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Evaluation of Interactions between Polymers and Low Molecular Weight Compounds by GLC I: Methodology and Interaction Evaluation

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Abstract ☐ Sorption-desorption thermodynamics were evaluated for two polymers and 13 low molecular weight compounds. The polymers, polyethylene and polyvinyl chloride, are of major use in packaging. The sorbents were bacteriostatic agents of pharmaceutical utility and organic solvents employed in package fabrication. GLC procedures were used where the polymers constituted the stationary phases and the sorbents constituted the mobile phases of the columns. Auxiliary methods were adapted for obtaining the data necessary for the thermodynamic computations. The measurement of interaction affinities was based on Raoult's law activity coefficients. The GLC methodology was fast and convenient to perform and could be a valuable tool for applied and theoretical investigations in this area.

Keyphrases ☐ Polymer-low molecular weight compound interactions—sorption-desorption thermodynamic methodology, GLC ☐ Sorption-desorption thermodynamic methodology—polyethylene and polyvinyl chloride, 13 low molecular weight compounds, GLC ☐ Package-content interactions—polymers and low molecular weight compounds, GLC method for evaluating sorption-desorption thermodynamics ☐ Plastics—interactions of packaging materials with low molecular weight compounds evaluated by GLC, sorption-desorption thermodynamics ☐ GLC—evaluation of polymer-low molecular weight compound interactions, sorption-desorption thermodynamic methodology

The use of polymers in packaging may result in sorption of ingredients from the contents or desorption of compounds used in package fabrication. These processes may render a product unsuited for use. The phenomena result from interactions between the polymer and the low molecular weight compound and diffusion. The thermodynamics of the interaction determine the equilibrium amount of sorbent in the polymer, and diffusion affects the rate for attaining the equilibrium. The available information on such interactions is insufficient for optimum use of polymers in packaging; thus the objectives of this work were to evaluate the interaction thermodynamics and to use the quantities obtained for relative grading of affinities and toward a better understanding of the systems. Since conventional methods for these objectives are cumbersome and time consuming, GLC procedures were adapted by using the polymers as the stationary phases and the low molecular weight compounds as the mobile phases of the columns.

GLC BACKGROUND

The retention of a compound in a GLC column is a function of its saturation vapor pressure at the column temperature and the energy of interaction with the stationary phase. The specific retention volume, V_g^0, is characteristic for a given mobile–stationary phase system and is derived from experimental quantities through (1):

$$V_g^0 = \frac{(t_r - t_a)\dot{V}j}{W_1}\frac{273}{T} \qquad \text{(Eq. 1)}$$

where t_r is the retention time for the compound, t_a is the injection pulse, $\dot{V}$ is the corrected inert gas flow rate, j is the compressibility

Table I—Solute-Solvent Systems

Low Molecular Weight Compounds Employed as Solutes

a. Organic Solvents[a]	b. Bacteriostatic Agents
n-Butanol	Chlorobutanol anhydrous USP[b]
tert-Butanol	Benzyl alcohol NF[c]
Methyl isobutyl ketone	Phenol USP[d]
2-Nitropropane	Methylparaben USP[b]
Butyl acetate	Propylparaben USP[b]
Hexane	
Octane	
Toluene	

Polymers Employed as Solvents

Polyethylene[e]
Polyvinyl chloride[f]

[a] Matheson, Coleman and Bell, E. Rutherford, N. J. [b] Fisher Scientific Co., Fair Lawn, N. J. [c] Eastman Chemical Products, Inc., Kingsport, Tenn. [d] J. T. Baker Chemical Co., Phillipsburg, N. J. [e] Marlex EMN TR-885, Phillips Petroleum Co., Bartlesville, Okla. [f] Allied Chemical Co., Morristown, N. J.

Table II—Column Characteristics and Applications

Stationary Phase	Column Length, m.	Stationary Phase Content, g.	Solutes for Which Employed
Polyethylene	3.0	6.5825	Organic solvents, chlorobutanol, phenol, benzyl alcohol
Polyethylene	1.5	2.4445	Methylparaben, propylparaben
Polyvinyl chloride	5.0	9.9674	Organic sorbents
Polyvinyl chloride	2.5	3.0226	Chlorobutanol, phenol, benzyl alcohol
Polyvinyl chloride	1.2	0.9640–0.8480	Methylparaben, propylparaben

correction, W_1 is the weight of the stationary phase, and T is the absolute temperature. The specific retention volume is unaffected by operational variables, such as column length and amount of stationary phase, but depends exclusively on the thermodynamic variables of the system. Raoult's law activity coefficient, γ, is derived from the specific retention volume through (2):

$$\gamma = \frac{273R}{V_g^0 P_2^0 M_1} = \frac{17.06 \times 10^6}{V_g^0 P_2^0 M_1} \qquad \text{(Eq. 2)}$$

where P_2^0 is the vapor pressure of the pure compound at the column temperature, M_1 is the molecular weight of the stationary phase, and R is the gas constant. The activity coefficient reflects only the interaction energy between the compound and the stationary phase; values decreasing from unity result from interactions favorable to mixing and those increasing from unity reflect interactions unfavorable to mixing. The coefficients derived from GLC are identical to those obtained from solutions where the polymers are the solvents and the low molecular weight compounds are the solutes. Since the coefficients are concentration dependent, their values are constants at infinite dilution. Rigorously, Eq. 2 requires fugacities; however, such values are rarely available and vapor pressures have been used instead, resulting in the apparent activity coefficients, γ_p.

Experimental Background—The solute-solvent systems used are presented in Table I. The solvents were polyethylene and polyvinyl chloride. The solutes consisted of organic solvents used in package fabrication and bacteriostatic agents employed in pharmaceuticals. The systems represent varying polarities, functional groups, and structures.

For the computations from Eq. 2, the number average molecular weight of the polymers, $\bar{M}_n$, was used. The vapor pressures for most of the organic solvents were computed by means of the Antoine equation (3) using empirical constants from *Reference 4*. For several of the solutes, the vapor pressures were determined experimentally.

The densities at the experimental temperatures for the thermodynamic computations (5) were derived as reasonable approximations through (6):

$$\rho_T = \rho\{1 - (T - T_\rho)10^{-3}\} \qquad \text{(Eq. 3)}$$

where ρ_T is the density at the experimental temperature T, and ρ is the literature density at temperature T_ρ. The error incurred in the computed values by this approximation is negligible. The melt densities for polyethylene were derived from Terry and Yang (7) as recommended by the supplier. Melt densities for polyvinyl chloride, chlorobutanol, phenol, methylparaben, and propylparaben were determined experimentally.

The elevated temperatures at which the experiments were conducted posed the question of chemical stability of the solutes and the polymers. The group of organic solvents was considered stable from the general knowledge of their properties. The literature contains reports on GLC of benzyl alcohol and phenol (8) and of methylparaben and propylparaben (9) without degradation. Chlorobutanol is known to be stable at the lower temperatures used. For the higher

temperatures, the stability of the solutes was monitored from the chromatograms, since degradation products would exhibit markedly different retention characteristics, and also from plots of retention values against temperature where deviation from linearity would indicate changes in the compound. This self-controlling capability of GLC obviated the need for further identification of an eluting compound as the temperature was raised, once a retention value was associated with it at a lower temperature. Preliminary experiments revealed degradation only for chlorobutanol starting from 210°, so experiments with this compound were conducted up to 200°.

The stability of the polymers was monitored by means of column performance experiments at the initiation of the experiments and periodically thereafter, using compounds of varying polarities. The stability of polyethylene did not present difficulties; however, for polyvinyl chloride, 3% stabilizer was incorporated. Retention attributable to the stabilizer was not observed during preliminary experiments. Although the temperature regimens for the two polymers were different because of stability reasons, overlapping at least at one temperature was always assured. The temperature ranges for polyethylene and polyvinyl chloride were 140-230 and 110-210°, respectively. For the polyvinyl chloride-parabens systems, which required high temperatures, a new column was used at each experimental temperature and only single runs were performed since replicates would have incurred greater error due to degradation of the polymer.

The loading of the column with stationary phase has usually been high in work of this kind in order to minimize adsorption effects on the solid support. Since the support used in this work was a silanized grade, the amount of loading was determined by feasibility. Polyvinyl chloride did not lend itself to heavy loading, so a lower level was used compared to polyethylene and compensation was achieved by a longer column.

The least amount of solute usable was determined by preliminary experiments and was found to be below any concentration effect on the retention. The solutes were applied as appropriate mixtures to save time and column deterioration.

Instrumentation—The dual-column gas chromatograph[1], equipped with flame-ionization detectors, was used. The temperature was measured with a millivolt potentiometer[2] and a set of four copper-constantan thermocouples.

Materials—The polyethylene was a narrow molecular weight distribution, high density homopolymer used for container molding. Its characteristics were: density, 0.965; molecular weight, $\bar{M}_n$ = 15,000; and antioxidant content, 0.015% 2,6-di(*tert*-butyl)-4-methylphenol[3]. The polyvinyl chloride was a blow molding grade homopolymer of $\bar{M}_n$ = 40,000 and $\bar{M}_n/\bar{M}_w \sim 2$. The organic solvents were chromato or spectral quality. The bacteriostatic agents were USP or NF grade. The stabilizer for polyvinyl chloride was di(*n*-butyl)tin β-mercaptopropionate[4] used at 3% (w/w) of polyvinyl chloride. The solid support for the columns was diatomaceous earth (acid and base washed and silanized)[5], 70-80 mesh.

[1] Model 5750B, Hewlett-Packard Analytical Instruments, Pasadena, Calif.
[2] Model 8690, Leeds and Northrup Co., Philadelphia, Pa.
[3] Ionol, Shell Chemical Co.
[4] Advastub, Carlisle Chemical Works, Inc., New Brunswick, N. J.
[5] Anakron ABS, Analabs, Inc., North Haven, Conn.

Tale III—Experimentally Determined Vapor Pressures

Compound	Temperature	Vapor Pressure, mm. Hg Experimental	Vapor Pressure, mm. Hg Literature	$\dfrac{\text{Experimental}}{\text{Literature}} \times 100$	Reference
Decane	119.8°	153.0	150	102	4
	140.9°	296.8	300	99	—
	150.5°	398.7	400	99.5	—
	158.4°	524.6	500	105	—
tert-Butanol	60.9°	309.0	300	103	4
	86.9°	921.0	900	102	—
Benzyl alcohol	141.7°	101.8	100	102	12
	183.0°	399.5	400	100	—
Butyl acetate	110°	528.6	—	—	—
	120°	666.0	—	—	—
	125°	749.0	760	98.5	12
	130°	841.5	—	—	—
	140°	1108.0	—	—	—
Chlorobutanol	120°	136.4	—	—	13
	135°	—	246	—	—
	140°	295	—	—	—
	150°	595	—	—	—
Methylparaben	190°	19.8	—	—	—
	200°	26.9	—	—	—
	210°	36.4	—	—	—
	220°	49.3	—	—	—
Propylparaben	190°	6.21	—	—	—
	200°	8.97	—	—	—
	210°	12.53	—	—	—
	220°	16.53	—	—	—

Preparation of Chromatographic Columns and Column Characteristics—The weighed polymer was dissolved in a suitable solvent in a refluxing apparatus. For polyethylene, the solvent was toluene; for polyvinyl chloride, a mixture of equal parts of toluene and methyl isobutyl ketone was employed. The stabilizer for the latter polymer was also dissolved in the solvent. The weighed amount of solid support was stirred into the solution, and the solvent was removed until dryness in a rotating vacuum evaporator. When preparing the polyvinyl chloride column material, glass beads were added as a stirring aid. The coated solid support was dried at 70° in a vacuum oven to constant weight, and the 50–70-mesh fraction was used for column packing. The stationary phase content of the packing was determined by extraction of a sample in a soxhlet apparatus, followed by gravimetric procedures. A correction was made for the amount of stabilizer in a polyvinyl chloride packing material. The prepared material was packed in 0.63-cm. (0.25-in.) stainless steel tubing. The amount of packing in the column was determined by weighing. The column characteristics are shown in Table II.

Chromatographic Procedures—The chromatographic columns were attached to the gas chromatograph through 6.4-cm. (2.5-in.) extension tubes to prevent the column ends from heating above the experimental temperature, since the blocks were maintained at 250°. The temperature of the column was measured by means of four thermocouples attached at various positions along the length and was controlled within 0.5°. The column was equilibrated for at least 10 min. before charging the solutes. The carrier gas was helium. Its flow rate was measured through the detector block at the experimental temperatures with a 50-ml. soap bubble flowmeter, and corrected values accounting for the water vapor pressure were employed in the computations. The inlet pressure was measured with a mercury manometer, and the outlet pressure was assumed to be 760 mm. Hg. The retention time was measured from the recorder chart using the front profiles of the elution peaks as recommended (10) for better consistency.

The main portion of experiments were run in triplicates, and the mean V_g^0 was used for further computations. Only those with methylparaben and propylparaben on polyvinyl chloride were performed individually. Every series of experiments was bracketed by column performance experiments to verify the column stability. The data were considered valid if the column performance parameters were within the experimental error.

Auxiliary Experiments—Vapor pressure and density data for several substances were unavailable and were determined within the framework of this investigation.

Vapor Pressure Determination by Means of GC—The method of

Table IV—Experimentally Determined Densities (g./ml.) at Varying Temperatures

Temperature	Chlorobutanol	Phenol	Methylparaben	Propylparaben	Polyvinylchloride
95°	—	1.0400	—	—	—
100°	—	1.0310	—	—	—
110°	1.2960	1.0170	—	—	—
120°	1.2720	1.0050	1.1575	1.0610	1.1834
130°	1.2490	0.9860	—	1.0520	1.1730
135°	—	—	1.1420	—	—
140°	1.1750	—	—	1.0380	1.1655
145°	—	—	1.1310	—	—
150°	—	—	—	1.0250	1.1590
155°	—	—	1.1200	—	—
160°	—	—	—	—	1.1490
170°	—	—	—	—	1.1425

Frank (11) using GC was employed. An apparatus suited for the compounds in this work was developed and placed in a gas chromatograph[6] equipped with a thermal conductivity detector[7]. The validity of the procedures and the performance of the apparatus were determined with substances of known vapor pressures. The experimental pressures are shown in Table III and appear to be within 5% of those determined by other means.

Density Determinations—These were performed at several temperatures with a modified Russell (14) pycnometer immersed in an oil bath, since the substances are solids at ambient conditions. The densities were plotted against temperature and the required values were read from the linear plots. The experimentally determined densities are shown in Table IV.

RESULTS AND DISCUSSION

The specific retention volumes computed from Eq. 1 depend on the vapor pressures of the solutes at the respective temperatures and, consequently, are not a reliable measure of the solute–solvent interaction energies. They are shown in the *Appendix* tables since they may
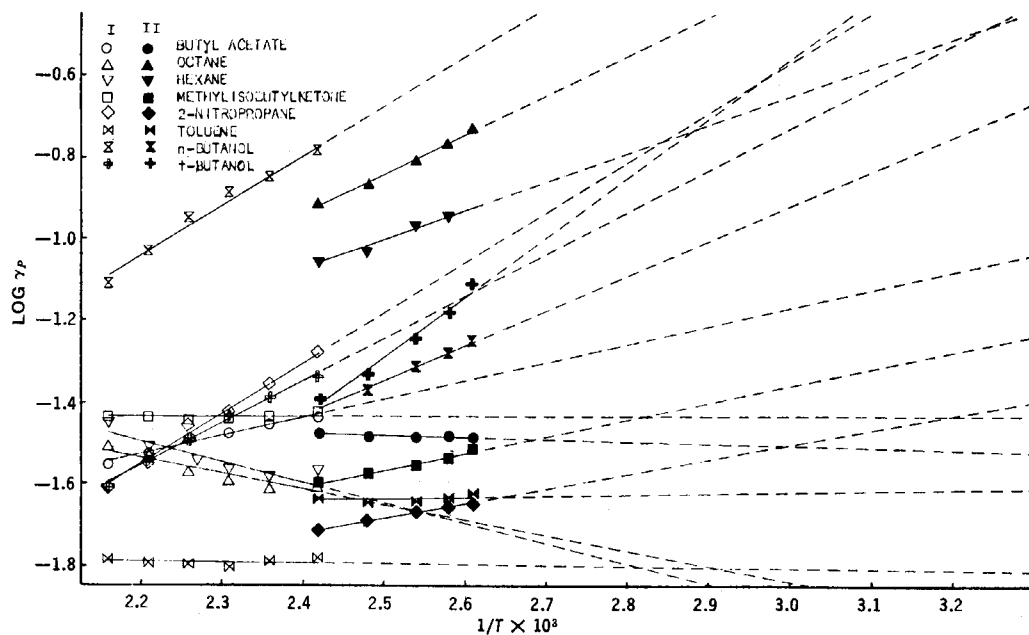
Figure 1—*Apparent activity coefficients at varying temperatures for the organic solvents. Key: I, polyethylene; and II, polyvinyl chloride.*

be of chromatographic utility. Their use in this work is in the computation of Raoult's law activity coefficients by means of Eq. 2. The coefficients depend exclusively on the interaction energies in the solute–solvent systems and were, therefore, used for relative grading of interaction affinities. The apparent activity coefficients are presented in Fig. 1 for the organic solvents and in Fig. 2 for the bacteriostatic agents. From the slopes of these plots, it may be seen that their temperature dependence varies considerably from solute to solute; therefore, the relative grading of the interaction affinities would depend on the temperature at which they are compared. To
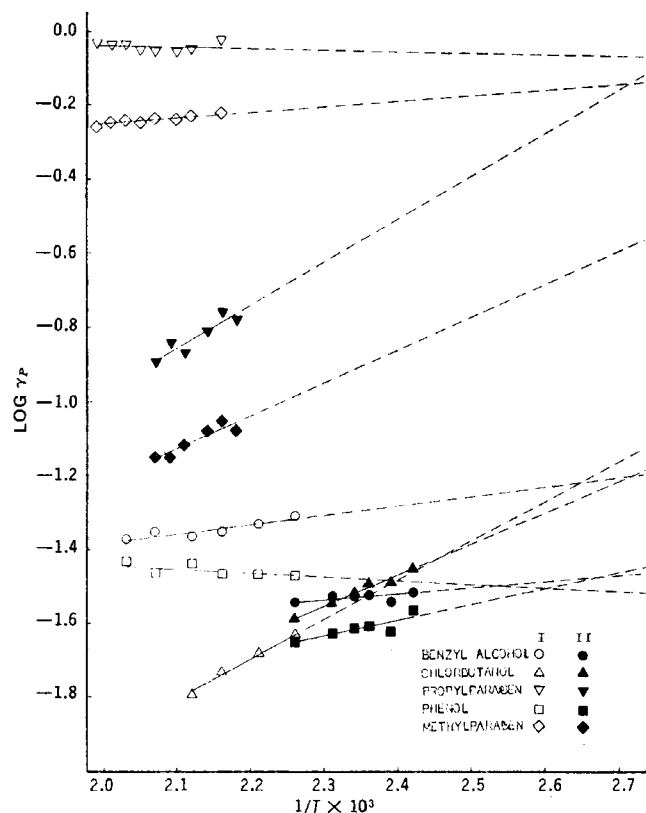

Figure 2 —*Apparent activity coefficients at varying temperatures for the bacteriostatic agents. Key: I, polyethylene; and II, polyvinyl chloride.*

obtain the coefficients at ambient temperature and the temperatures used for accelerated stability testing, the plots were extrapolated as shown by the dashed lines. The theoretical premises and good linear fit of the data permitted this extrapolation which employed the regression points to reduce the error. Since the polymers contain crystalline regions at these temperatures, the extrapolated activity coefficients are pertinent only to the amorphous portion of the polymer (15).

The extrapolated activity coefficients are presented in Table V. The solutes are arranged in the order of decreasing activity coefficient values, *i.e.*, in increasing order of interaction affinity, within each group. These values reveal that, for the most part, the sorption of the solutes by the polymers is favored even at ambient temperatures, as evidenced by the values smaller than unity. Referring to the organic solvents in respect to polyethylene, it may be seen that the highest activity coefficients were exhibited by *n*-butanol, 2-nitropropane, and *tert*-butanol. The lowest values were observed for toluene, hexane, and octane, and this finding agrees with the general knowledge on miscibility and sorption for these systems. Comparing the values in relation to the polymers reveals a general trend for reversal of the interaction affinity grading. Thus, octane and hexane were among the least miscible with polyvinyl chloride, and 2-nitropropane showed a relatively strong mixing tendency with that polymer. The relative ordering of normal and *tert*-butanol was reversed, and the same was observed with methyl isobutyl ketone and butyl acetate.

Concerning the bacteriostatic agents, the least interaction affinity with both polymers was exhibited by the parabens. Propylparaben was more miscible with polyethylene than methylparaben and vice versa in respect to polyvinyl chloride. The miscibility of the

Table V—Apparent Activity Coefficients at 25° [a]

On Polyethylene		On Polyvinyl Chloride	
n-Butanol	3.800	Octane	1.000
2-Nitropropane	0.813	*tert*-Butanol	0.902
tert-Butanol	0.432	Hexane	0.393
Butyl acetate	0.097	*n*-Butanol	0.237
Methyl isobutyl ketone	0.040	Methyl isobutyl ketone	0.062
Toluene	0.017	2-Nitropropane	0.042
Hexane	0.009	Butyl acetate	0.030
Octane	0.009	Toluene	0.026
Methylparaben	1.070	Propylparaben	1.335
Propylparaben	0.776	Methylparaben	0.891
Chlorobutanol	0.367	Chlorobutanol	0.182
Benzyl alcohol	0.089	Phenol	0.048
Phenol	0.038	Benzyl alcohol	0.039

[a] The coefficients at 30, 40, and 60° are presented in the dissertation. The relative grading remained the same at these temperatures.

others was in the following increasing order: with polyethylene—chlorobutanol < benzyl alcohol < phenol, and with polyvinyl chloride—chlorobutanol < phenol < benzyl alcohol. It may also be seen that at ambient conditions the miscibility of chlorobutanol and benzyl alcohol was less with polyethylene than with polyvinylchloride; for phenol, it was slightly less with polyvinyl chloride than with polyethylene.

The experimental error incurred during the GLC procedures did not exceed 3%, as evidenced by the fluctuations of replicate runs and the linear fit in Figs. 1 and 2. The errors arising from gas imperfections, approximations of vapor pressures, and other similar data are estimated to be within 5%.

CONCLUSIONS

This work has shown that GLC techniques may be applied successfully for evaluating and grading interaction affinities between some polymers employed in packaging and low molecular weight compounds contained in food and pharmaceuticals or used for package fabrication. The methodology is applicable for low molecular weight compounds which are volatile and chemically stable at the temperatures employed for GLC. The chemical stability of the polymers at these temperatures is also a necessary requirement. The procedures were fast and relatively simple to perform. The required instrumentation is widely available. The proposed auxiliary techniques were accessible and adequate for generating the data needed for the computations, and values not previously reported were obtained by this means. The methodology employed may, therefore, be considered a useful tool for evaluating and grading sorption–desorption propensities for polymer–low molecular weight compound systems.

The thermodynamics of the interactions and their elucidation are presented in the second part of this work (5).

APPENDIX

Table VI—Specific Retention Volumes at Varying Temperatures for the Organic Solvents

Solute	Polymer	383°K.	V_g^0, ml. 413°K.	443°K.
n-Butanol	Polyethylene	—	4.398	2.812
2-Nitropropane	Polyethylene	—	15.350	8.643
tert-Butanol	Polyethylene	—	5.117	3.556
Methyl isobutyl ketone	Polyethylene	—	21.050	11.206
Butyl acetate	Polyethylene	—	28.186	15.300
Hexane	Polyethylene	—	9.119	4.876
Octane	Polyethylene	—	41.650	18.857
Toluene	Polyethylene	—	41.740	22.206
Octane	Polyvinyl chloride	4.781	3.178	—
Hexane	Polyvinyl chloride	—	1.059	—
tert-Butanol	Polyvinyl chloride	2.737	2.174	—
n-Butanol	Polyvinyl chloride	13.202	6.887	—
Butyl acetate	Polyvinyl chloride	25.069	11.626	—
Methyl isobutyl ketone	Polyvinyl chloride	22.063	11.763	—
Toluene	Polyvinyl chloride	24.079	11.262	—
2-Nitropropane	Polyvinyl chloride	34.448	15.778	—

Table VII—Specific Retention Volumes at Varying Temperatures for the Bacteriostatic Agents

Solute	Polymer	413°K.	V_g^0, ml. 443°K.	463°K.
Benzyl alcohol	Polyethylene	—	85.02	51.64
Phenol	Polyethylene	—	59.38	30.08
Chlorobutanol	Polyethylene	—	56.45	35.13
Benzyl alcohol	Polyvinyl chloride	141.95	54.46	—
Chlorobutanol	Polyvinyl chloride	40.01	19.15	—
Phenol	Polyvinyl chloride	83.78	33.64	—

Solute	Polymer	463°K.	V_g^0, ml. 483°K.	503°K.
Propylparaben	Polyethylene	193.86	102.53	55.90
Methylparaben	Polyethylene	94.59	53.13	30.87
Propylparaben	Polyvinyl chloride	389.42	263.65	—
Methylparaben	Polyvinyl chloride	240.57	164.70	—

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