE-PVC F

 Table A-22 Flory-Huggins interaction parameter (XI) and percent weight gain (W%)
 of Toluene-PVC H

No:	#1	#2
Vtoluene	3.8191	3.8017
Vpvc	4.4358	4.4124
Gpvc	6.4782	6.444
V1	0.4626	0.4628
V2	0.5374	0.5372
Average V1	0.4627	2
STD DVN	0.0001	ายาลย
RSD	0.00028	
%W	51.11	51.14
Average %W	0.51	
STD DVN	0.0265	
RSD	0.0518	
X1	0.81	0.81
Average X1	0.81	
STD DVN	0.0001	
RSD	0.0002	

No:	#1	#2
Vtce	3.0935	3.0737
Vpvc	4.3198	4.2944
Gpvc	6.3087	6.2717
V1	0.4173	0.4172
V2	0.5827	0.5828
Average V1	0.4172	
STD DVN	0.0001	
RSD	0.0002	
%W	71.79	71.75
Average %W	71.77	
STD DVN	0.0267	
RSD	0.0004	
X1	0.86	0.86
Average X1	0.86	
STD DVN	0.0001	
RSD	0.0001	

Table A-23 Flory-Huggins interaction parameter (X<sub>I</sub>) and percent weight gain (W%) of TCE-PVC H

 Table A-24 Flory-Huggins interaction parameter (XI) and percent weight gain (W%)
 of Benzene-PE A

No:	#1	#2
Vbenzene	0.1968	0.2070
Vpe	2.4120	2.5351
Gpe	2.2797	2.396
V1	0.0754	0.0755
V2	0.9246	0.9245
Average V1	0.0755	2
STD DVN	0.0000	ายาลย
RSD	0.0006	
% W	7.59	7.59
Average %W	7.59	
STD DVN	0.0052	
RSD	0.0007	
X1	1.94	1.94
Average X1	1.94	
STD DVN	0.0005	
RSD	0.0003	

No:	#1	#2
Vtoluene	0.3029	0.2842
Vpe	3.4422	3.2406
Gpe	3.2534	3.0628
V1	0.0809	0.0806
V2	0.9191	0.9194
Average V1	0.0808	
STD DVN	0.0002	
RSD	0.00215	
%W	8.07	8.04
Average %W	8.06	
STD DVN	0.0189	
RSD	0.0023	
X1	1.89	1.89
Average X1	1.89	
STD DVN	0.0016	
RSD	0.0009	

Table A-25 Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%) of Toluene-PEA

 Table A-26 Flory-Huggins interaction parameter (XI) and percent weight gain (W%)
 of Ethylbenzene-PE A

No:	#1	#2
Vethyl-	0.2156	0.2069
Vpe	2.6679	2.5712
Gpe	2.5215	2.4301
V1	0.0748	0.0745
V2	0.9252	0.9255
Average V1	0.0746	2
STD DVN	0.0002	ายาลย
RSD	0.0026	
%W	7.35	6.98
Average %W	7.16	
STD DVN	0.2636	
RSD	0.0368	
X1	1.95	1.95
Average X1	1.95	
STD DVN	0.0020	
RSD	0.0010	

No:	#1	#2
Vxylene	0.2145	0.2421
Vpe	2.374	2.680
Gpe	2.2436	2.5331
V1	0.0829	0.0828
V2	0.9171	0.9172
Average V1	0.0829	
STD DVN	0.0000	
RSD	0.0003	
%W	8.26	8.26
Average %W	8.26	
STD DVN	0.0023	
RSD	0.0003	
X1	1.87	1.87
Average X1	1.87	
STD DVN	0.0002	
RSD	0.0001	

Table A-27 Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%) of Xylene-PE A

**Table A-28** Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%)of TCE-PE A

No:	#1	#2
VTCE	0.2700	0.2710
Vpe	2.5014	2.5106
Gpe	2.3642	2.3729
V1	0.0974	0.0974
V2	0.9026	0.9026
Average V1	0.0974	ายาลย
STD DVN	0.0000	
RSD	0.00007	
%W	16.72	16.72
Average %W	16.72	
STD DVN	0.0014	
RSD	0.0001	
X1	1.75	1.75
Average X1	1.75	
STD DVN	0.0001	
RSD	0.0000	

No:	#1	#2
Vmtbe	0.17376	0.17745
Vpvc	3.1955	3.2107
Gpvc	3.0202	3.0346
V1	0.05157	0.05237
V2	0.94843	0.94763
Average V1	0.94803	
STD DVN	0.0006	
RSD	0.00060	
%W	4.26	4.33
Average %W	4.30	
STD DVN	0.0494	
RSD	0.0115	
X1	2.24	2.23
Average X1	2.24	
STD DVN	0.0088	
RSD	0.0039	

Table A-29 Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%) of MTBE-PE A

**Table A-30**Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%)of Regular gasoline-PE A

No:	#1	#2
Vgasoline	0.2584	0.2521
Vpvc	2.6972	2.6127
Gpvc	2.5492	2.4694
V1	0.0874	0.0880
V2	0.9126	0.9120
Average V1	0.0877	າຍາລຍ
STD DVN	0.0004	
RSD	0.0046	
%W	7.24	7.29
Average %W	7.26	
STD DVN	0.0400	
RSD	0.0050	
X1	1.83	1.83
Average X1	1.83	
STD DVN	0.0034	
RSD	0.0019	

No:	#1	#2
Vgasoline	0.3053	0.2723
Vpvc	3.1484	2.8027
Gpvc	2.9757	2.6489
V1	0.0884	0.0886
V2	0.9116	0.9114
Average V1	0.0885	
STD DVN	0.0001	
RSD	0.0013	
%W	7.32	7.34
Average %W	7.33	
STD DVN	0.0100	
RSD	0.0014	
X1	1.82	1.82
Average X1	1.82	
STD DVN	0.0009	
RSD	0.0005	

**Table A-31** Flory-Huggins interaction parameter  $(X_I)$  and percent weight gain (W%)of Premium gasoline-PE A

**Table A-32** Flory-Huggins interaction parameter (XI) and percent weight gain (W%)of 10% ethanol gasoline-PE A

No:	#1	#2
Vgasoline	0.2249	0.2140
Vpvc	2.4827	2.3306
Gpvc	2.3465	2.2027
V1	0.0831	0.0841
V2	0.9169	0.9159
Average V1	0.0836	າຍາລຍ
STD DVN	0.0007	1011012
RSD	0.0087	
%W	6.84	6.93
Average %W	6.89	
STD DVN	0.0700	
RSD	0.0095	
X1	1.87	1.86
Average X1	1.86	
STD DVN	0.0065	
RSD	0.0035	

### **APPENDIX B**

### Determination of volume of pipe specimens by water displacement

The overflow can was over-filled with distilled water of known temperature and allowed to drain for 5 minutes. The edge of a beaker was touched to the tip of the spout to remove the remaining drop of water. The collection can was weighed and placed beneath the spout. The pipe specimen was immersed in the overflow can and the displaced water was allowed to drain completely (about 5 minutes). The edge of the collection can was touched to the tip of the spout in order to collect the last drop of water before weighing. The collection can plus the displaced water was weighed and the volume of the specimen calculated as follows:

$$\mathbf{V} = (\mathbf{W}_{c+w} - \mathbf{W}_c)/\mathbf{d}_t$$

where

V = volume of the specimen, cm<sup>3</sup>

 $W_{c+w}$  = weight of the collection can plus displaced water

 $W_c$  = weight of the empty collection can

 $d_t$  = density of the distilled water at the measured temperature, g/cm<sup>3</sup>



# BIOGHAPHY

Miss. Parichat Prasomsat was born on November 14, 1980 in Nakornratchasima, Thailand. She graduated in high school level in 1999 from Phimai Wittaya School. She achieved her Bachelor's degree in Science from Department of Biotechnology, Faculty of Technology, KhonKaen University in 2003.

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