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REVERSE OSMOSIS REMOVAL OF ORGANIC COMPOUNDS I. A PRELIMINARY LITERATURE REVIEW

ARCADIO P. SINCERO, DSc, PE

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U.S. ARMY BIOMEDICAL RESEARCH & DEVELOPMENT LABORATORY
Fort Detrick
Frederick, MD 21702-5010

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The subject review concludes that present reverse osmosis (RO) technology cannot insure production of drinking water quality effluent in all instances, but that the technology may be improved by staging the RO units. This may be done by optimizing power cost and permeate water flux (objective functions) with the sole constraint that an arrangement effect 100 percent rejection of solute, The report also recommends serching for a better membrane than polyethylenimine (PEI) and continuation of the literature review to cover membrane fouling, disposal of concentrates, operation costs, and removal of bacteria, protozoa and viruses. The review finds that PEI is the best candidate membrane for optimization studies at this time.

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PREFACE

Dr. Arcadio P. Sincero is Assistant Professor of Civil Engineering, Morgan State University, Baltimore, MD. This literature review was conducted under Contract DAAL03-86-D-0001 of the U. S. Army Summer Faculty Research and Engineering Program, sponsored by the Department of the Army and the U.S. Army Biomedical Research and Development Laboratory (USABRDL), Fort Detrick, MD, through Battelle, Columbus Division. The Contracting Officer's Representative for USABRDL was Catherine A. Connor. This study supported the research program of the USABRDL Unit Process Laboratory, W. Dickinson Burrows, director.

INTRODUCTION

The purpose of this review is to gather relevant information regarding the current state-of-the-art of removing organic chemicals in water using reverse osmosis (RO). The review includes theories that have been developed regarding organic solute removals from water and organic compounds that have been studied.

The basics of a normal osmotic process are shown in Figure 1.[1] A bag of semipermeable membrane is shown placed inside a bigger container containing pure water. Inside the membrane bag is a solution of a solute, sucrose. Because sucrose has osmotic pressure, it "sucks" water from outside the bag causing the water to pass through the membrane. Introduction of the water into the membrane bag, in turn, causes the solution level to rise as indicated by the height, B, in the figure. It follows that if sufficient pressure is applied to the tip of the tube in excess of that of the osmotic pressure the height, B, will be suppressed and the flow of water through the membrane will be reversed, i.e., it would be from inside the bag towards the outside into the bigger container. For sucrose, this pressure would have to be in excess of 1.05 psia (pounds per sq. in. absolute) if the concentration is of the order of 1,000 mg/L; for NaCl, it would have to be in excess of 398 psia if the concentration is in the order of 35,000 mg/L.[2] The operation just described, i.e., applying sufficient pressure to the tip of the tube to reverse the flow of water, is the fundamental description of a basic reverse osmosis process.

Reverse osmosis is just one of the three allied membrane methods that have been used to separate solute from solvent water: ultrafiltration (UF), nanofiltration, and reverse osmosis. According to Jacangelo, ultrafiltration removes particles ranging in sizes from 0.001 to 10 microns while reverse osmosis can remove particles ranging in sizes from 0.0001 to 0.001 microns.[3] As far as size removals are concerned, nanofiltration stays in-between ultrafiltration and reverse osmosis, being able to remove particles in the size range of the order of 0.001 microns. Ultrafiltration is normally operated in the range of 15 to 75 psig (pounds per sq. in. gage); nanofiltration, in the range of 75-250 psig; and, reverse osmosis, in the range of 200-1,200 psig. These pressures corrected for osmotic pressure are the excess pressures referred to above in the fundamental description of the process of reverse osmosis.

Jacangelo has developed a brief chronology on the growth of membrane technology for water treatment as follows:[3] In the 1950s, the cellulose acetate (CA) membrane was developed. In the 1960s, there were several developments: the casting techniques for CA membranes were improved, flux relationships for RO membranes were developed, the first UF manufacturing firm was founded, spiral wound membranes were developed, and large-scale manufacturing of CA membranes was made possible. The first thin film composite membrane was developed, and the first large-scale desalination plant was put on-line and operated satisfactorily in the 1970s. The 1980s saw the following further developments: water softening membranes developed and marketed, the largest desalination plant of 72 mgd constructed in Yuma, Arizona, and particulates removal in drinking water by ultrafilters investigated.

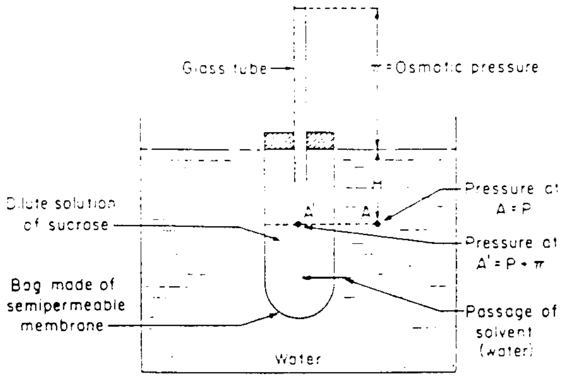


Figure 1. A normal osmosis process (1)

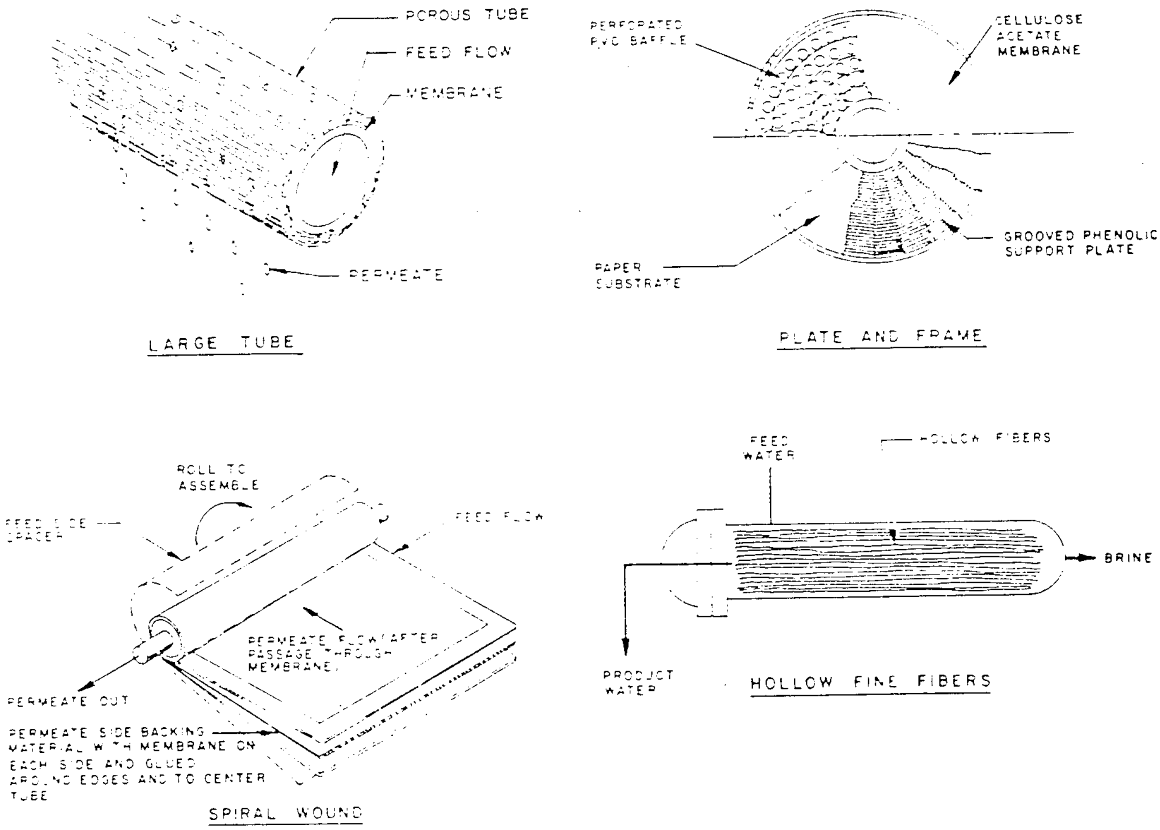


Figure 2. Reverse osmosis module designs (2)

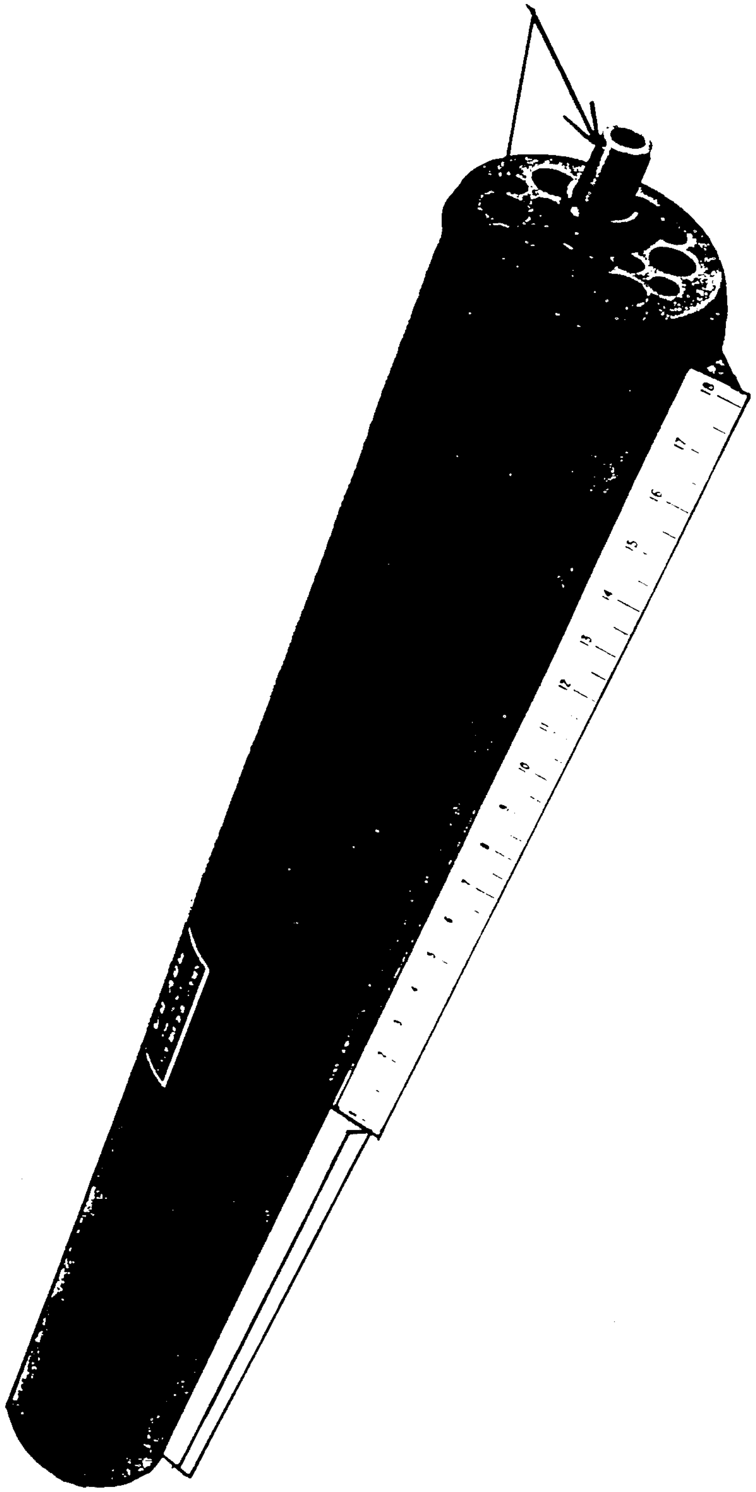


Figure 3. A tubular reverse osmosis module (4)

Over the course of the development of membrane technology, modular designs as shown in Figure 2 evolved. They are tubular, plate-and-frame, spiral wound, and hollow fine fiber modules. In the tubular design, the membrane is lined inside the tube, which is made of ordinary tubular material.[1] Wastewater or other feedwater is allowed to pass through the inside of the tube under excess pressure, causing the water to permeate through the membrane and to collect at the outside of the tube. The plate-and-frame design is similar to the plate-and-frame press as used in chemical engineering practice. In the case of RO, the semipermeable membrane replaces the filter cloth. The spiral wound design consists of two flat sheets of membranes separated by porous spacers. The two sheets are sealed on the three sides; the fourth side is attached to a central collector pipe and the sealed sheets are rolled around the central pipe. As the sheets are rolled around the pipe, a second spacer is provided between the sealed sheets. In the final configuration, the spiral wound sealed membrane forms a cylinder, as shown in Figure 3.[4] Note the arrows pointing towards the central pipe collector and the holes leading to the second spacer, respectively, of the spiral-wound module. Wastewater is introduced under pressure into the second spacer, thereby allowing the water to permeate through the membrane and into the spacer between the sealed membranes. Henceforth, the permeate continues on to the central pipe collector and discharges as a permeate product. The concentrated wastewater exits at the other end of the cylindrical module. The hollow fiber modules are a bundle of thousands of parallel, self-supporting, hairlike fibers enclosed in a fiberglass or epoxy-coated steel vessel.[3] Wastewater is introduced into the hollow bores of the fibers under pressure. The permeate water exits through one or more module ports. The concentrate exits through one or more module ports, depending on design. These modules can be combined into banks of modules, and they may be connected in parallel or in series.

THEORIES OF SOLUTE REMOVAL

This chapter aims to unify the theories of solute removal from available literature. Theoretically, if the solute did not reach the membrane, it would be totally separated from its solvent water. It would simply go out with the concentrate (or retentate) and have no chance of ever passing through the thickness of the membrane. Invoking Prandtl's hypothesis, developed in 1904, if there is velocity in the bulk solution region, a fluid boundary layer exists as a thin film close to the surface of the membrane. This layer is laminar with velocity approaching zero as the flow field encounters the membrane surface. Because of the stagnation at the surface, solute species from the bulk solution would be transported through the film and be lodged at the surface, unless some means is provided to dislodge them. By analogy to the fluid boundary layer, a concentration boundary layer is also created (See Figure 4).[5] If there were no bulk flow, it would be immaterial whether or not a concentration boundary layer exists. The solute would still lodge on the membrane surface if solute and membrane have affinity towards each other, increasing the chance of permeating through and not being removed or rejected.

In Figure 4, XA1 and XA2 are the solute concentrations in the bulk and the membrane surface, respectively. Since the velocity in the bulk stream is greater than that near the surface of the

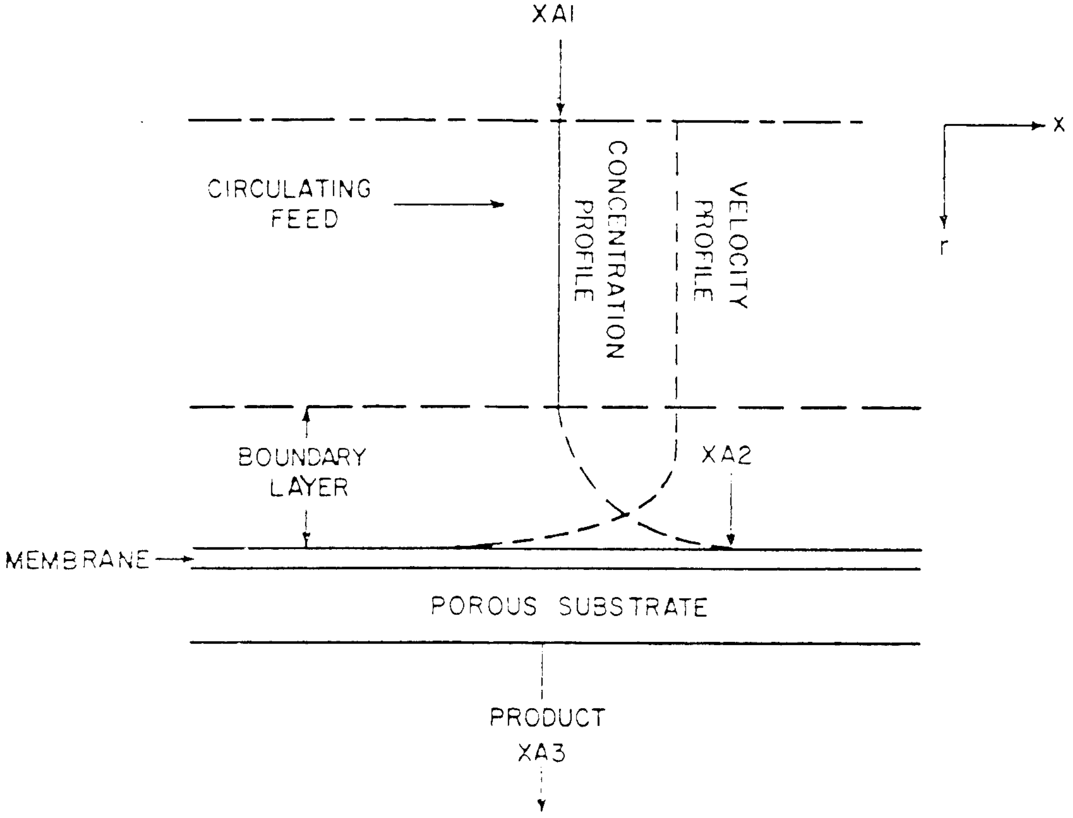


Figure 4. Fluid velocity and concentration boundary layers (5)

membrane, while the corresponding values for solute concentration are the opposite, the velocity and concentration profiles in the boundary layer are the inverse of each other, as shown in the figure. Because of solute accumulation, the concentration at the membrane surface is greater than that in the bulk solution, a situation called concentration polarization. This much is well accepted in fluid mechanics theory. The question is, once the solute is entrapped in the boundary layer and lodged on the membranes surface, how does the solute (or the solvent) move through the membrane thickness from the inflow side to the other to form the permeate? For this, several theories have been developed which are the subject of this chapter.

Dependence of Removal on Functional Groups

Duvel and Helfgott[6] and Fang et al.[7] demonstrated that removals are dependent on the chemical nature of the organic functional groups. Table 1 presents the functional groups: carboxylic acid, amine, amide, alcohol, sulfone, aldehyde, ketone, and two isomeric esters in order of increasing rejection. The lowest removal is represented by the acid, butyric and the highest removal, by the ester, ethyl acetate. (See Figure 5 for the functional groups.[8]) When one of the two atoms in a molecule shares more of the electrons and the other less, one end of the molecule becomes more negative than the other; this situation makes the molecule behave like a bar magnet, or dipole. One end of the dipolar molecule attracts the unlike end of another molecule, creating an electrostatic bond between them. When one atom involved is hydrogen, the bond is called a hydrogen bond. A molecule that can both donate and accept a proton forms stronger hydrogen bonds than those that can accept only. Since alcohols, amides, amides, and carboxylic acids are capable of both donating and accepting protons, they form strong hydrogen bonds.[6] Referring to Table 1, it has been theorized that butyric acid bonds itself to an active site on the RO membrane by hydrogen bonding.[6] Since the bonding is strong, more solute population species are available for migration across the membrane; they then appear in large amounts on the permeate side resulting in a very poor rejection. Just how the population migrates across the membrane will be discussed shortly.

The data indicate an increasing trend of removals from butyric acid to ethyl acetate. This suggests a decreasing trend of species population at the membrane surface as we go down the table from butyric acid to ethyl acetate. Since less is available, less will emerge at the permeate side, resulting in progressively higher rejection. This decreasing trend of solute population (increasing trend of removals) can be explained in terms of the decrease in hydrogen bonding force. Thus, ethyl acetate has the least bonding force, resulting in the lowest population at the membrane surface and the highest rejection of the group studied. (The order shown in Table 1 may or may not hold for other membranes.)

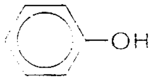
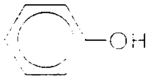
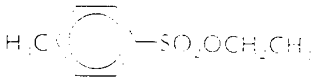
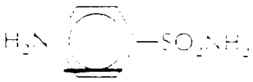
FUNCTIONAL GROUP NAME	FUNCTIONAL GROUP	EXAMPLE	USE OR OCCURRENCE OF EXAMPLE
ALCOHOLS, PHENOLS, ETHERS, AND SULFUR ANALOGUES:			
Alcohol	$-C-OH$	CH_3CH_2OH	beverage alcohol
Phenol			antiseptic, local anesthetic
Ether	$-C-O-C-$	$CH_3CH_2OCH_2CH_3$	diethyl ether—general anesthetic
Thiol	$-C-SH$	$CH_3CH_2CH_2SH$	propanethiol—from fresh onions
Sulfide	$-C-S-C-$	$CH_2{=}CHCH_2-S-CH_2CH{=}CH_2$	allyl sulfide—flavor and odor of garlic
ORGANIC NITROGEN COMPOUNDS:			
Amine	$-N-$	CH_3NH_2	methylamine—fishy odor of herring brine
Nitrile	$-C\equiv N$	$CH_2{=}CH-C\equiv N$	acrylonitrile—used to make Orlon
ORGANIC HALOGEN COMPOUNDS:			
Halides	$-C-X$	CCl_2F_2	freon—refrigerant and aerosol propellant
SULFONIC ACIDS AND DERIVATIVES:			
Sulfonic acid	$-C-SO_3H$		p-toluenesulfonic acid—used in dye chemistry and in manufacture of antidiabetic drugs
Sulfonyl chloride	$-C-SO_2Cl$	CH_3SO_2Cl	methanesulfonyl chloride—organic synthesis
Sulfonate ester	$-C-SO_2OR$		ethyl *p*-toluenesulfonate—organic synthesis
Sulfonamide	$-C-SO_2N-$		sulfanilamide—sulfa drug

Figure 5. Organic functional groups (8)

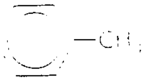
FUNCTIONAL GROUP NAME	FUNCTIONAL GROUP	EXAMPLE	USE OR OCCURRENCE OF EXAMPLE
HYDROCARBONS:			
Alkane	$-\overset{\mid}{\underset{\mid}{C}}-\overset{\mid}{\underset{\mid}{C}}-$	$CH_3CH_2CH_3$	propane—rural or camping gas
Alkene	$>C=C<$	$CH_2=CH_2$	ethene—precursor of polyethylene
Alkyne	$-C\equiv C-$	$HC\equiv CH$	acetylene—used in oxy-acetylene torches
Aromatic			benzene, toluene—high octane gasoline components
CARBOXYLIC ACIDS AND DERIVATIVES:			
Carboxylic acid	$-\overset{O}{\overset{\|}{C}}OH$	$CH_3\overset{O}{\overset{\|}{C}}OH$	acetic acid—vinegar acid
Acid chloride	$-\overset{O}{\overset{\|}{C}}Cl$	$CH_3\overset{O}{\overset{\|}{C}}Cl$	acetyl chloride—organic synthesis
Acid anhydride	$-\overset{O}{\overset{\|}{C}}O\overset{O}{\overset{\|}{C}}-$	$CH_3\overset{O}{\overset{\|}{C}}O\overset{O}{\overset{\|}{C}}CH_3$	acetic anhydride—organic synthesis
Ester	$-\overset{O}{\overset{\|}{C}}O\overset{\mid}{C}-$	$H\overset{O}{\overset{\|}{C}}OCH_2CH_3$	ethyl formate—artificial rum-flavoring agent
Amide	$-\overset{O}{\overset{\|}{C}}-\underset{\mid}{N}-$	$H_2N\overset{O}{\overset{\|}{C}}NH_2$	urea—found in urine
ALDEHYDES AND KETONES:			
Aldehyde	$-\overset{O}{\overset{\|}{C}}H$	$H\overset{O}{\overset{\|}{C}}H$	formaldehyde—biological preservative
Ketone	$-\overset{\mid}{\underset{\mid}{C}}-\overset{O}{\overset{\|}{C}}-\overset{\mid}{\underset{\mid}{C}}-$	$CH_3\overset{O}{\overset{\|}{C}}CH_3$	acetone—fingernail polish remover

Figure 5 (cont). Organic functional groups (8)

Table 1. Rejection of Selected Organics Based on Functional Groups[6]

Solute	Solute Rejection, %
Butyric Acid	16.4
1-Butylamine	39.2
Butyramide	40.5
1-Butanol	41.3
Diethyl sulfone	67.9
Butyraldehyde	72.1
2-Butanone	72.9
Methyl propionate	89.1
Ethyl acetate	91.1

The relationship of functional group to percentage rejection can also be seen in Figures 6-9.[5,9,10] Test results are presented graphically for alcohols (methanol. ethanol and isopropyl alcohol), a carboxylic acid (acetic acid), an aldehydes (formaldehyde), a ketones (acetone), an ether (diethyl ether), a polyhydroxy alcohol (glycerol), phenols (hydroquinone and phenol), an amide (urea) and an ester (methyl acetate). For example, Figure 6 shows results for three different cellulose acetate membranes, CA_{blend}, CA_{KP98}, and CAB.[9] For the CAB membrane (solid line) the alcohols range in removals from negative for the one-carbon alcohol to around 42% for isopropyl alcohol. For the two-carbon acid the removal is approximately 12 percent, contrasted with approximately 1 percent removal for the two-carbon atom alcohol . For the one-carbon aldehyde removal is approximately 42 percent, to be contrasted with negative removal for the one-carbon alcohol. These results show that for the same number of carbon atoms per molecule, different functional groups (alcohol, acid, aldehyde) have different percentage removals. Note that for glycerol the percentage removal is around 90 percent; much, much greater than for the lower alcohols. The reason for this will be discussed later. Note also that the order of rejection is not the same as in Table 1.

One observes parallel behavior for the other cellulose acetate membranes. In fact, similar observations hold for Figures 7-9, i.e., <u>percentage removal is dependent on functional groups</u>.[6,7,11] It should be noted that as one scans through the figures, percentage removals for the groups are not consistent. For example, phenol in Figure 6 is shown to be 10 percent removed by the CAB membrane; the same phenol is shown in Figure 7 to be ca. 90 percent removed by a polyamide membrane (1500 psig). This means that the <u>percentage removal is dependent not only on functional groups but also on the nature of the membrane surface</u>.

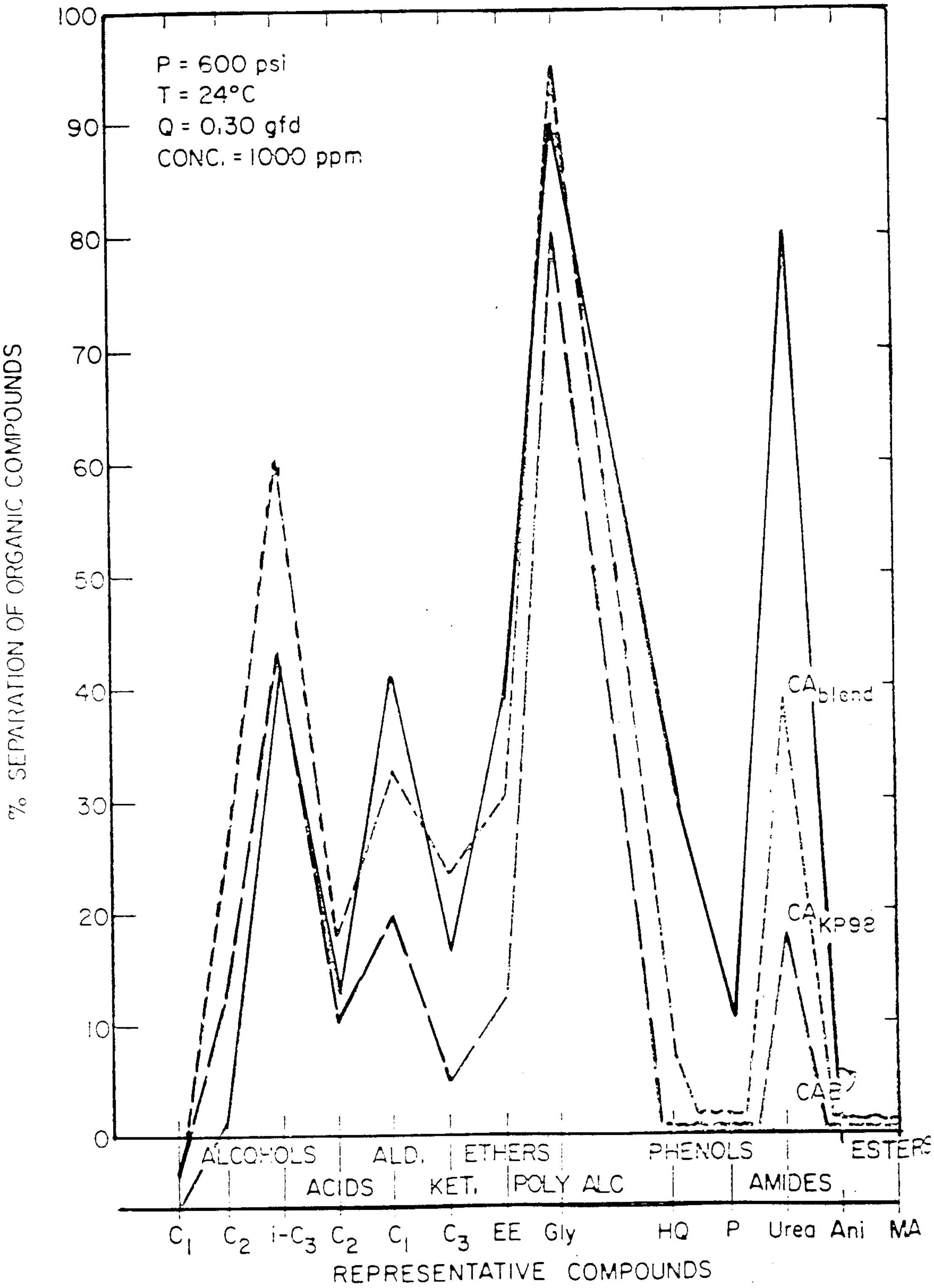


Figure 6. Representative functional group compounds vs. percentage separation (9)

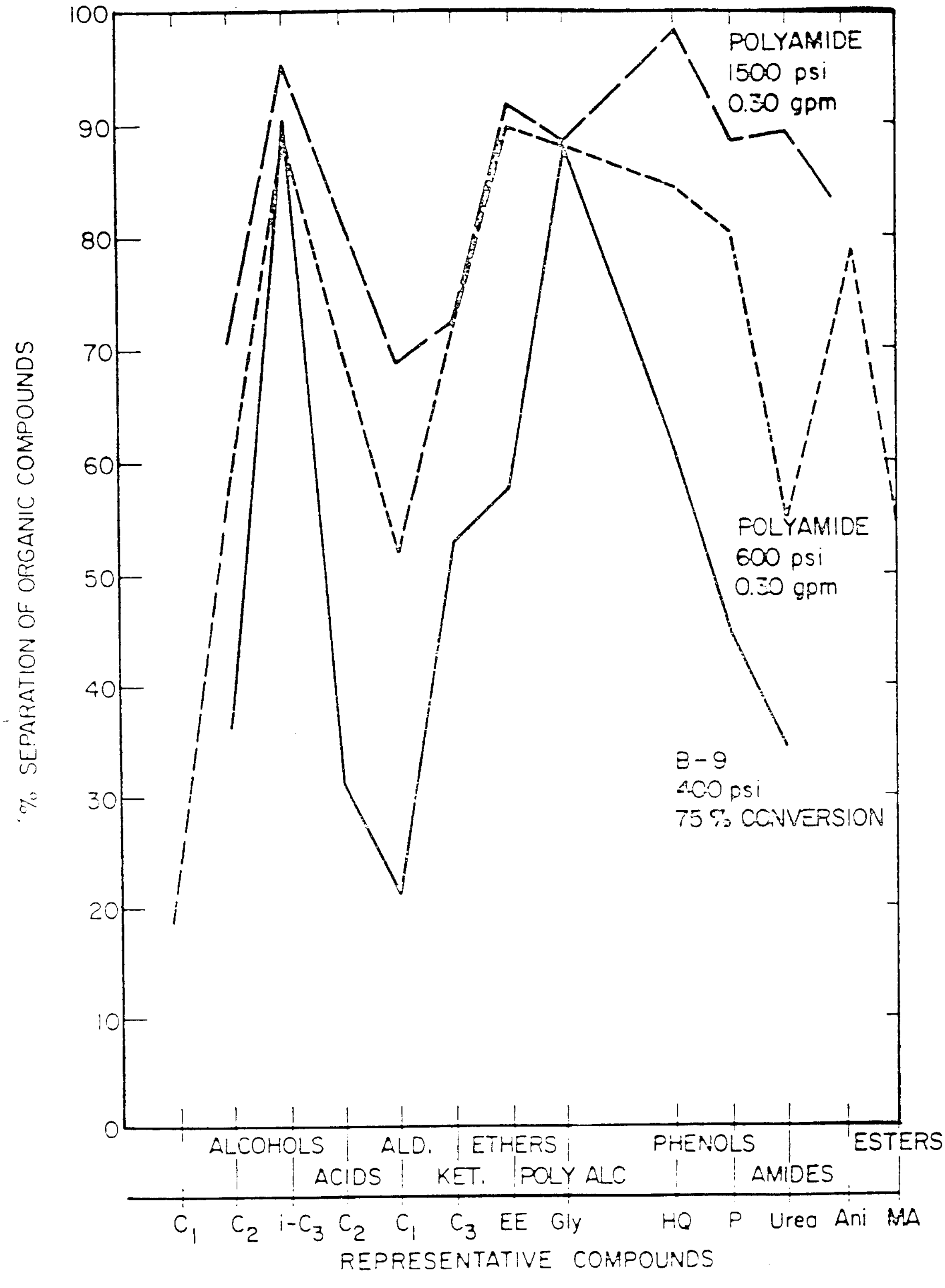


Figure 7. Representative functional group compounds vs. percentage separation (9)

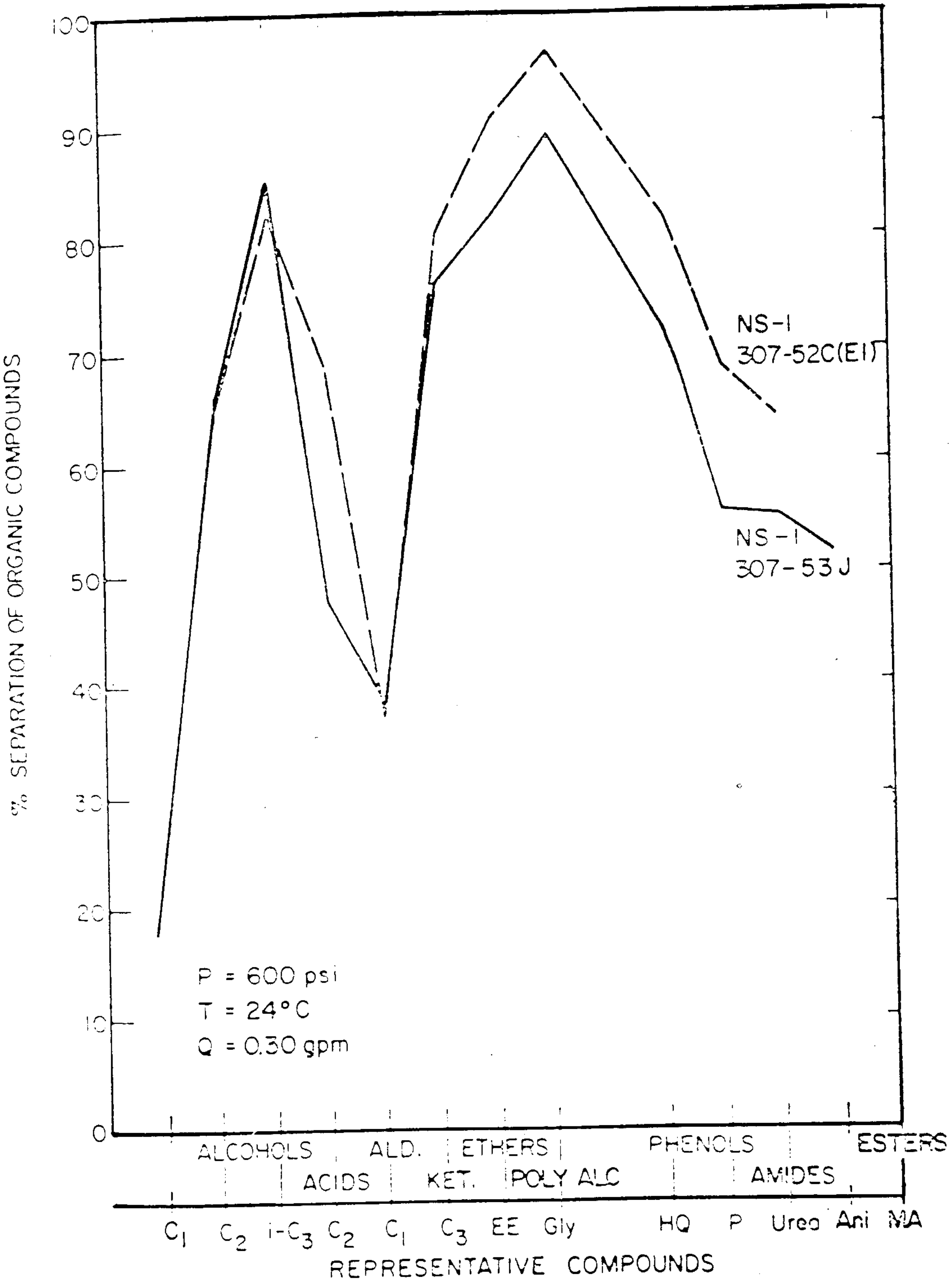


Figure 8. Representative functional group compounds vs. percentage separation (5)

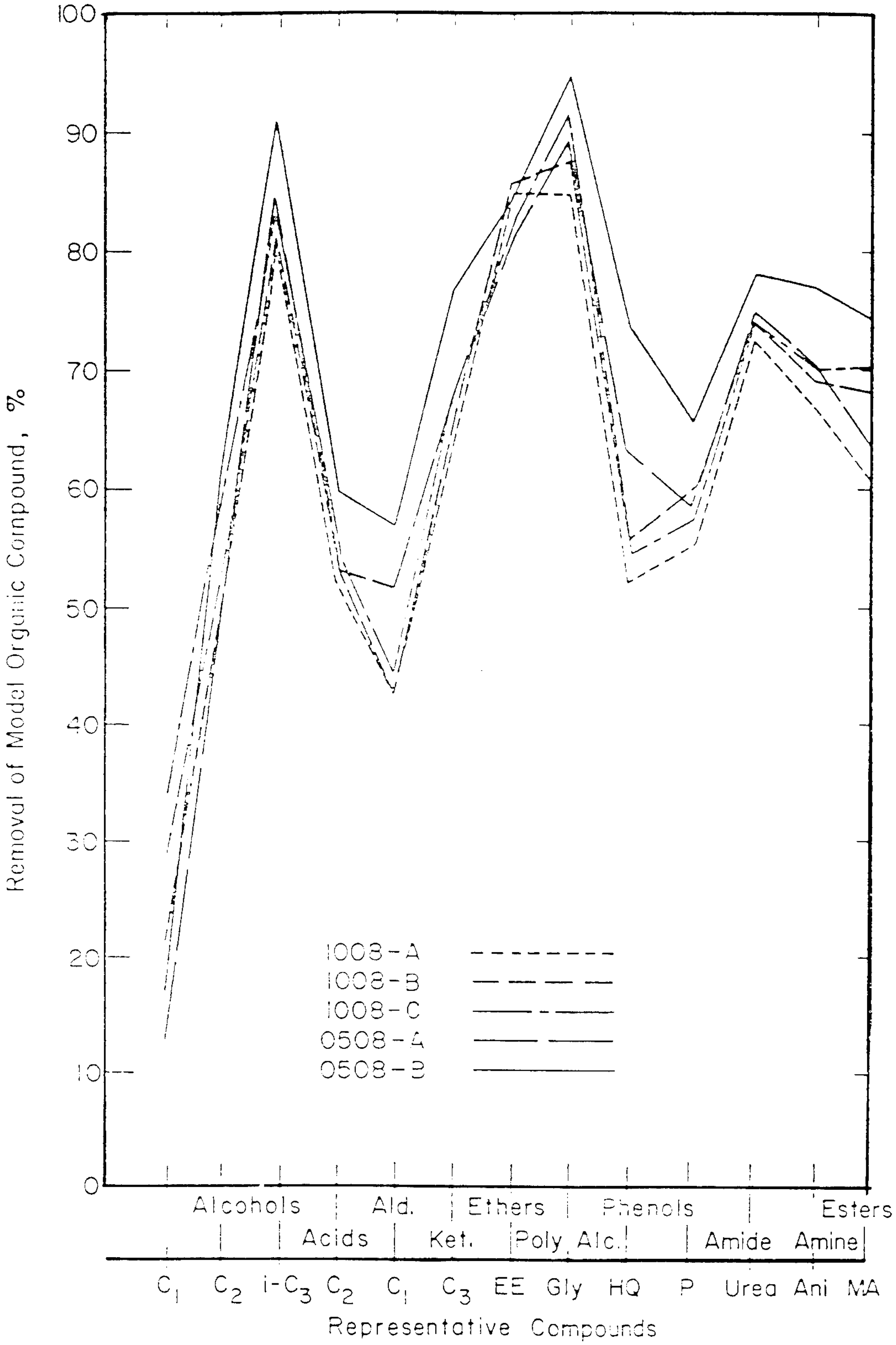


Figure 9. Representative functional group compounds vs. percentage separation (10)

Dependence of Removal on Molecule Weight and Size

The two curves in Figure 10 and the three curves in Figure 11 illustrate percentage removals using different membranes for six normal alkyl alcohols and seven normal undissociated alkyl acids, respectively.[10] For example, in Figure 10, the lower curve is for Membrane 0316 B and the upper curve for Membrane 0316 A. Figure 11 shows percentage removals for the aliphatic acid group using three different membranes, 0517 A, 0517C, and 0517B. Fang and Chian observed that for a homologous series of a given functional group removal increases with molecular weight.[10] In Figures 10 and 11, the number of carbon atoms increases from one to six or seven, respectively, corresponding to increases in molecular weights. Therefore, as shown, for a homologous series removal increases as the molecular weight increases.

For straight chain compounds, Duvel and Helfgott explained the increase in percentage removals with increase in molecular weight.[6] They stated that data indicate elongated molecules diffuse through membranes with the long axis parallel to the direction of diffusion. As diffusion progresses the molecules bend and twist randomly [due to thermal energy] creating an increased projected area in the direction of diffusion. Hence, they become geometrically larger, i.e., the longer they are, the larger they become. Duvel and Helfgott further suggested that the membrane has an active layer [called the skin layer] which is responsible for retention. The layer is a continuum having alternate amorphous and crystalline regions. For the molecule to pass through, a gap must be produced. This is created by the random thermal motion of the polymer chains of the membrane. Up to a limit, the size of the gap is a function of the thermal energy imparted to the polymer chain. Larger gaps require larger energy. Since the larger molecules will naturally require larger gaps, more energy is required to let them through. However, because gaps are normally small, consistent with existing thermal energy of the solution, the larger molecules must diffuse slower. This condition results in lower permeation rates and higher percentage removals.

Figures 12-14 show percentage removals of alcohols, amines, and acids, respectively.[5] As before, one can see that as the molecular weight increases, percentage removal increases. For example, the curves in Figure 12 all have positive slopes. Additional information, however, has been added to these figures. Consider the lower curves in Figure 12 for the normal alcohols, then consider the iso- and tertiary curves. As shown, the percentage removals increase from normal to tertiary alcohols, i.e., with increased branching. For a molecule which must pass through a gap, a tertiary alcohol has a broader cross section than an iso-alcohol of the same carbon number, which in turn is broader than a normal alcohol. The greater apparent molecular size with increased branching explains the increase in percentage removal from normal to tertiary alcohols. The same explanation holds for the primary and secondary amines in Figure 13 and the normal and the iso-acids in Figure 12. In other words, the branching configuration affects removals. Hence, percent removal is also a function of the apparent size of the molecule.[5,6,10] (However, shielding of the functional group may also play a role.)

This brings us to the case of the polyalcohol, glycerol, which was removed to approximately 90

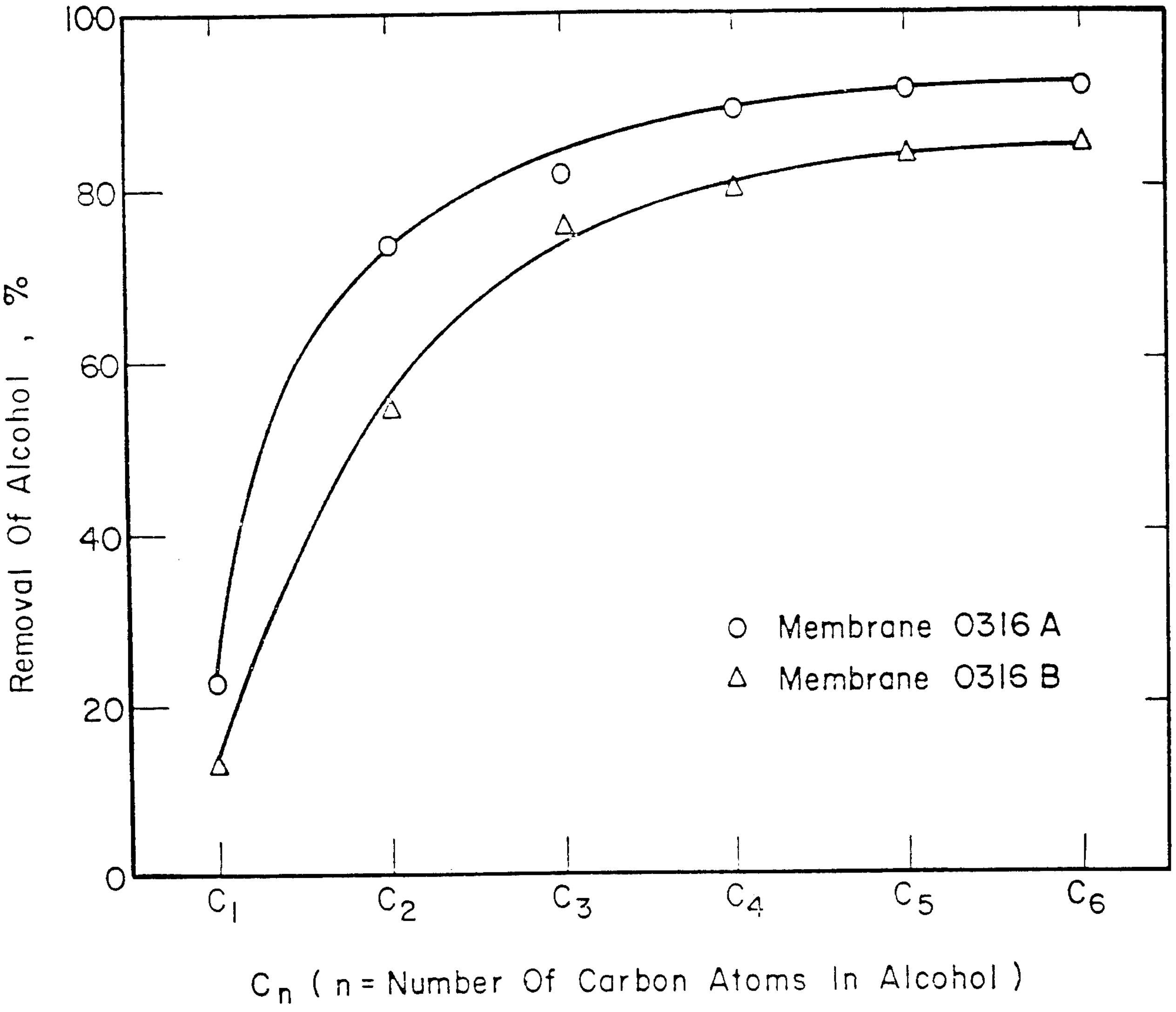


Figure 10. Effect of molecular weight on percentage removal (10)

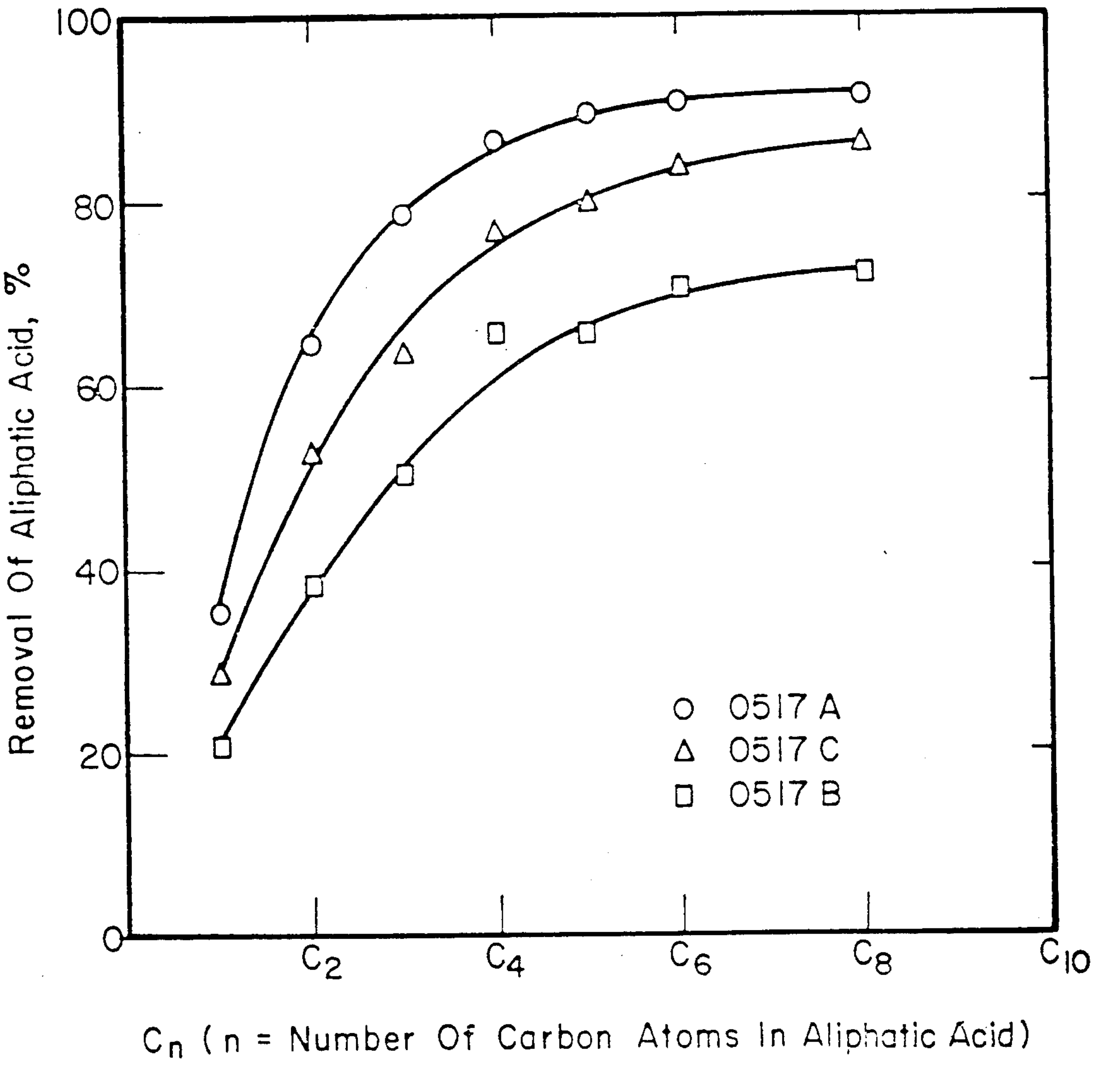


Figure 11. Effect of molecular weight on percentage removal (10)

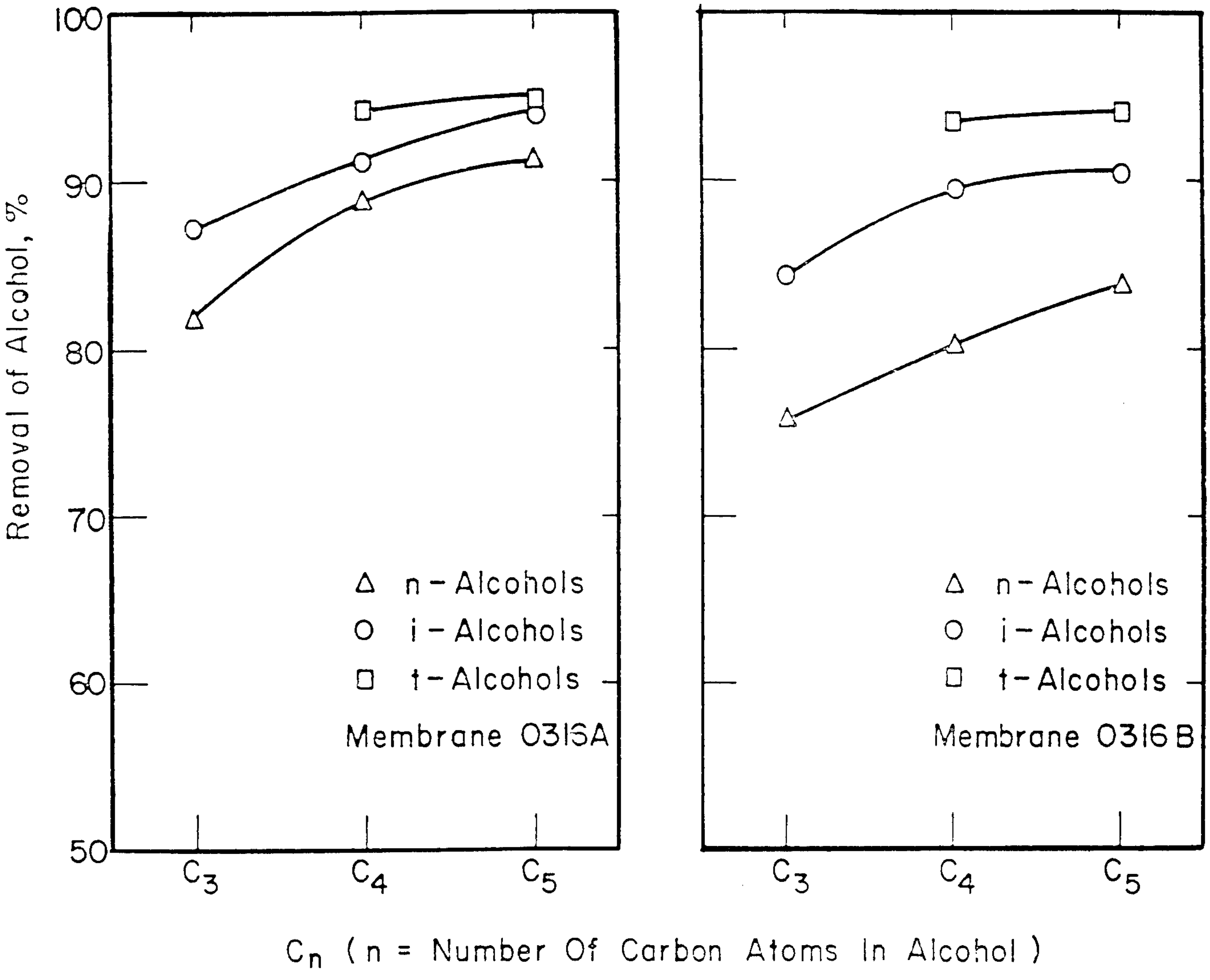


Figure 12. Effect of molecular configuration on percentage removal (5)

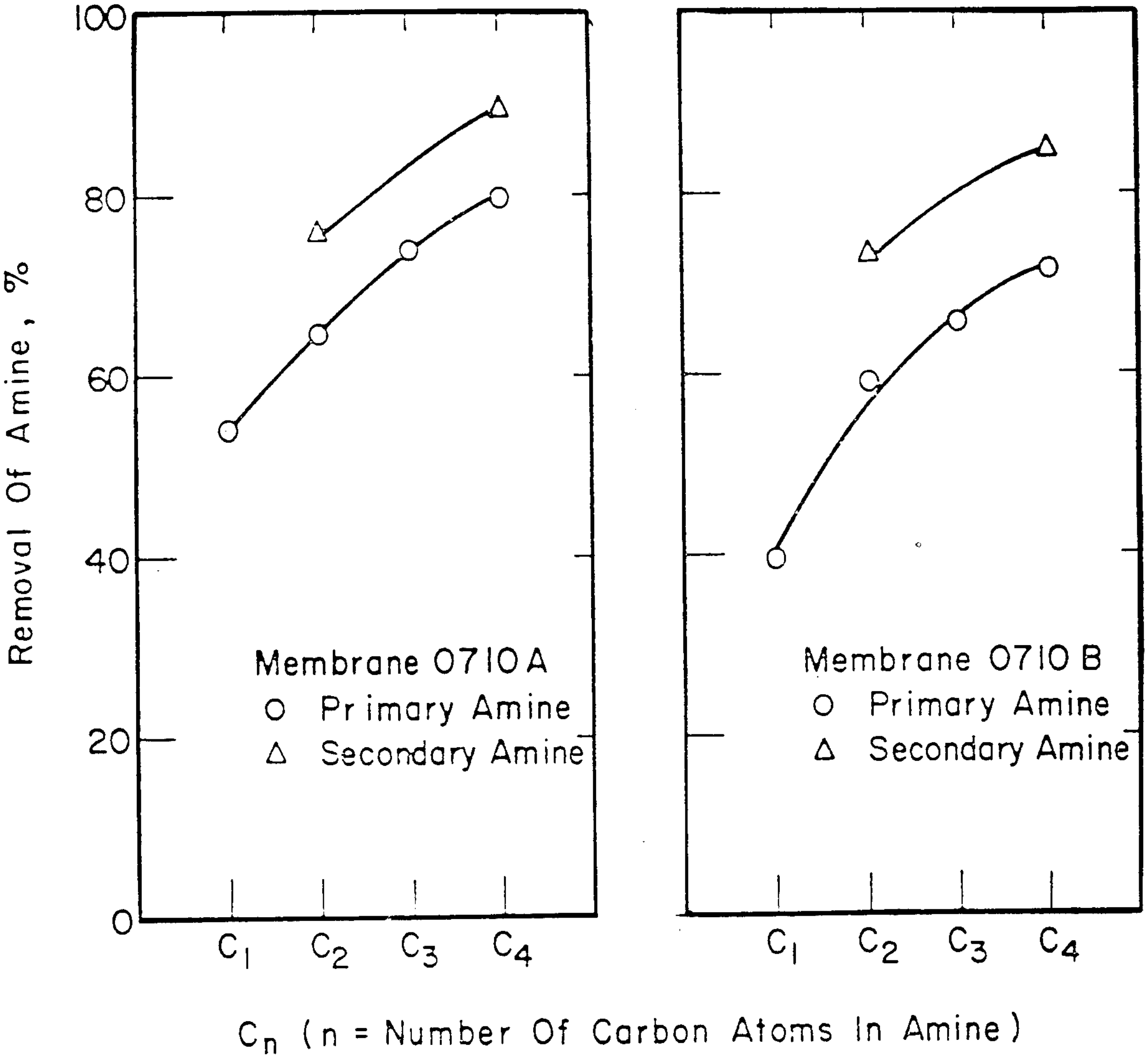


Figure 13. Effect of molecular configuration on percentage removal (5)

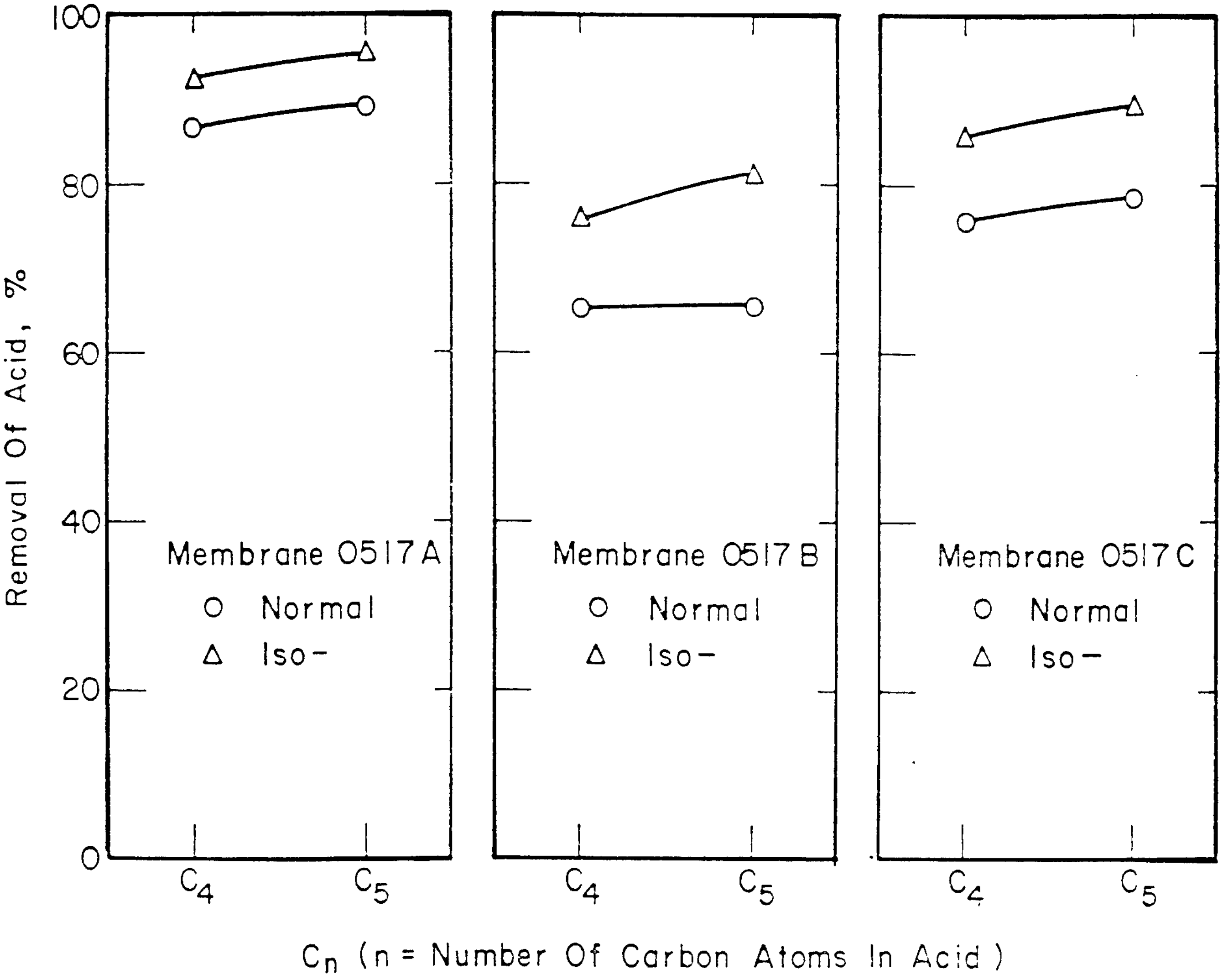


Figure 14. Effect of molecular configuration on percentage removal (5)

percent (Figure 6).[9] There can be two considerations for the removal of glycerol. In the first case, glycerol has three hydroxyls that can help in binding itself to the membrane surface, increasing the glycerol solute species population there. With more "bodies" available for passage through the membrane, the rejection should decrease. On the other hand, glycerol is also a secondary alcohol, which makes the molecule expand its size. In addition, the three hydroxyls are attached to the side of the chain, augmenting the spatial volume of the molecule. Insofar as the gap theory is correct, it would appear that apparent molecular size (configuration) is more effective in hindering passage than is accumulation of more solute on the membrane surface or hydrogen bonding in promoting passage.[12] Two experimental results of Duvel and Helfgott using cellulose acetate as the membrane may also be contrasted.[6] They obtained the following percentage removals: for straight-chain alcohols: 2-buten-1-ol, 18.3 percent; 3-buten-1-ol, 28.3 percent; 3-butyn-1-ol, 12.3 percent; for diols: 1,2-ethanediol, 52.2 percent; 1,3-propanediol, 64.1 percent; 1,4-butanediol, 65.9 percent; 1,5-pentanediol, 62.0 percent. The high removals of the diols compared to those of the straight-chain alcohols follows the same argument as that for glycerol, i.e., the diols have larger spatial configuration. In addition, the two hydroxyl groups of the diols at opposite ends may have the tendency to "curl" the molecules through intramolecular hydrogen bonding, increasing their apparent sizes all the more.[6]

Dependence of Removal on Degree of Dissociation and pH

Anderson et al. have shown that percentage removals can be enhanced by changing the pH.[13] When they increased the pH of the solution from 3.0 to 11.0, the percentage removal of p-nitrophenol increased from -40 percent to +84 percent. Figure 15 shows the effect of pH and degree of dissociation on percentage removal of phenol. As shown, the removal increased from a negative value to ca. 90 percent. The pKa of phenol is 9.9 at 25 °C.[14] Therefore, at a pH of 9.9, the degree of dissociation is 50 percent (since pH = pKa + logB/A). From Figure 15, at 50 percent dissociation, the percentage removal is approximately 40 percent; this corresponds to a pH of ca. 10. (It is uncertain if the temperature in Figure 15 was 25 °C.) At a pH of 12.5, the percentage removal is approximately 90 percent, corresponding to practically complete dissociation. Figure 16 is a plot of percentage removal of acetic acid against its degree of dissociation. Figures 17 and 18 are similar plots for formic acid and methyl amine, respectively. These plots clearly show a positive correlation. Therefore, percentage removal increases as the percentage ionization increases. It is to be noted that unionized methylamine is a relatively small molecule with molecular weight of 31, not quite twice that of water.

The positive correlation of removal of carboxylic acids with degree of dissociation may be explained in more than one way. Since the molecule has already been dissociated, it is "dissolved" in water. At the boundary layer, the ions have the choice of whether to adhere to the skin and be available for migration across the membrane or to stay in solution. Since the ions are more soluble in water than they would be in the solid membrane skin, they should stay in solution. Another explanation would be that the ions are hydrated. In the case of the reverse osmosis process for inorganic solutes, it is known that the cations and anions are surrounded by waters of hydration.[5] This condition makes the ions bulky for passage across the membranes. The same could also be

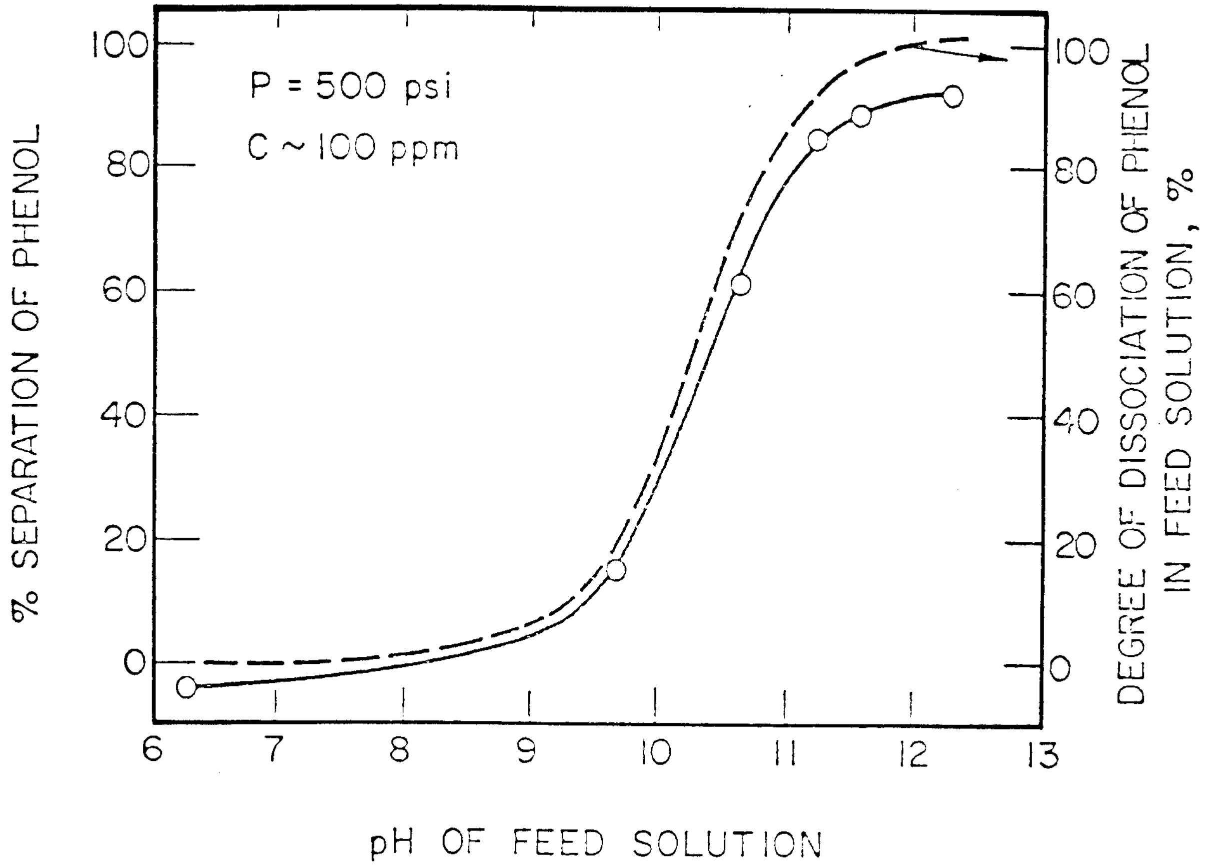


Figure 15. Effect of pH and degree of dissociation on percentage removal (5)

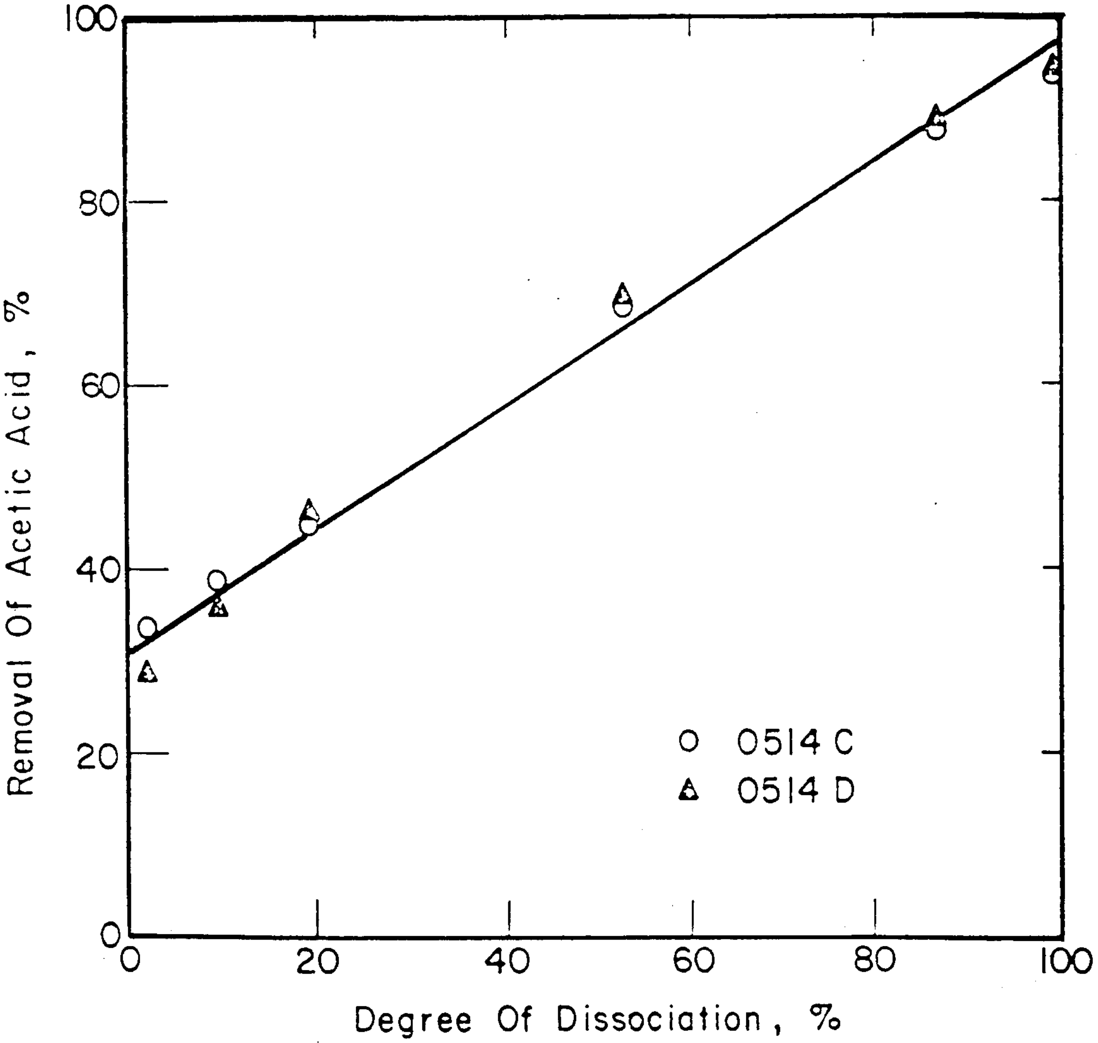


Figure 16. Effect of degree of dissociation on percentage removal (10)

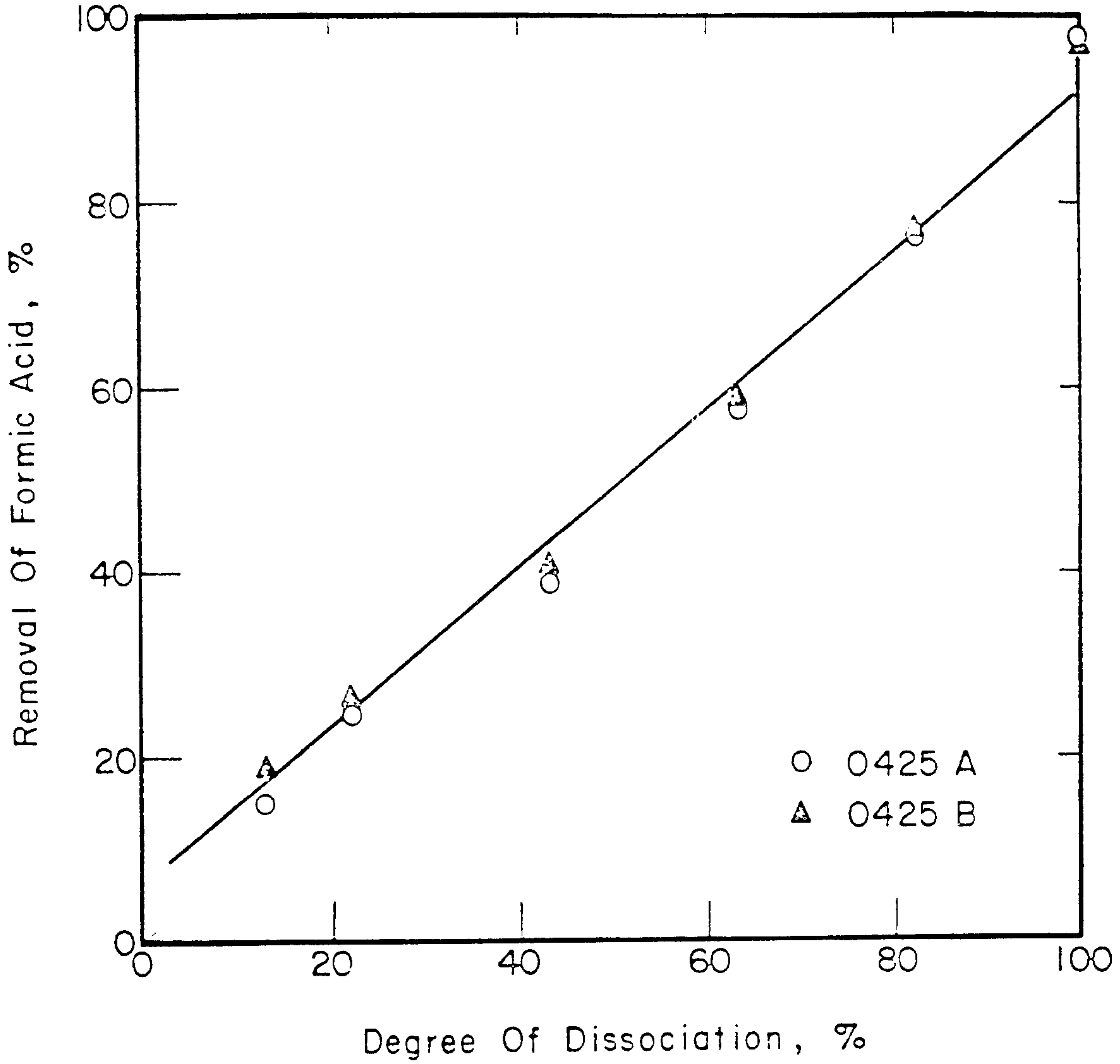


Figure 17. Effect of degree of dissociation on percentage removal (10)

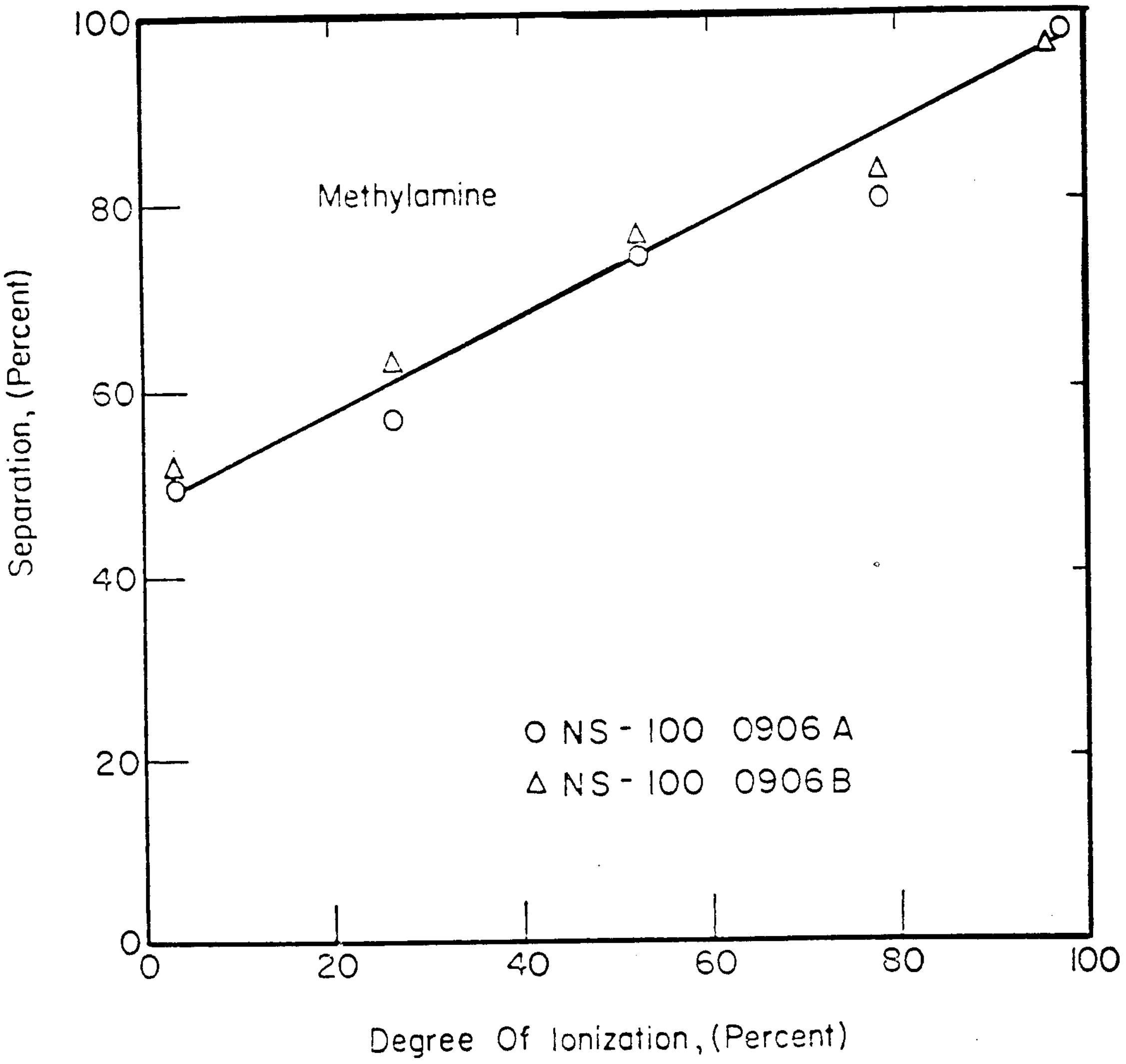


Figure 18. Effect of degree of ionization on percentage removal (10)

true for organic ions and water. Hence, as the ions are formed from the parent molecule when the pH is adjusted, hydration reactions immediately ensue, surrounding the organic ion with waters of hydration. The resulting bulkiness, then, increases separation.

Adsorbate-adsorbent Partitioning and Transport across Membrane

At the beginning of this chapter we stated that if the solute molecules were not to reach the membrane skin, they would all be removed. They do, however, reach the skin. To determine how membrane and solution interact, Anderson and coworkers performed a partitioning experiment.[13] They used cellulose acetate (CA) and cellulose acetate butyrate (CAB) membranes. Weighed amounts of the dried CA and CAB membranes were added to aqueous solutions of the various solutes they were experimenting upon. Changes in the concentration of the solutions were then determined. In addition, they also performed diffusion studies and RO experiments. Diffusion coefficients D were obtained by following the sorption or desorption of solute from plane sheets of membrane having a known thickness that ranged from 20 to 50 microns. The solutes saturated into the membrane were generally eluted into a known volume of distilled water. Their results along with the corresponding RO experimental results are shown in Table 2. As shown, the applied pressure is 68 atmospheres (1000 psig). The percentage removals are discouragingly low and do not correlate with the diffusion coefficient D..

Table 2 also shows ranges of values for the partition coefficient K.[13] Higher values mean the solute has more affinity for the membrane than for water. High values therefore mean that more solute is available for crossing the membrane, which would predict a low percentage removal. Although the indicated removals are generally low, they are not, at first glance, consistent with the increase or decrease of the value of K. There is, however, a weak negative correlation between K and R, the percent rejection. Looking back at Table 2, solutes with high and low values of K all have low removals. A general conclusion suggested by these results is that once the solute is incorporated into the membrane, its passage through to the permeate side is relatively unobstructed, causing the very low removals.

The results of Anderson et al.[13] may be compared with those of Chian et al.,[15] who studied the removal of pesticides using RO (Table 3). As shown, although the percentages of solute adsorbed onto the membrane are very high, signifying more solute particles available for migration, the percentage removals are, indeed, also very high. This is in apparent contradiction with results of Anderson and coworkers; Table 2 indicates that as long as the solutes are already in the membrane, they pass right through, causing low removals.

Table 2. Diffusion Coefficients, Partition Coefficients and RO Rejection of Organic Solutes by CA and CAB Membranes[13]

Solute	Mol. Wt.	Membrane	D x 10^{10} cm^2/sec[a]	K[b]	% Rejection (68 atm)
Phenol	94	CA	10	37 ± 1	- 10
		CAB	1.5	51 ± 1	
2,4-dichloro-phenol	163	CA	1.5	332 ± 7	- 33
		CAB	0.7	405 ± 5	
p-bromophenol	173	CA	3.8	165 ± 2	
		CAB	0.84	175 ± 2	
Acetone	58	CA	300	0.3 ± 0.1	+ 17
		CAB	100	0.49 ± 0.05	
Hydroquinone	110	CA	-	3.5 ± 0.2	
		CAB	-	5.4 ± 1.5	
Nitromethane	61	CA	150	2.1 ± 0.5	- 6
		CAB	100	4.0 ± 0.6	
Nitrobenzene	123	CA	8.0	54 ± 4	
		CAB	3.5	105 ± 5	
Pyridine	79	CA	75	0.7	
		CAB	50	1.33 ± 0.03	
Urea	60	CA	130	0.49 ± 0.03	+ 45
Aniline	93	CA	20	20 ± 2	+ 4
		CAB	3.5	52 ± 3	
3,5-dicarbethoxy-phenol	238	CA	1.8	129 ± 12	
		CAB	-	110 ± 10	

a. D = diffusion coefficient.

b. K = partition coefficient.

Table 3. Removal of Chlorinated Pesticides by Reverse Osmosis[15]

Solute	Mol. Wt.	Membrane	Percentage Adsorbed	Percentage Removed
Aldrin	365	NS-100	95.15	100
		CA	79.55	100
Lindane	291	NS-100	12.03	98.95
		CA	68.40	99.51
Heptachlor	373	NS-100	96.28	100
		CA	80.63	100
Heptachlor epoxide	389	NS-100	91.50	99.84
		CA	76.47	99.77
DDE	318	NS-100	94.06	100
		CA	80.29	100
DDT	354.5	NS-100	94.29	100
		CA	100	100
Dieldrin	381	NS-100	95.36	100
		CA	76.31	99.88

The second set of solutes are larger than those in the first set studied by Anderson and coworkers. In addition, these pesticides are highly substituted. For example, heptachlor epoxide has the following chemical structure: 1,4,5,6,7,8,8a-heptachloro-a,2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindene. As can be observed, there are seven chlorine groups substituted into the molecule. This would give the molecule a bigger steric configuration. Therefore, although the pesticides had been sorbed into the membrane and, thus, available for passage to the permeate side, the bigger spread of the molecule had been a hindrance. The percentage removals are much higher, indeed, as compared to the removals reported by Anderson and coworkers in part because of the difference in steric configuration. It is to be noted that although phenol in Table 2 is reported to have a negative removal, it had been rejected by 90% when the pH was adjusted to 12.5, as shown in Figure 15.[5]

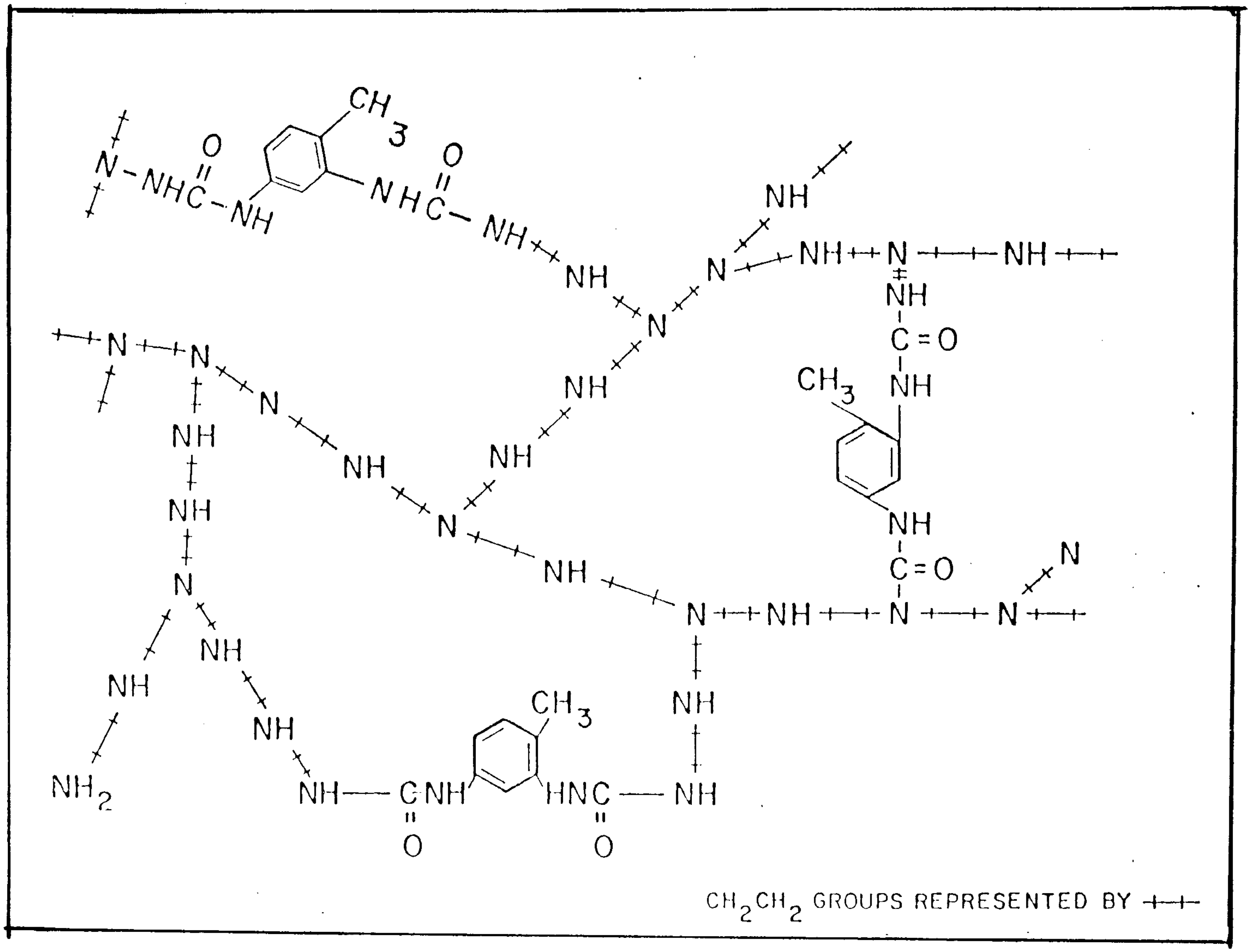
Table 3 shows that the NS-100 membrane consistently sorbed more pesticide compared with CA except for lindane and DDT. Chian et al. attributed these observations to the relative apolarity of NS-100 compared with CA.[15] Figure 19 shows the idealized structure of NS-100. NS-100 is a polyethylenimine (PEI) membrane cross-linked with toluene-2,4-diisocyanate. As shown, the ethylene backbone and the cross-linked benzene groups are the nonpolar regions while the peptide bonds and amines are the polar regions. CA, on the hand, is more polar because of the excess of OH^- compared with the acetyl group in its structure. Acetyl is the nonpolar while the hydroxyl is the polar part of the CA membrane. According to Chian et al., most chlorinated pesticides are nonpolar, and since nonpolar molecules attract each other, nonpolar pesticides are more strongly sorbed by NS-100, as shown in Table 3. Since lindane and DDT are more sorbed by CA, Chian et al. concluded that they are more polar. Again, however, despite their abundance on the membrane, they were unable to break through to the permeate side.

Going back to Table 2, when the value of the partition coefficient, K, is high, the value of the diffusion coefficient, D, is low, i.e., the relationship is inverse. Intuitively, passage of the solute across the membrane should depend on the diffusion and on the amount of solute in the membrane (partition coefficient). Also, intuitively, the large K values should correspond to large diffusion coefficients. However, Anderson et al. suggest that free and bound solutes interact as in other chemical equilibria; for high partition coefficients, the equilibrium favors the production of more bound solutes, hence, less of the free.[13] Armed with this concept, it is now easy to account for the high K values yielding low diffusion coefficients (because of the bonding of the solute to the membrane) and, conversely, for the lower K.

This concept of bound and free solute may have a bearing on the conclusion reached by Schutte and Belfort when studying the rejection of phenol using CA and an aromatic polyamide membrane.[12] They concluded that phenol was sorbed into both membranes but that the force of attraction between phenol and CA was weaker compared to the attraction between phenol and polyamide, resulting in a faster rate of diffusion in CA across the membrane and hence poor rejection. The force of attraction, which can impede diffusion, may be due to the binding of the solute by the membrane. We may rationalize that the binding is due to hydrogen bonding or to the outright polarity or nonpolarity of the solute.

Anderson et al. discussed the concept of the KD product.[13] They wrote that a solute will not be removed if the KD product is comparable to or greater than 2×10^{-8}. The K and D of NaCl are 0.038 and 3.2×10^{-9} cm^2/sec, respectively (1). The KD for NaCl is therefore 1.22×10^{-10}, much smaller than 2×10^{-8}. We know that sodium chloride is readily removed by the reverse osmosis process. If the KD products in Table 2 are computed, they will be shown to be comparable to 2×10^{-8}; hence, the solutes are poorly rejected, as observed. KD is, therefore, a measure of solute transport across membranes.

Lonsdale et al. discovered that transport of phenol in a cellulose acetate membrane is not purely by diffusion.[16,17] In their experiment, a piece of CA was supported between two coarse Monel grids in a circular hole through a central dividing partition in a rectangular plastic box. Phenol



CH_2CH_2 GROUPS REPRESENTED BY —+—+—

Figure 19. Idealized structure of NS-100 membrane (14)

(100 ppm) was placed in one chamber and water was placed in the other. Using various concentrations of sucrose, they manipulated the direction of flow of water such that phenol and water could cross the membrane in the same direction or opposite direction. The value of the KD product for phenol was 2.68×10^{-8} when in opposite direction and 3.67×10^{-8} when in the same direction. Insofar as the KD product is a measure of transport across the membrane, the transport of phenol is larger when the direction is the same as water and smaller when the direction is opposite. It is therefore clear that the transport across the membrane is not pure diffusion in the case of phenol but that there is coupling of the transport of phenol with water. The coupling may be due in part to the relatively small size of the phenol molecule, hence, it can easily "ride" with water. However, when the molecule is large, the principal process may still be diffusion across the membrane interposed with the alternate closing and opening of the chain gap due to thermal energy.

Removal of Mixtures of Solutes

The theories discussed have been developed from data using single solutes. For mixtures, there is the possibility of interactions among solutes. For example, if one solute is a base and the other an acid, they will neutralize each other. Because of the possibility of interactions, any mathematical formula developed for a single solute would not guarantee applicability to mixtures. For example, Ohya and Sourirajan developed rather complicated design equations for reverse osmosis process using parameters based on a single solute.[18] For single solutes, their equations would probably have good applicability. Lonsdale et al. also developed diffusion models based on a single solute.[17] In water treatment processes, the solutes involved are numerous; in fact, they may be unknown. Hindin et al. have shown that methyl formate experienced a drastic change in percentage removal when mixed with ethyl formate, methyl propionate and ethyl propionate.[19] When in a single solute the removal was 14 percent but, when in a mixture, the removal increased to 66.6 percent. It is therefore envisaged that design of reverse osmosis processes is done better by obtaining criteria utilizing laboratory or pilot plant testing of the water to be treated.

ORGANIC COMPOUNDS STUDIED

Although the following tables do not give an exhaustive listing of all the organic compounds studied, they should give a fairly accurate indication of how much they are removed. The tables are composed of three types of listing: Table 4 for removal of organic compounds when they exist as a single solute, Table 5 for their removal when they exist in solute mixtures, and Table 6 for their removal when they are in mixtures with sewage. The abbreviations used in the tables are explained in the Glossary of Terms (Appendix).

Table 4. Organic Compounds Removed as a Single Solute

Solute	Mem	MW	Conc mg/L	Flux	pH atm	P %	R	Ref
Acetaldehyde	CA	44.1	1,000	68 ccd	-	17	60	20
	PEI	44.1	1,000	47 ccd	-	54	75	20
	PEI	44.4	1,000	24 ccd	-	54	81	20
Acetic acid	CA	60.1	100	68 ccd	-	17	24	20
	CA	60.1	1,000	35 ccd	3.97	41	10	20
	CA	60.1	1,000	21 ccd	3.97	41	18	20
	CA	60.1	1,000	88 ccd	3.97	102	18	20
	CA	60.1	1,000	53 ccd	3.97	102	30	20
	PEI	60.1	1,000	39 ccd	3.77	41	69	20
	PEI	60.1	1,000	46 ccd	3.77	41	48	20
	PEI	60.1	1,000	47 ccd	-	54	71	20
	PEI	60.1	1,000	24 ccd	-	54	80	20
	PEI	60.1	1,000	114 ccd	3.77	102	70	20
	CAB	60.1	1,000	7 ccd	3.77	41	12	20
	CAB	60.1	1,000	20 ccd	3.77	102	26	20
	PA	60.1	682	-	-	27	31	20
	PA	60.1	500	-	-	-	40	20
	PA	60.1	1,000	15 ccd	3.77	41	70	20
	PA	60.1	1,000	38 ccd	3.77	102	82	20
Acetone	CA	58.1	100	68 ccd	-	17	22	20
	CA	58.1	100	-	-	102	17	13
	CA	58.1	1[illegible]	-	-	100	47	20
	CA	58.1	1,000	35 ccd	5.48	41	5	20
	CA	58.1	1,000	21 ccd	5.48	41	23	20
	CA	58.1	1,000	88 ccd	5.48	102	6	20
	CA	58.1	1,000	53 ccd	5.48	102	30	20
	PEI	58.1	1,000	39 ccd	5.48	41	80	20
	PEI	58.1	1,000	46 ccd	5.48	41	76	20
	PEI	58.1	1,000	47 ccd	-	54	93	20
	PEI	58.1	1,000	24 ccd	-	54	96	20
	PEI	58.1	1,000	97 ccd	5.48	102	78	20
	PEI	58.1	1,000	114 ccd	5.48	102	77	20
	CAB	58.1	1,000	7 ccd	5.48	41	17	20
	CAB	58.1	1,000	20 ccd	5.48	102	6	20
	PA	58.1	856	-	-	27	53	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH atm	P %	R	Ref
Acetone	PA	58.1	1,000	15 ccd	5.48	41	72	20
	PA	58.1	1,000	38 ccd	5.48	102	72	20
Acetophenone	CA	120.1	100	68 ccd	-	17	17	20
	PEI	120.1	500	47 ccd	-	54	97	20
	PEI	120.1	500	24 ccd	-	54	98	20
Adipic acid	CA	146.1	100	68 ccd	-	17	41	20
Adonitol	CA	152.2	100	68 ccd	-	17	97	20
Aldrin	PEI	365.0		-	-	-	100	15
	CA	142.3	-	-	-	-	100	15
m-aminobenzoic acid	CA	137.1	137	-	3.9	40	57	21
	CA	137.1	137	-	3.1	40	70	21
	CA	137.1	137	-	2.4	40	88	21
	CA	137.1	137	-	2.0	40	90	21
	CA	137.1	137	-	1.8	40	96	21
p-aminobenzoic acid	CA	137.1	137	68 ccd	-	17	13	20
	CA	137.1	137	-	4.2	40	2	21
	CA	137.1	137	-	3.2	40	12	21
	CA	137.1	137	-	2.3	40	59	21
	CA	137.1	137	-	1.7	40	75	21
p-aminophenol	CA	109.1	100	68 ccd	-	17	27	20
n-amyl acetate	CA	130.2	100	68 ccd	-	17	50	20
n-amyl alcohol	CA	88.2	100	68 ccd	-	17	23	20
Anisic acid	CA	152.1	100	68 ccd	-	17	20	20
Aniline	CA	93.1	100	68 ccd	-	17	4	20
	CA	93.1	1,000	35 ccd	6.62	41	-9	20
	CA	93.1	1,000	21 ccd	6.62	41	22	20
	CA	93.1	1,000	88 ccd	6.62	102	-5	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH atm	P %	R	Ref
Aniline	CA	93.1	1,000	53 ccd	6.62	102	17	20
	CA	93.1	-	-	6.4	34	88	13
	CA	93.1	-	-	7.3	34	9	13
	CA	93.1	-	-	6.4	68	78	13
	CA	93.1	-	-	7.5	68	4	13
	PEI	93.1	1,000	39 ccd	6 62	41	31	20
	PEI	93.1	1,000	46 ccd	6.62	41	52	20
	PEI	93.1	1,000	47 ccd	-	54	92	20
	PEI	93.1	1,000	27 ccd	-	54	96	20
	PEI	93.1	1,000	97 ccd	6.62	102	15	20
	PEI	93.1	1,000	114 ccd	6.62	102	31	20
	CAB	93.1	1,000	7 ccd	6.62	41	26	20
	CAB	93.1	1,000	20 ccd	6.62	102	-5	20
	PA	93.1	440	-	-	27	47	20
	PA	93.1	1,000	15 ccd	6.62	41	78	20
	PA	93.1	1,000	38 ccd	6.62	102	82	20
o-anisidine	CA	123.2	100	68 ccd	-	17	8	20
p-anisidine	CA	123.2	100	68 ccd	-	17	4	20
Anisole (ether)	CA	108.1	100	68 ccd	-	17	51	20
Arabitol	CA	152.2	100	68 ccd	-	17	97	20
Atrazine	PEI	216.0	-	-	-	-	98	15
	CA	216.0	-	-	-	-	84	15
Azelaic acid	CA	188.2	100	68 ccd	-	17	15	20
Beef extract	CA	-	255	39 gfd	-	20	97	22
Benzaldehyde	CA	106.1	100	68 ccd	-	17	10	20
	PEI	106.1	1,000	47 ccd	-	54	88	20
	PEI	106.1	1,000	24 ccd	-	54	95	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH atm	P %	R	Ref
Benzene	CA	78.1	100	68 ccd	-	17	76	20
	CA	78.1	100	13 ccd	-	3.4	97	20
	CA	78.1	100	27 ccd	-	6.8	89	20
	CA	78.1	100	134 ccd	-	34	54	20
Benzene hexachloride	CA	-	638 ug	14 gfd	-	100	52	19
Benzoic acid	CA	122.1	100	68 ccd	-	17	19	20
	PEI	122.1	500	47 ccd	-	54	66	20
	PEI	122.1	500	24 ccd	-	54	82	20
Benzyl alcohol	CA	108.1	100	68 ccd	-	17	5	20
Benzyl methyl ketone	CA	134.2	100	68 ccd	-	17	17	20
Boric acid	CA	43.8	-	-	7.0	34	43	13
	CA	43.8	-	-	7.0	68	67	13
	CA	43.8	-	-	11.0	34	97	13
	CA	43.8	-	-	11.0	68	97	13
1,3-butanediol	CA	-	100	68 ccd	-	17	72	20
2,3-butanediol	CA	90.1	100	68 ccd	-	17	80	20
n-butyl alcohol	CA	74.1	100	68 ccd	-	17	16	20
	PEI	74.1	1,000	47 ccd	-	54	94	20
	PEI	74.1	1,000	24 ccd	-	54	96	20
i-butyl alcohol	CA	74.1	100	68 ccd	-	17	41	20
s-butyl alcohol	CA	74.1	100	68 ccd	-	17	34	20
t-butyl alcohol	CA	74.1	100	68 ccd	-	17	79	20
i-butyraldehyde	CA	72.1	100	68 ccd	-	17	78	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH atm	P %	R	Ref
n-butyraldehyde	CA	72.1	1,000	68 ccd	-	17	60	20
t-butylbenzene	CA	134.1	100	68 ccd	-	17	99	20
i-butyric acid	CA	88.1	100	68 ccd	-	17	35	20
n-butyric acid	CA	88.1	100	68 ccd	-	17	25	20
Caprylic acid	CA	144.2	100	68 ccd	-	17	19	20
Captan	PEI	301.0	-	-	-	-	100	15
	CA	301.0	-	-	-	-	98	15
Casein	CA	-	1,000	17 gfd	7.8	100	99	19
Cellulose	CA	-	47	25 gfd	-	100	69	19
o-chloroaniline	CA	127.6	100	68 ccd	-	17	19	20
m-chloroaniline	CA	127.6	100	68 ccd	-	17	21	20
o-chlorobenzoic acid	CA	156.6	100	68 ccd	-	17	45	20
p-chloronitrobenzene	CA	-	0.098	9 gfd	5.3	100	27	19
Chlorophenol	CA	128.6	100	68 ccd	-	17	22	20
m-chlorophenol	CA	128.6	100	68 ccd	-	17	20	20
p-chlorophenol	CA	128.6	100	68 ccd	-	17	21	20
	CA	128.6	128	68 ccd	-	17	42	20
	CA	128.6	128	162 ccd	-	41	20	20
	CA	128.6	128	270 ccd	-	68	18	20
	CA	128.6	128	405 ccd	-	102	1	20
	PEI	128.6	1,000	47 ccd	-	54	81	20
	PEI	128.6	1,000	15 ccd	-	17	66	20
	PEI	128.6	1,000	35 ccd	-	41	79	20
	PEI	128.6	1,000	59 ccd	-	68	80	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
p-chlorophenol	PEI	128.6	1,000	88 ccd	-	102	80	20
	PEI	126.1	1,000	24 ccd	-	54	83	20
m-cresol	CA	108.1	100	68 ccd	-	17	2	20
p-cresol	CA	108.1	100	68 ccd	-	17	2	20
Crotonaldehyde	CA	70.1	100	68 ccd	-	17	15	20
Cumene	CA	120.2	100	68 ccd	-	17	80	20
	CA	120.2	100	27 ccd	-	6.8	98	20
	CA	120.2	100	134 ccd	-	34	71	20
	CA	120.2	100	268 ccd	-	68	53	20
	CA	120.2	100	405 ccd	-	102	35	20
Cycloheptatriene	CA	92.1	100	68 ccd	-	17	66	20
Cyclohexane	CA	84.2	100	68 ccd	-	17	90	20
Cyclohexene	CA	82.1	100	68 ccd	-	17	65	20
cis-and-trans-1,2-cyclohexanediol	CA	116.2	100	405 ccd	-	102	83	20
trans-1,2-cyclohexanediol	CA	116.2	100	405 ccd	-	102	86	20
Cyclohexanol	CA	100.2	100	68 ccd	-	17	60	20
Cyclohexanone	CA	98.1	100	68 ccd	-	17	39	20
	PEI	98.1	500	47 ccd	-	54	97	20
	PEI	98.1	500	24 ccd	-	54	99	20
Cyclopentane	CA	70.1	100	68 ccd	-	17	70	20
Cyclopentanone	CA	84.1	100	68 ccd	-	17	26	20
Cyclopentene	CA	68.1	100	68 ccd	-	17	60	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
DDD (Technical TDE)	CA	-	0.532	14 gfd	-	100	199	19
DDE	PEI	318.0	-	-	-	-	100	15
	CA	318.0	-	-	-	-	100	15
DDT	CA	354.5	0.007	17 gfd -	100	100	19	
	PEI	354.5	-	-	-	-	100	15
	CA	354.5	-	-	-	-	100	15
Dextrose	CA	180.2	940	-	7.00	102	100	20
Diazinon	PEI	304.0	-	-	-	-	98	15
	CA	304.0	-	-	-	-	98	15
2,4-dichlorophenol	CA	163.0	35	-	-	120	-34	20
2,4-dichlorophenoxy-acetic acid	CA	221	35	-	-	102	93	20
Dieldrin	PEI	381	-	-	-	-	100	15
	CA	381	-	-	-	-	100	15
Diethyl ether	CA	74.1	100	68 ccd	-	17	57	20
	CA	74.1	1,000	35 ccd	5.59	41	12	20
	CA	74.1	1,000	21 ccd	5.59	41	30	20
	CA	74.1	1,000	88 ccd	5.59	102	10	20
	CA	74.1	1,000	53 ccd	5.59	102	24	20
Diisobutyl ketone	CA	142.2	100	68 ccd	-	17	59	20
Diisopropyl amine	CA	101	-	-	-	17	72	20
Diisopropyl ether	CA	102.2	100	68 ccd	-	17	84	20
Diisopropyl ketone	CA	114.2	100	68 ccd	-	17	67	20
Dimethylamine	CA	45.1	100	68 ccd	-	17	16	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
N,N-dimethylaniline	CA	121.2	100	68 ccd	-	17	33	20
N,N-dimethylbenzyl-amine	CA	135.2	100	68 ccd	-	17	56	23
Di-n-butylamine	CA	129.5	100	68 ccd	-	17	63	20
1,4-dioxane (ether)	CA	88.1	100	68 ccd	-	17	49	20
Dulcitol	CA	182.2	100	68 ccd	-	17	99	20
Epichlorohydrin (ether)	CA	92.5	100	68 ccd	-	17	18	20
1-erythritol	CA	122.1	100	68 ccd	-	17	94	20
Ethyl acetate	CA	88.1	100	68 ccd	-	17	45	20
	PEI	88.1	1,000	47 ccd	-	54	96	20
	PEI	88.1	1,000	24 ccd	-	54	97	20
Ethyl alcohol	CA	46.1	1,000	34 ccd	5.42	41	12	
20	CAB	46.1	1,000	7 ccd	5.42	41	2	20
	CA	46.1	100	68 ccd	-	17	10	20
	CA	46.1	1,000	21 ccd	-	41	24	20
	CA	46.1	1,000	35 ccd	5.42	41	13	20
	CA	46.1	1,000	49 ccd	5.42	102	37	20
	CA	46.1	1,000	88 ccd	-	102	18	20
	PEI	46.1	1,000	47 ccd	-	54	80	20
	PEI	46.1	1,000	24 ccd	-	54	87	20
	PEI	46.1	1,000	39 ccd	5.42	41	65	20
	PEI	46.1	1,000	46 ccd	5.42	41	66	20
	PEI	46.1	1,000	97 ccd	5.42	102	70	20
	PEI	46.1	1,000	114 ccd	5.42	102	74	20
	CAB	46.1	1,000	7 ccd	5.42	41	2	20
	CAB	46.1	1,000	20 ccd	5.42	102	14	20
	PA	46.1	677	-	-	27	36	20
	PA	46.1	2,000	-	-	-	28	20
	PA	46.1	1,000	15 ccd	5.42	41	57	20
	PA	46.1	1,000	38 ccd	5.42	102	70	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
Ethyl alcohol	CA	46.1	1,000	-	-	41	12	9
	PA	46.1	677	-	-	27	36	9
Ethylamine	PEI	45.1	1,000	47 ccd	-	54	86	20
	PEI	45.1	1,000	24 ccd	-	54	93	20
Ethylbenzene	CA	106.2	100	68 ccd	-	17	78	20
Ethyl ether	PEI	74.1	1,000	39 ccd	5.59	41	91	20
	PEI	74.1	1,000	46 ccd	5.59	41	82	20
	PEI	74.1	1,000	97 ccd	5.59	102	66	20
	PEI	74.1	1,000	114 ccd	5.59	102	77	20
	CAB	74.1	1,000	7 ccd	5.59	41	6	20
	CAB	74.1	1,000	20 ccd	5.59	102	2	20
	PA	74.1	388	-	-	27	58	20
	PA	74.1	1,000	15	5.59	41	90	20
	PA	74.1	1,000	38 ccd	5.59	102	92	20
Ethyl vinyl ether	CA	72.1	100	68 ccd	-	17	55	20
Formaldehyde	CA	30.0	1,000	21 ccd	4.64	41	33	20
	CA	30.0	1,000	35 ccd	4.64	41	20	20
	CA	30.0	1,000	53 ccd	4.64	102	48	20
	CA	30.0	1,000	88 ccd	4.64	102	30	20
	PEI	30.0	1,000	39 ccd	4.64	41	37	20
	PEI	30.0	1,000	46 ccd	4.64	41	39	20
	PEI	30.0	1,000	47 ccd	-	54	55	20
	PEI	30.0	1,000	24 ccd	-	54	70	20
	PEI	30.0	1,000	97 ccd	4.64	102	50	20
	PEI	30.0	1,000	114 ccd	4.64	102	51	20
	CAB	30.0	1,000	7 ccd	4.64	41	41	20
	CAB	30.0	1,000	20 ccd	4.64	102	50	20
	PA	30.0	1,278	-	-	27	21	20
	PA	30.0	1,000	15 ccd	4.64	41	52	20
	PA	30.0	1,000	38 ccd	4.64	102	67	20
Furfuryl aldehyde	CA	96.1	100	68 ccd	-	17	2	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
Glucose	CA	-	972	41 gfd	-	20	100	22
Glutamic acid	CA	-	564	40 gfd	-	20	100	22
Glycerol	CA	92.1	100	68 ccd	-	17	82	20
	CA	92.1	1,000	21 ccd	5.97	41	95	20
	CA	92.1	1,000	35 ccd	5.97	41	81	20
	CA	92.1	1,000	53 ccd	5.97	102	98	20
	CA	92.1	1,000	88 ccd	5.97	102	85	20
	PEI	92.1	1,000	39 ccd	5.97	41	97	20
	PEI	92.1	1,000	46 ccd	5.97	41	89	20
	PEI	92.1	1,000	97 ccd	5.97	102	96	20
	PEI	92.1	1,000	114 ccd	5.97	102	92	20
	CAB	92.1	1,000	7 ccd	5.97	41	90	20
	CAB	92.1	1,000	20 ccd	5.97	102	96	20
	PA	92.1	765	-	-	27	88	20
	PA	92.1	2,000	-	-	-	90	20
	PA	92.1	1,000	15 ccd	5.97	41	88	20
	PA	92.1	1,000	38 ccd	5.97	102	88	20
Heptachlor	PEI	373.0	-	-	-	-	100	15
	CA	373.0	-	-	-	-	100	15
Heptachlor oxide	PEI	389.0	-	-	-	-	100	15
	CA	389.0	-	-	-	-	100	15
1,6-heptadiene	CA	96.2	100	68 ccd	-	17	94	20
1,6-heptadiyne 20		CA	92.1	100	68 ccd	-	17	42
n-heptyl alcohol	CA	116.2	100	68 ccd	-	17	9	20
1-heptyne	CA	96.2	100	68 ccd	-	17	77	20
1,5-hexadiene	CA	82.1	100	68 ccd	-	17	59	20
n-hexane	CA	86.2	100	68 ccd	-	17	99	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
1,6-hexanediol	CA	118.2	100	405 ccd	-	102	45	20
1,2,6-hexanetriol	CA	134.2	100	68 ccd	-	17	85	20
1-hexene	CA	84.2	100	68 ccd	-	17	90	20
n-hexyl alcohol	CA	102.2	100	68 ccd	-	17	11	20
1-hexyne	CA	82.1	100	68 ccd	-	17	54	20
Humic acid	CA	-	-	12 gfd	9.9	100	99	19
	CA	-	-	11 gfd	6.5	100	99	19
Hydroquinone	CA	110.1	100	68 ccd	-	17	1	20
	CA	110.1	1,000	35 ccd	5.2	41	-0.6	20
	CA	110.1	1,000	21 ccd	5.2	41	7	20
	CA	110.1	1,000	88 ccd	5.2	102	-8	20
	CA	110.1	1,000	53 ccd	5.2	102	18	20
	CA	110.1	100	68 ccd	-	17	4	20
	PEI	110.1	1,000	39 ccd	5.2	41	82	20
	PEI	110.1	1,000	46 ccd	5.2	41	72	20
	PEI	110.1	1,000	97 ccd	5.2	102	86	20
	PEI	110.1	1,000	114 ccd	5.2	102	78	20
	CAB	110.1	1,000	7 ccd	5.2	41	31	20
	CAB	110.1	1,000	20 ccd	5.2	102	38	20
	PA	110.1	551	-	-	27	60	20
	PA	110.1	1,000	15 ccd	5.2	41	84	20
	PA	110.1	1,000	38 ccd	5.2	102	99	20
m-hydroxybenzoic acid	CA	138.1	100	68 ccd	-	17	19	20
Isopentane	CA	72.2	100	68 ccd	-	17	87	20
Isoprene	CA	68.1	100	68 ccd	-	17	52	20
Isopropanol	PEI	60.1	1,000	47 ccd	-	54	91	20
	PEI	60.1	1,000	24 ccd	-	54	92	20
	PEI	60.1	1,000	39 ccd	5.91	41	82	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
Isopropanol	PEI	60.1	1,000	46 ccd	5.91	41	85	20
	PEI	60.1	1,000	97 ccd	5.91	102	56	20
	PEI	60.1	1,000	114 ccd	5.91	102	40	20
	CAB	60.1	1,000	7 ccd	5.91	41	41	20
	CAB	60.1	1,000	20 ccd	5.91	102	58	20
	PA	60.1	1,174	-	-	27	90	20
	PA	60.1	2,000	-	-	-	75	20
	PA	60.1	1,000	15 ccd	5.91	41	89	20
	PA	60.1	1,000	38 ccd	5.91	102	96	20
	CA	60.1	1,000	-	-	41	43	9
	PA	60.1	1,174	-	-	27	90	9
2,4-D isopropyl ester	CA	-	97 ug	12 gfd	-	100	96	19
Lactic acid	PEI	90.1	1,000	47 ccd	-	54	88	20
	PEI	90.1	1,000	24 ccd	-	54	89	20
L-leucine	CA	131.2	1,312	-	3.6	40	96	21
	CA	131.2	1,312	-	4.5	40	98	21
	CA	131.2	1,312	-	5.8	40	99	21
Lindane	CA	290.8	500 ng	16 gfd	-	100	84	19
	PEI	290.8	-	-	-	-	99	15
	CA	290.8	-	-	-	-	100	15
Linear alkyl benzene sulfonate	CA	-	10	18 gfd	5	100	99	19
	CA	-	98	41 gfd	-	20	99	22
Malathion	PEI	330.0	-	-	-	-	100	15
	CA	330.0	-	-	-	-	99	15
Malonic acid	CA	104.1	100	68 ccd	-	17	55	20
Methanol	CA	32.0	100	68 ccd	-	17	5	20
	CA	32.0	1,000	21 ccd	5.77	41	-20	20
	CA	32.0	1,000	35 ccd	5.77	41	-8	20
	CA	32.0	1,000	53 ccd	5.77	102	-12	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
Methanol 20	PEI	32.0	1,000	47 ccd	-	54	33	
	PEI	32.0	1,000	24 ccd	-	54	41	20
	PEI	32.0	1,000	39 ccd	5.77	41	-6	20
	PEI	32.0	1,000	46 ccd	5.77	41	18	20
	PEI	32.0	1,000	97 ccd	5.77	102	1	20
	PEI	32.0	1,000	114 ccd	5.77	102	10	20
	CAB	32.0	1,000	7 ccd	5.77	41	-9	20
	CAB	32.0	1,000	18 ccd	5.77	102	1	20
	PA	32.0	536	-	-	27	28	20
	PA	32.0	2,000	-	-	-	0	20
	PA	32.0	1,000	15 ccd	5.77	41	19	20
	PA	32.0	1,000	38 ccd	5.77	102	5	20
p-methoxyphenol	CA	124.1	100	68 ccd	-	17	0	20
Methyl acetate	CA	74.1	100	68 ccd	-	17	40	20
	CA	74.1	1,000	35 ccd	5.34	41	-3.5	20
	CA	74.1	1,000	21 ccd	5.34	41	21	20
	CA	74.1	1,000	88 ccd	5.38	102	11	20
	CA	74.1	1,000	53 ccd	5.38	102	12	20
	PEI	74.1	1,000	39 ccd	5.38	41	36	20
	PEI	74.1	1,000	46 ccd	5.38	41	28	20
	PEI	74.1	1,000	47 ccd	-	54	89	20
	PEI	74.1	1,000	24 ccd	-	54	92	20
	PEI	74.1	1,000	97 ccd	5.38	102	23	20
	PEI	74.1	1,000	114 ccd	5.38	102	47	20
	CAB	74.1	1,000	7 ccd	5.38	41	-25	20
	CAB	74.1	1,000	20 ccd	5.38	102	-11	20
	PA	74.1	370	-	-	27	57	20
	PA	74.1	1,000	15 ccd	5.38	41	54	20
	PA	74.1	1,000	38 ccd	5.38	102	44	20
Methyl acrylate	CA	86.1	100	68 ccd	-	17	35	20
N-methylaniline	CA	107.2	100	68 ccd	-	17	12	20
Methyl benzoate	CA	136.1	100	68 ccd	-	17	25	20

Table 4. - Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
Methyl benzyl ether	CA	122.2	100	68 ccd	-	17	26	20
Methyl n-butyrate	CA	102.1	100	68 ccd	-	17	44	20
Methyl chloroacetate	CA	108.5	100	68 ccd	-	17	13	20
Methyl cyclopentane	CA	84.2	100	68 ccd	-	17	93	20
Methyl ethyl Ketone	CA	72.1	100	68 ccd	-	17	24	20
	CA	72.1	-	-	-	163	18	14
	PEI	72.1	1,000	47 ccd	-	54	91	20
	PEI	72.1	1,000	24 ccd	-	54	96	20
Methyl formate	CA	-	80	10 gfd	5.4	100	14	19
Methyl methacrylate	CA	100.1	100	68 ccd	-	17	40	20
Methylparathion	PEI	263.0	-	-	-	-	100	15
	CA	263.0	-	-	-	-	100	15
2-methyl-1-pentene	CA	84.2	100	68 ccd	-	17	92	20
4-methyl-1-pentene	CA	84.2	100	68 ccd	-	17	90	20
Motor oil, SAE-20	CA	-	10	19 gfd	-	100	100	19
m-nitroaniline	CA	138.1	100	68 ccd	-	17	8	20
m-nitrobenzoic acid	CA	167.1	100	68 ccd	-	17	37	20
o-nitrobenzoic acid	CA	167.1	100	68 ccd	-	17	76	20
p-nitrobenzoic acid	CA	167.1	100	68 ccd	-	17	35	20
Nitromethane	CA	61.0	-	-	7.2	34	-6	13
	CA	61.0	-	-	7.2	68	-9	13
m-nitrophenol	CA	139.1	100	68 ccd	-	17	2	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
p-nitrophenol	CA	139.1	-	-	3.0	20	-10	13
	CA	139.1	-	-	8.5	20	70	13
	CA	139.1	-	-	3.0	27	-13	13
	CA	139.1	-	-	8.5	27	78	13
	CA	139.1	-	-	3.0	41	-21	13
	CA	139.1	-	-	8.5	41	80	13
	CA	139.1	-	-	3.0	55	-28	13
	CA	139.1	-	-	8.5	55	81	13
	CA	139.1	-	-	3.0	68	-37	13
	CA	139.1	-	-	8.5	68	82	13
	CA	139.1	100	68	-	17	0	20
	CA	139.1	-	-	3.0	0	0	13
	CA	139.1	-	-	3.0	68	-40	13
	CA	139.1	-	-	11.0	68	84	13
	CA	139.1	-	-	3.0	20	-10	13
	CA	139.1	-	-	3.0	27	-14	13
	CA	139.1	-	-	3.0	41	-20	13
	CA	139.1	-	-	3.0	54	-27	13
n-octyl alcohol	CA	130.2	100	68 ccd	-	17	23	20
Oxalic acid	CA	90.0	100	68 ccd	-	17	94	20
Parathion	PEI	291.0	-	-	-	-	100	15
	CA	291.0	-	-	-	-	100	15
1,5-pentanediol	CA	104.2	100	405 ccd	-	102	54	20
n-pentanol	PEI	88.2	1,000	47 ccd	-	54	95	20
	PEI	88.2	1,000	24 ccd	-	54	98	20
3-pentanol	CA	88.2	100	68 ccd	-	17	40	20
	PEI	88.2	1,000	47 ccd	-	54	98	20
	PEI	99.2	1,000	24 ccd	-	54	99	20
Peptone	CA	-	1,000	17 gfd	4.0	100	99	19
Phenethyl alcohol	CA	122.2	100	68 ccd	-	17	8	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
Phenetole (ether)	CA	122.2	100	68 ccd	-	17	46	20
Phenol	CA	94.1	1,000	88 ccd	6.25	10	-18	20
	CA	94.1	1,000	21 ccd	6.25	41	-6	20
	CA	94.1	100	68 ccd	-	17	1	20
	PEI	94.1	1,000	86 ccd	6.25	41	68	20
	PEI	94.1	1,000	46 ccd	6.25	41	56	20
	PEI	94.1	1,000	47 ccd	6.25	54	84	20
	PEI	94.1	1,000	24 ccd	6.25	54	87	20
	PEI	94.1	1,000	97 ccd	6.25	102	70	20
	PEI	94.1	1,000	114 ccd	6.25	102	66	20
	CAB	94.1	1,000	7 ccd	6.25	41	11	20
	CAB	94.1	1,000	20 ccd	6.25	102	-12	20
	PA	94.1	773	-	-	27	45	20
	PA	94.1	2,000	-	-	-	55	20
	PA	94.1	1,000	15 ccd	6.25	41	80	20
	PA	94.1	1,000	38 ccd	6.25	102	89	20
Phenylacetic acid	CA	136.1	100	405 ccd	-	102	26	20
4-phenylbutyric acid	CA	164.2	100	405 ccd	-	102	22	20
m-phenylenediamine	CA	108.2	100	68 ccd	-	17	15	20
o-phenylenediamine	CA	108.2	100	68	-	17	14	20
p-phenylenediamine	CA	108.2	100	68	-	17	14	20
3-phenylpropionic acid	CA	150.2	100	405 ccd	-	102	18	20
Pimelic acid	CA	160.2	100	68 ccd	-	17	36	20
Pinacol	CA	118.2	100	68 ccd	-	17	92	20
Piperidine	CA	85.2	100	68 ccd	-	17	88	20
Pivalic acid	CA	102.1	100	48 ccd	-	17	66	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
Polyoxyethylene nonylphenyl ether	CA	661.4	52	106 ccd	-	40	84	20
	CA	925.5	100	-	-	-	60	24
	CA	1,410	238	-	-	-	92	24
1,3-propanediol	CA	76.1	100	405 ccd	-	102	48	20
n-propanol	CA	60.1	100	68 ccd	-	17	24	20
	PEI	60.1	1,000	47 ccd	-	54	93	20
	PEI	60.1	1,000	24 ccd	-	54	94	20
I-propanol	CA	60.1	100	68 ccd	-	17	37	20
Propionaldehyde	CA	58.1	1,000	68 ccd	-	17	75	20
Propionic acid	CA	74.1	100	68 ccd	-	17	24	20
Propylbenzene	CA	120.2	100	68 ccd	-	17	98	20
Propylene glycol	CA	76.1	100	68 ccd	-	17	68	20
Propylene oxide (ether)	CA	58.1	100	68 ccd	-	17	33	20
Pyrocatechol	CA	110.1	100	68 ccd	-	17	2	20
Randox	PEI	174.0	-	-	-	-	99	15
	CA	174.0	-	-	-	-	72	15
Resorcinol	CA	110.1	100	68 ccd	-	17	-1	20
Sodium dodecylbenzene-sulfonate (ABS)	CA	349.5	210	37 ccd	-	40	100	24
	CA	349.5	1	16 gfd	-	100	94	19
	CA	349.5	105	40 gfd	-	20	100	22
Sodium laurate	CA	-	25	10 gfd	-	100	100	19
Sodium oleate	CA	304.5	-	-	basic	100	100	20
	CA	304.5	38	7 gfd	-	100	100	19

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R	Ref
Sodium stearate	CA	-	40	10 gfd	-	100	100	19
	CA	-	961	38 gfd	-	20	100	22
D-sorbitol	CA	182.2	100	68 ccd	-	17	99	20
Starch (soluble)	CA	-	13	13 gfd	-	100	97	19
	CA	-	110	41 gfd	-	20	100	22
Succinic acid	CA	118.1	100	68 ccd	-	17	50	20
Sucrose	CA	-	2,110	40 gfd	-	20	100	22
Styrene	CA	104.1	100	68 ccd	-	17	70	20
Styrene oxide (ether)	CA	120.1	100	68 ccd	-	17	26	20
Suberic acid	CA	174.2	100	68 ccd	-	17	25	20
Tannic acid	CA	-	56	11 gfd	3.9	100	95	19
Tetradecylbenzyl-ammonium chloride	CA	704.6	188	139 ccd	-	40	94	24
Tetrahydropyran (ether)	CA	86.1	100	68 ccd	-	17	47	20
	CA	72.1	100	68 ccd	-	17	53	20
Toluene	CA	91.13	100	68 ccd	-	17	73	20
m-Toluic acid	CA	136.1	100	68 ccd	-	17	23	20
o-toluidine	CA	107.2	100	68 ccd	-	17	7	20
m-toluidine	CA	107.2	100	68 ccd	-	17	7	20
p-toluidine	CA	107.2	100	68 ccd	-	17	7	20
Trimethylamine	CA	59.1	100	68 ccd	-	17	77	20

Table 4. Organic Compounds Removed as a Single Solute, Continued

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R	Ref
Triethylamine	CA	101.2	100	68 ccd	-	17	95	20
	PEI	101.2	1,000	47 ccd	-	54	100	20
	PEI	101.2	1,000	24 ccd	-	54	99	20
Triethylene glycol	CA	150.0	-	-	-	100	37	20
Trifluralin	PEI	335.3	-	-	-	-	100	15
	CA	353.3	-	-	-	-	100	15
Urea	CA	60.0	-	-	-	100	45	20
	CA	60.0	1,000	35 ccd	7.72	40	18	20
	CA	60.0	1,000	21 ccd	7.72	40	38	20
	CA	60.0	1,000	88 ccd	7.72	102	27	20
	PEI	60.0	1,000	39 ccd	7.72	41	64	20
	PEI	60.0	1,000	46 ccd	7.72	41	55	20
	PEI	60.0	1,000	97 ccd	7.72	102	74	20
	PEI	60.0	1,000	114 ccd	7.72	102	63	20
	CAB	60.0	1,000	7 ccd	7.72	41	8	20
	CAB	60.0	1,000	20 ccd	7.72	102	31	20
	PA	60.0	1,188	-	-	27	34	20
	PA	60.0	1,000	15 ccd	7.72	41	55	20
	PA	60.0	1,000	38 ccd	7.72	102	89	20
Valeric acid	CA	102.1	100	68 ccd	-	17	19	20
o-xylene	CA	106.2	100	68 ccd	-	17	86	20
m-xylene	CA	106.2	100	68 ccd	-	17	84	20
p-xylene	CA	106.2	100	68 ccd	-	17	85	20
Xylitol	CA	152.2	100	68 ccd	-	17	97	20

Table 5. Removal of Organic Compounds in Solute Mixtures

Solute	Mem	MW	Conc mg/L	Flux	pH	P atm	R %	Ref
Phenol	CA	-	1031	9 gfd	9.0	100	18	19
Chlorophenol	-		1165				87	
Methyl formate	CA	-	98	10 gfd	4.2	100	67	19
Ethyl formate	-	-	87	-	-	-	68	
Methyl propionate	-	-	92	-	-	-	51	
Ethyl propionate	-	-	92	-	-	-	57	
L-glycine	CA	-	10	12 gfd	-	100	50	19
L-lysine.HCl	-	-	10	-	-	-	95	
dl-phenylalanine	-	-	10	-	-	-	95	
Sodium acetate	CA	-	104	15 gfd	-	100	77	19
Sodium propionate	-	-	99	-	-	-	74	
Sodium butyrate	-	-	95	-	-	-	73	
Sodium valerate		-	92	-	-	-	69	
Sodium isovalerate	-	-	93	-	-	-	71	
2,4-D isopropyl ester	CA	-	66	8 gfd -	100	54	19	
p-chloronitrobenzene	-	-	82	-	-	-	26	
Phenol	-	-	10	-	-	-	14	
Chlorophenol	-	-	10	-	-	-	37	
Acetone	-	-	101	-	-	-	33	
Ethyl acetate	-	-	100	-	-	-	60	
VX	CA	-	25	-	-	-	99	25
GB (Sarin)	-		30	-	-	-	78	
BZ (3-quinuclidinyl benzilate)	-		30	-	-	-	99	
BT	-		-	-	-	-	100	

Table 6. Removal of Organic Compounds in Mixtures with Sewage

Solute	Mem	MW	Conc mg/l	Flux	pH	P atm	R %	Ref
Solute Mixtures in Sewage Effluent								
In activated sludge:								
2,4-D isopropyl ester	CA	-	0.087	9 gfd	-	100	94	19
p-chloronitrobenzene	-	-	0.119	-	-	-	40	
Phenol	-	-	10	-	-	-	21	
Chlorophenol	-	-	10	-	-	-	50	
DDT	-	-	0.098	-	-	-	98	
Ethyl acetate	-	-	0.073	-	-	-	66	
Acetone	-	-	0.096	-	-	-	39	
TDS	CA	-	734	-	6.2	20	79	22
Conductivity, umho	-	-	1,090	-	-	-	82	
Ammonia as N	-	-	13	-	-	-	74	
Nitrates as N	-	-	1.2	-	-	-	53	
Orthophosphates as P	-	-	1.3	-	-	-	100	
COD	-	-	109	-	-	-	96	
TOC	-	-	27	-	-	-	96	
Permanganate oxygen	CA	-	-	-	-	-	88	26
BOD	PA	-	5.3	-	-	-	97	27
	PA		4.1	-	-	-	98	9

DISCUSSION, CONCLUSIONS, AND RECOMMENDATIONS

Referring to Table 4, it is not appropriate to contend that removal increases with the increase in molecular weight, which would seem logical. Acetaldehyde, e.g., with a molecular weight of 44.1, is removed by 60% using CA, while benzaldehyde, with a molecular weight of 106.1, more than twice that of acetaldehyde; is removed to the extent of only 10% using CA. Boric acid, with a molecular weight of only 43.8, is shown to be removed by 97% at pH 11 using CA, while benzoic acid, with a molecular weight of 122.1, about three times that of boric acid, is removed by only 19%. One may continue to analyze Table 4 and find similar comparisons as those of acetaldehyde and benzaldehyde and boric acid and benzoic acid. From these comparisons, one may conclude that increase in molecular size may not increase removal of a particular solute over another, even when the same functional group is involved. Of course, studies performed by different investigators may not give comparable results.

The effect of the change of membrane on removal is readily seen using PEI (polyethylenimine). For example, in the case of acetaldehyde, the removal increased from 60% using CA to 81% with PEI. For acetic acid, the increase is also very conspicuous, from 10% to 80%, also from CA to PEI. Benzaldehyde increased from 10% to 95%; benzoic acid, from 19% to 82%; and, boric acid, from 43% to 97%, and urea from 18% CA to 74% using PEI. Some noticeable increase is also shown by PA (polyamide). For example, urea increased its removal from 18% to 89%. Similar observations can be seen in the rest of the table. CAB (cellulose acetate butyrate) is similar to CA in removal of organics. For example, urea is shown removed to only 8%, which means it is worse than CA. However, CAB can be better than CA; for example, hydroquinone was removed by 38% using CAB as compared to 18% only or negative removals using CA. All these changes in removals with changes in membranes indicate that removal is dependent on the nature of the membrane.

Fang and Chian[7] studied the separation of polar organic compounds using the following types of membranes: cellulose acetate (CA), cellulose acetate butyrate CAB), cellulose triacetate, cross-linked polyethylenimine (PEI), the furfural alcohol membrane (NS-200)[5], poly-2,2'-(m-phenylene)-5,5'-bibenzimidazole (PBI), sulfonated polyphenylene oxide (SPPO), and the polyamide membranes. Their results showed PEI and NS-200 had the best removals. The removal ability of PEI can also be seen in Table 4, which demonstrates the superiority of PEI over the others.[5] The table shows excellent removals for the pesticides aldrin, lindane, heptachlor, heptachlor oxide, DDE, DDT, dieldrin, diazinon, methylparathion, malathion, parathion, randox, trifluralin, atrazine, and captan using either CA or PEI, although PEI shows a marked superiority over CA in removing randox and atrazine. Although we have seen that removals do not necessarily increase with molecular weight, big molecules are, however, without exceptions, always removed efficiently by any of the membranes in Table 4. For example, sodium laurate, sodium oleate, sodium stearate, starch, sodium dodecylbenzene sulfonate (ABS), motor oil, and linear alkyl benzene sulfonate (LAS) are removed almost 100%. However, Table 4 does not support the idea of a molecular cut-off point for efficient removal.

As far as reverse osmosis removal is concerned, we may categorize solutes into three: those that donate protons, those that accept protons, and those that simply stay neutral. For acidic and basic solutes, removal may be a very pronounced function of pH. For example, boric acid, molecular weight, 43.8, was removed to only 43 percent at pH 7.0; whereas, at pH 11.0, it was removed to 97 percent. For p-nitrophenol, molecular weight, 139.1, removal is -37 percent at pH 3 and 82 percent at pH 8.5. Hence, for the same molecular weight, we can have a very efficient or a very inefficient removal. Changes in pH, of course, involve interplay among the hydronium ion, the proton, and the hydroxyl ion. As the pH is raised, acidic organic species will tend to become dissociated, while basic species become increasingly neutral (except for a few strong organic bases, the quaternary ammonium hydroxides, that are completely dissociated at high pH); as the pH is lowered, on the other hand, organic bases will tend to become protonated, while acids become increasingly neutral (except for a few strong organic acids, such as trifluoroacetic acid, which are completely dissociated at low pH). Inasmuch as ionic species (inorganic salts in particular) are more effectively rejected than neutral molecules, we would expect that organic acids would be best removed from basic solution and organic bases best removed from acidic solution, and this is observed. For example, as discussed previously, rejection of the weak acid p-nitrophenol increases from negative values at pH 3 to 84 percent at pH 11, corresponding to dissociation of the phenolic hydroxyl group.

Amphoteric solutes, such as the amino acids, might be expected to have minimum RO rejection at their isoelectric point, at which they are electrically neutral. This may explain the variation in removal of p-aminobenzoic acid with pH change. This solute has a pK_1 of 2.38 (corresponding to dissociation of $-NH_3+$), a pK_2 of 4.89 (corresponding to dissociation of $-CO_2H$) and an isoelectric point of pH 3.6. As shown in Table 4, at pH values closely bracketing the isoelectric point, 4.2 and 3.2, the removals were only 2 and 12 percent, respectively. As the pH was decreased to 1.7, the removal accordingly increased to 75%. The ion then partitioned more toward the water and less toward the membrane, resulting in improved removal. (The ion could also have been hydrated, making its steric configuration larger, thus hindering the passage through the membrane.) Improved removal with increasing pH above the isoelectric point would also be anticipated. A similar explanation applies to the improvement in removal of m-aminobenzoic acid with decreasing pH from 57 to 96 percent.

It is to be noted that no amount of pH change is required when the molecule is already large. This fact is exemplified by humic acid where the removal stayed the same at 99% when the pH was changed from 6.5 to 9.9. Humic acid could be an important aid for the removal of small pesticide molecules. Pesticide molecules are known to sorb into humic acids, thus, with humic acids shown to be effectively removed, small pesticide molecules would also be effectively removed. In sewage treatment, degradable organic substances are converted to humic acids at advanced degrees of treatment; hence, any undegraded pesticide may simply sorb into these humics and be removed if further treatment by RO is considered.

We mentioned that we can classify solutes in RO process into three categories. The first two have been discussed. The third ones are those that simply stay neutral unaffected by change in pH, i.e., staying as molecules. Unfortunately, Table 4 does not contain results of an experiment of this nature. Intuitively, however, the removal would depend upon the affinity of the solute species toward the membrane. Again, if the solute does not interact with the membrane, it will be completely removed according to the theories presented. It is only when the solute partitions into the membrane will there be passage into the permeate side. For this, we do not need any proof; it is axiomatic.

The removal of ethanol as shown in Table 4 is a case of removal or nonremoval by affinity to the membrane. Since alcohols have very small dissociation constants, they stay as molecules. Hence ethanol in Table 4 at pH 5.42 is in molecular form. As shown, using CAB and CA, the removals average in the range of 20 percent while, when using PEI, the removals average around 75%. Ethyl alcohol must have partitioned more into the membrane and diffused faster into the permeate side producing low removals; whereas, in the case of PEI, it partitioned lesser and more into water, thus, resulting in decidedly higher removals. These data indicate that ethyl alcohol has more affinity towards CA and CAB than it has towards PEI. In the case of glycerol, however, affinity or no affinity is not the question. Despite its relatively low molecular weight, it is shown to be efficiently removed. This is due to its steric configuration (the three hydroxyls dangling to the side).

All that Table 4 is showing is removals of solutes when they are in single solute solutions. The situation may, however, be different when the solutes are mixed into one solution. In this situation, the possibility of interaction among solutes cannot be avoided. There may be antagonistic or synergistic effects. For example, Table 5 shows that phenol was removed by 18% when removed along with chlorophenol using CA. When removed alone, it was removed by only 1% or even by negative removals as shown in Table 4 using also CA membrane. A similar situation is shown in the case of chlorophenol. Its removal is improved when mixed with phenol. When alone in a single solute, chlorophenol was removed to only 22 percent (Table 4) but when mixed with phenol it was removed to 87 percent which is, of course, a very high improvement in removal. Methyl formate in solution with ethyl formate, methyl propionate and ethyl propionate was removed by 67 percent (Table 5) but only by 14 percent when alone in solution.

These results show that design formulations derived for single solutes will not apply for mixtures. Design of reverse osmosis processes is therefore done better using data derived from laboratory or pilot plant studies. This is similar to the case of the operation of a municipal water treatment plant coagulation basin. Optimum dosage for alum and optimum pH are achieved better not by a theoretical equation but by performing the standard jar test.

Table 6 shows the removals of various solutes when mixed with sewage. Phenol is now shown removed by 21 percent, which is very close to its removal in a binary solute mixture with chlorophenol. On the other hand, chlorophenol removal has now been reduced to 50 percent down from 87 percent when it was mixed with phenol; but, this is still, however, a much higher removal than when it existed as a single solute. DDT, a big molecule, is showing a consistently high removal; 98 percent when mixed with sewage and 100 percent when alone in a single solute solution. For practical engineering purposes, 98 percent and 100 percent are equal. Ethyl acetate shows an improvement in removal by CA membrane when mixed with sewage (66 percent) compared with the single solute (45 percent). All these different values of removals for a given solute when alone in a single solute solution and when in a mixture are testimonies that design parameters are obtained better conducting laboratory or pilot plant studies.

In general, the percent removals indicated in Tables 4-6 will not ensure meeting drinking water standards. For example, let us take the case of benzene, a solvent commonly used in the manufacture of industrial chemicals, pharmaceuticals, pesticides, paints and plastics.[28] The MCL (maximum contaminant level) for benzene based on the Federal Primary Drinking Water Standard is 0.005 mg/L (56). Table 4 suggests that benzene can be removed by approximately 90 percent. Assuming the feed to the RO contains 100 mg/L, the product will contain 0.1 x 100 = 10 mg/L of benzene, much greater than 0.005. (The value we assumed for the influent feed concentration, 100 mg/L, is much less than the solubility of benzene in water.).

Again, let us take another solute, phenol, and assume a standard to be met of 0.001 mg/L.[29] Phenol is a taste-producing substance. From Table 4, let us assume a removal of 90 percent. Assume an influent of 100 mg/L. This will then correspond to a permeate of 10 mg/l which is much greater than 0.001. Finally, let us take the case of BZ (3-quinuclidinyl benzilate), which is one of the chemical warfare agents. Assume a maximum permissible concentration in the permeate of 0.003-0.007 mg/L (13). From Table 4, BZ is shown to be removed by 99%. Assuming an influent of 30 mg/l, the permeate will contain 0.01 x 30 = 0.3 mg/L. This, again, is much higher than the required effluent standard.

Our calculations show that the use of reverse osmosis may not be practical for producing a potable water from a heavily contaminated source. (Of course, the results are based upon our assumed influent concentrations.) To use RO for removing organics to produce drinkable water quality, two things needs to be investigated further. First, a membrane better than PEI needs to be found. This review has encountered only PEI as the best membrane, but the search was only through the literature and was not very thorough. Suppliers were not contacted. It is possible that a membrane may now exist that can remove the organics very efficiently to a drinking water quality. If this is the case, a pilot study needs to be done on this membrane using selected organics. PEI membranes are of excellent materials and can withstand a pH range of 1 to 13.[5] Since removal of many organic chemicals in water is a very strong function of pH and since PEI has a wide pH range of applicability, the pH of the influent water can be varied at will to effect a good removal. However, this percentage removal may still be short of the required efficiency to meet drinking water standards. The trend, therefore, would have to be toward research on the unit operations of RO as indicated below.

It is axiomatic that transport through a membrane is a function of the amount of solute species enmeshed in the membrane. Assume that we have passed a sample of water through a membrane. The amount of solute on the permeate side is less than that on the feed side, say by 90 percent. If the solute on the permeate side becomes the solute on the feed side of a second membrane, the solute concentration at the surface of the feed side of this second membrane will be much less than that on the feed side of the first membrane. All independent variables constant, the rate of flow of water in the first and the second membranes will be approximately the same. However, because of the reduction in the amount of solute, the rate of diffusion of the solute in the second membrane will be smaller compared to the rate of diffusion in the first membrane. In addition, if the concentration of solute on the feed side of the second membrane is insufficient to create a concentration driving force, there may not even be a solute diffusion. The constant rate of water flow through the second membrane coupled with the decreased rate of diffusion will therefore have a diluting effect on the solute on the permeate side, resulting in greater percentage removal for the second membrane compared to the first. Indeed, if the concentration on the feed side of the second membrane has become insufficient to cause a concentration driving force, the removal will be 100 percent effective, assuming the rejection mechanisms discussed above. The same reasoning would apply for multiple membranes in series, i.e., the efficiency of removal is expected to increase from stage 1 to stage 2 to stage n. This scenario would form a basis for unit operations research of RO. Various combinations of units may be attempted to attain the desired removals.

To sum up, the functionality of solute removals may be simply stated as follows: Removal is a function of pH, removal is function of the size of the solute molecule, and removal is a function of solute-membrane affinity. Dependence on pH means that we have a method at our disposal to convert or dissociate the solutes into ions, if they are amenable to this treatment, and remove them efficiently. Dependence on size means that if the size of the solute molecule is small, its removal must depend on the availability of a membrane "tight" enough to prevent the diffusion of the molecule, other conditions remaining constant. Of course, if the size of the molecule is large, it would be efficiently removed. Solute-membrane affinity is a factor responsible for the inefficiency of the membrane.

Based upon review of the literature and the discussions above, the following conclusions regarding the state-of-the-art of removing organic solutes in water treatment may be drawn:

1. The present RO technology can not insure the production of drinking water quality permeate in all instances.

2. RO technology has the potential to produce a water safe from organic chemicals. To this end, a search for a better membrane should be undertaken.

3. A drinking water quality effluent may be obtained by improvement on the unit operations. This may be obtained by staging and by optimizing various combination of RO units. The polyethylenimine-based membrane has been found to be the best in this review. This membrane and any better membrane may be used on a study for the improvement of the unit operations of reverse osmosis.

Of necessity, this literature review has been brief. All aspects of the current state-of-the-art of RO technology for the removal of organics in water treatment have not been covered. The discussions above and the conclusions drawn from them may even be erroneous based on information from the literature not yet reviewed. As a result of this literature review, the following recommendations are made:

1. Undertake a search for a new and better membrane.

2. Perform a study on the improvement of the unit operations of reverse osmosis using PEI membranes and other better membranes that may be found. This may be done by optimizing power cost (objective function) and water permeate flux (objective function) on the various unit system arrangements with the sole constraint that an arrangement effect a 100 percent rejection of the solute.

3. Continue the literature review on removal of organic substances in water treatment which shall include the following:

    (a) Membrane fouling
    (b) Disposal of concentrates
    (c) Operation costs
    (d) Removal of bacteria, protozoa, and viruses

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APPENDIX

GLOSSARY OF TERMS

atm	atmospheres
BOD	biochemical oxygen demand
CA	cellulose acetate
CAB	cellulose acetate butyrate
ccd	cubic centimeter per square centimeter per day
COD	chemical oxygen demand
Conc	concentration
D	diffusion coefficient
gfd	gallons per square foot per day
K	partition coefficient
Mem	membrane
MW	molecular weight
P	applied pressure
PA	polyamide
PEI	polyethylenimine
psig	pounds per square inch (gauge)
R	removal or rejection
Ref	reference
RO	reverse osmosis
TDS	total dissolved solids
TOC	total organic carbon
UF	ultrafiltration

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