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Mechanism of liquid adsorption chromatography in thin-layer chromatography with ternary mobile phases

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ABSTRACT

A model of the chromatographic process which takes into account molecular association in the mobile phase is discussed. This model is applied to the study of the association effects in liquid adsorption chromatography with ternary mobile phases. Jaroniec's equation was used to describe the molecular interactions in the systems investigated. The assumed model of the molecular mechanism of the chromatographic process significantly broadens the possibility of interpretation of experimental chromatographic data.

INTRODUCTION

The optimum separation conditions of mixtures in liquid adsorption chromatography very often require application of mixed (multi-component) mobile phases. A great number of the mobile phase components used in the chromatographic separation process cause this process to become more complex. Physicochemical interpretation of experimental data in such chromatographic systems is difficult.

In our previous papers [1–3] a new method of presentation of the retention data obtained from systems with ternary mobile phases was described. It was stated that this method facilitated a complete interpretation of the process realized in such a system.

The process of liquid adsorption chromatography is determined by molecular interactions in the surface and mobile phases. Lately Jaroniec and co-workers [4–7] as well as Ościk-Mendyk and co-workers [8–10] have presented a simple model of the chromatography process mechanism and then verified it in thin-layer chromatography (TLC) and high-performance liquid chromatography (HPLC) systems. The investigations have until now shown the applicability of this model for interpretation of experimental retention data. It can be also concluded that the results of these investigations have not yet provided complete information relating to the molecular mechanism of the chromatographic process in real systems with a multi-component mobile phase.

THEORETICAL

The chromatographic process in solid-liquid systems is determined by competitive adsorption of the chromatographed substance molecules and the components of a mobile phase. The adsorption phenomenon can be presented as the exchange reaction of the molecules of the chromatographic system components between surface and mobile phases. Molecular interactions in both phases, including interactions with the adsorbent surface, determine the results of such exchange. The strong specific interactions leading to the formation of molecular complexes (associates) in both phases play an especially important role. In many cases a model of the chromatographic process which assumes the existence of double associates in the mobile phase is sufficient to describe the properties of the chromatographic systems [11,12]. The strong localization effects of the test substance and solvent molecules during the adsorption process on silica gel or other oxide adsorbents cause the destruction of the associates in the surface phase. It can be assumed that in such systems only single molecules are adsorbed.

In liquid adsorption chromatography with binary and ternary mobile phases three most important types of associates can be distinguished: association of the molecules of the more polar mobile phase component (autoassociates of the 1-1 type); association of the chromatographed substance molecules and those of the more polar solvent (associates of the s-1 type); and association of both mobile phase components; (associates of the 1-2 and 1-3 types).

The non-specific interactions between the molecules of non-polar or weakly polar components of the mobile phase (components 2 and 3) can be ignored.

In the earlier investigations it was stated that only the first two types of association played a significant role in the chromatographic process. For such systems the equation describing the effect of the above specific molecular interactions on the retention parameter k'_s has been derived [4-7].

$$k'_s = K_{s1} q^{-1} \frac{y_1}{x_1} [1 + (C_1 - 2L_2)]^{-1} \quad (1)$$

where $k'_s = q^{-1} y_s/x_s$ is the capacity factor of the chromatographed substance; x_1, y_1 are the molar fractions of the most polar mobile phase component in the mobile and surface phase, respectively; x_s, y_s are the molar fractions of the chromatographed substance in the mobile and surface phases, respectively; q is a proportionality coefficient, characteristic of a given adsorbent and independent of the nature of the mobile phase; $K_{s1} = y_s x_1/x_s y_1$ is the thermodynamic equilibrium constant of the phase exchange reaction between the molecules of the substance and of the more polar component of the mobile phase; $C_1 = x_{s-1}/x_s x_1$ is the equilibrium constant of formation of s-1 type bimolecular associates; x_{s-1} is the molar fraction of bimolecular associates of the s-1 type; $L_2 = x_{1-1}/x_1^2$ is the equilibrium constant of the formation of bimolecular associates of the 1-1 type (double autoassociates of the most polar mobile phase component); and x_{1-1} is the molar fraction of bimolecular associates of the 1-1 type.

In the theoretical considerations leading to eqn. 1, the model of the liquid adsorption chromatography process which introduces several simplifying assump-

tions was accepted [4]. Among others, the assumption of a very small concentration of the chromatographed substance in the chromatographic system considered was accepted. For this reason y_s and x_s are often neglected in theoretical considerations. Eqn. 1 can be written in the linear form [5-7]:

$$\frac{y_1}{k'_s x_1} = \alpha + \beta x_1 \quad (2)$$

where

$$\alpha = qK_{s1}^{-1} \text{ and } \beta = q(C_1 - 2L_2)K_{s1}^{-1}$$

From the boundary conditions of the above equation α and β values can be calculated. These parameters are connected with the basic physicochemical factors determining the chromatographic process. The reciprocal of α is proportional to the adsorption equilibrium constant of a given chromatographed substance (K_{s1}). Numerical value of the ratio $\beta/\alpha = C_1 - 2L_2$ permits quantitative specification of the predominant association effects occurring in the mobile phase of the system investigated. These effects are expressed by the equilibrium constants C_1 and L_2 [8,9]. The positive value of β betokens that $C_1 > 2L_2$. In such a system, apart from adsorption expressed by the equilibrium constant K_{s1} , the second factor, association of s-1 type, has a predominant effect on the retention process. Negative β values betoken that autoassociation of most polar solvent molecules of a mobile phase (association of the 1-1 type) plays a significant role in the retention process. It can be said that $\beta = (C_1 - 2L_2)\alpha$ expresses the intensity of the association effect in the retention data of a given chromatographic system.

The relationships described by eqn. 2 include the experimental data relating to the chromatographic retention k'_s and adsorption of the more polar mobile phase component (y_1). In earlier papers [5-7] it has been stated that for a strongly polar solvent it can be assumed that $y_1 = 1.0$ in the whole concentration range of this solvent in the mobile phase. It has also been concluded that the linear relationship $1/k'_s x_1$ vs. x_1 for $x_1 \geq 0.5$ is identical with the relationship y_1/k'_s vs. x_1 taking into account the changes in y_1 values for $x_1 < 0.5$ [13,14]. It can be stated that for many systems with low concentrations of the most polar mobile phase component the conditions of the chromatographic process do not correspond to those of the assumed theoretical model. Thus, in the study of the chromatographic process we can use as a good approximation a simpler form of eqn. 2:

$$\frac{1}{k'_s x_1} = \alpha + \beta x_1 \quad (3)$$

Eqn. 3 permits description of the chromatographic process on the basis of the experimental retention data (k'_s) only. For binary mobile phases the equilibrium constants K_{s1} , C_1 and L_2 are the functions of the nature of mobile phase components and temperature. As all measurements were carried out at constant temperature the changes in numerical values of the above equilibrium constants depend only on the nature of mobile phase components and of the tested substances. The chromato-

graphic process becomes more complex with the increase in number of the mobile phase components. The description of the chromatographic process with a ternary mobile phase 1 + 2 + 3 (component 1 is the most polar) may also be based on eqn. 3. In this case one can assume that the ratio $r = x_2/x_3$ is constant. Changes in the value of r cause changes in α and β and thus changes in the equilibrium constants K_{s1} , C_1 and L_2 . Analysis of the parameter changes of eqn. 3 due to the changes in r may allow complete interpretation of the chromatographic process mechanism in such a complicated systems. An attempt to carry out such an analysis for a few selected chromatographic systems with a ternary mobile phase is the main object of this paper.

The aim of study is to establish:

(1) Whether the proposed model can simplify the explanation of some physicochemical aspects of the molecular mechanism of the liquid adsorption chromatography process with a ternary mobile phase.

(2) How far eqn. 3 permits description of real chromatographic systems in complex conditions when a multi-component mobile phase is used.

(3) Whether the data obtained from eqn. 3 are realistic enough to have a definite physical meaning.

EXPERIMENTAL

The measurements were made at 293K using TLC. Silica gel 60H (Merck, Darmstadt, Germany) was used as the adsorbent. The components of mobile phases were: *n*-heptane, carbon tetrachloride, toluene, benzene, trichloroethylene, acetone and *n*-propanol. In all mixed mobile phases (binary and ternary) concentration of the more or most polar solvent was equal to 0.1, 0.3, 0.5, 0.7 and 0.9 of the molar fraction value. The criterion of polarity was solvent dipole moment (Table I). Naphthalene, *o*-nitrophenol, *o*-nitroaniline, fluorenone and isoquinoline were the test substances. Other details concerning the TLC measurements were described in the previous paper [9].

RESULTS AND DISCUSSION

The mechanism of the chromatographic process may be determined by molecular interactions of the 1-1 and s-1 types depending on the nature of the

TABLE I

DIPOLE MOMENTS AND CLASS, ACCORDING TO PIMENTEL AND McCLELLAN, OF MOBILE PHASE COMPONENTS

Solvent	Dipole moment (D)	Class
<i>n</i> -Heptane	0.00	N
Carbon tetrachloride	0.00	N
Benzene	0.00	N/B
Toluene	0.39	N/B
Trichloroethylene	0.94	A
<i>n</i> -Propanol	1.69	AB
Acetone	2.76	B

chromatographic system. In this connection interpretation of the experimental data was divided into two parts.

In the first part, chromatographic systems with ternary mobile phases containing two non-polar and one polar solvent from A, B or AB class according to the Pimentell and McClellan classification [15] were examined. Non-polar solvents differed in their physicochemical properties (carbon tetrachloride, benzene). Fig. 1 shows the mobile phase composition presented on Gibbs triangle. The vertex S_1 of this triangle corresponds to trichloroethylene (A), acetone (B) and *n*-propanol (AB), respectively. The analysis of the molecular mechanism was based on eqn. 3 for $x_1 \geq 0.5$ [13,14]. For such chosen concentrations of mobile phase components and because of the preadsorption process the demixing phenomenon may be ignored [16].

In each chromatographic system the measurements were made for five mixed phases according to the scheme presented in Fig. 1. From this scheme it can be seen that the measurements were made for two binary mobile phases, $S_1 + C_6H_6(S_2)$ and $S_1 + CCl_4(S_3)$ (systems 1 and 5 in Fig. 1), and for three ternary mobile phases, $S_1 + S_2 + S_3$. In the ternary mobile phase the ratio of mole fractions $r = x_{S_2}/x_{S_3}$ was constant and equal to 1/3, 1 and 3, respectively (systems 2, 3, and 4 in Fig. 1). The chromatographic measurements obtained for the selected substances are presented in Fig. 2 in the form of $1/k'_s x_1$ vs. x_1 relationships. Values of the eqn. 3 parameters obtained in three systems with different ternary mobile phases used are listed in Table II. Fig. 3 illustrates the changes of these parameters with the changes of r values. Complete interpretation of the results presented in Figs. 2 and 3 and Table II is based on the analysis of the changes of α and β in eqn. 3 with the changes of the mobile phase composition.

It can be seen that only negative values of β were obtained in the first part of the experiments. It can be stated that in these systems autoassociation of the molecules of polar mobile phase components (association of the 1-1 type) plays an important role in the chromatographic retention mechanism.

Values of $1/\alpha$ and their changes with the changes of the nature of the mobile phase depend, above all, on the properties of a polar component of mobile phase. Strong adsorption of *n*-propanol molecules on the silica gel surface leads to low adsorption of the chromatographed substance molecules. In this case numerical values of the equilibrium constant K_{s_1} are very low and do not differ significantly from one

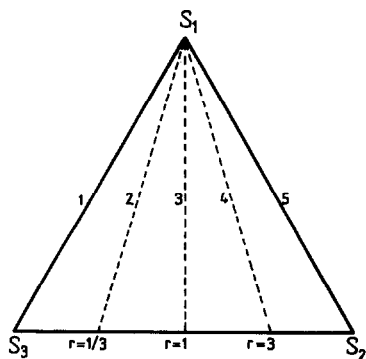


Fig. 1. Mobile phases used in the investigated systems. S_1 is the most polar solvent, S_2 and S_3 are non-polar or weakly polar solvents.

another. Variations of the mole fractions ratio of non-polar mobile phase components do not influence the value of $1/\alpha$, which increases slightly with the increase in benzene concentration in the mobile phase. This slight increase of $1/\alpha$ is observed mainly in the case of *o*-nitrophenol and *o*-nitroaniline. It can be said that the increase in benzene concentration in mobile phase causes the increase of the interactions between the molecules of *n*-propanol (AB) and benzene (N/B). In this connection both substances mentioned above adsorb strongly on the silica gel surface.

The numerical values of $1/\alpha$ are also low and decrease linearly with increasing benzene concentration in the mobile phase when acetone is used as a polar solvent (Figs. 2 and 3). Molecules of acetone can form autoassociates due to a very strong dipole moment (2.76 D). In this system benzene molecules can compete more strongly with the chromatographed substance molecules for active centres on the adsorbent surface. Thus, the greatest changes in $1/\alpha$ are again observed for *o*-nitrophenol and *o*-nitroaniline. The tendency of these substances to form intramolecular hydrogen bonds can also be taken into account. Apart from strong adsorption of *n*-propanol and acetone molecules, the association of the mobile phase components can also play a very important role in the chromatographic process. This can be confirmed from numerical values of β and β/α . For *n*-propanol, values of β/α are practically the same for all tested substances and do not change with the changes of the concentration of

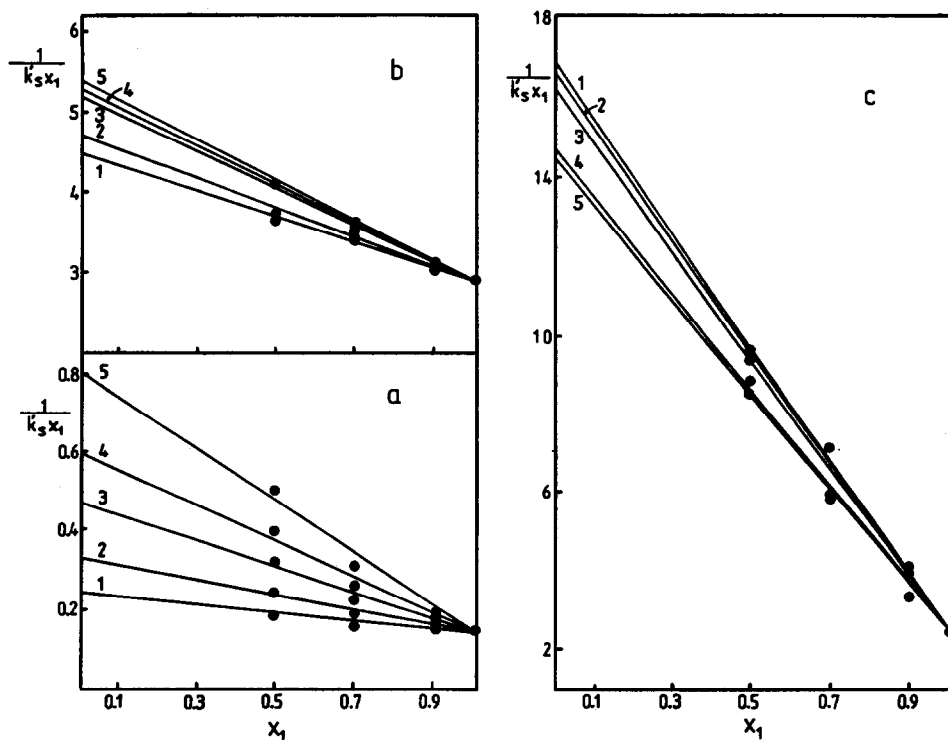


Fig. 2. $1/k_s x_1$ vs. x_1 plotted for *o*-nitroaniline in (a) carbon tetrachloride–benzene–trichloroethylene, (b) carbon tetrachloride–benzene–acetone and (c) carbon tetrachloride–benzene–*n*-propanol. Numbers 1–5 indicate mobile phase compositions according to the scheme in Fig. 1.

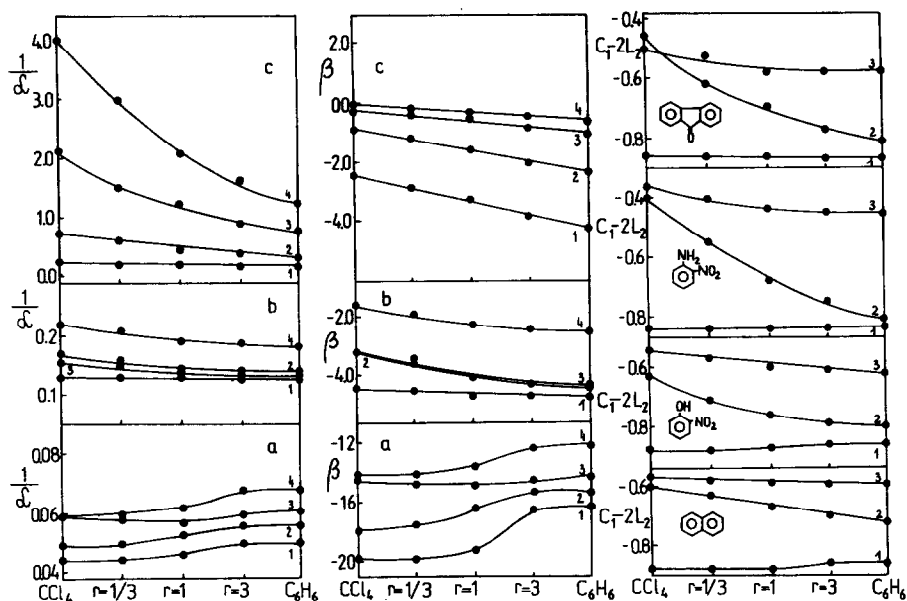


Fig. 3. Dependence of $1/\alpha$ (left panel) and β (centre panel) on different values of r for (1) naphthalene, (2) *o*-nitrophenol, (3) fluorenone and (4) *o*-nitroaniline in (a) carbon tetrachloride-benzene-*n*-propanol, (b) carbon tetrachloride-benzene-acetone and (c) carbon tetrachloride-benzene-trichloroethylene. Dependence of $C_1 - 2L_2$ values (right panel) on different values of r in (1) carbon tetrachloride-benzene-*n*-propanol, (2) carbon tetrachloride-benzene-acetone and (3) carbon tetrachloride-benzene-trichloroethylene.

non-polar mobile phase components. One can suppose that such results are determined mainly by strong autoassociation of molecules of a polar mobile phase component. In the system containing acetone, lower negative values of β/α depend, in great part, on the properties of the tested substance and on the concentration of non-polar mobile phase components (decrease with the increase of benzene concentration). Assuming a constant value of the equilibrium constant for the autoassociation of acetone molecules in a given system, the observed changes in $C_1 - 2L_2$ values may be due to the changes in solvation of the chromatographed substance only, *i.e.* to the changes in the equilibrium constant C_1 (Fig. 3).

The values of $1/\alpha$ depend on the properties of the chromatographed substances when trichloroethylene is used as a polar component of the mobile phase. For example, for fluorenone and *o*-nitroaniline high values of this parameter indicate a strong adsorption of these substances on the silica gel surface in comparison with that of *o*-nitrophenol and especially of naphthalene. The magnitude of the adsorption decreases significantly with increasing benzene concentration in the mobile phase. This is due to significantly lower adsorption of trichloroethylene molecules on the silica gel surface in comparison with the adsorption of *n*-propanol and acetone molecules. The adsorption of trichloroethylene on the silica gel surface is comparable to that of benzene. Great changes in the numerical values of β/α due to changes in concentration of non-polar mobile phase components illustrate the significant effect of molecular interactions in a mobile phase on the chromatographic process.

TABLE II
VALUES OF $1/\alpha$, β , AND $\beta/\alpha = C_1 - 2L_2$ OBTAINED FOR DIFFERENT MOBILE PHASE COMPOSITIONS

Mobile phase composition ^a	Naphthalene			<i>o</i> -Nitrophenol			<i>o</i> -Nitroaniline			Fluorenone		
	$1/\alpha$	β	$C_1 - 2L_2$	$1/\alpha$	β	$C_1 - 2L_2$	$1/\alpha$	β	$C_1 - 2L_2$	$1/\alpha$	β	$C_1 - 2L_2$
$S_1 + S_3$	0.25	-2.45	-0.60	0.74	-0.85	-0.63	4.00	-0.10	-0.40	2.17	-0.21	-0.46
$S_1 + r = 1/3$	0.22	-2.75	-0.62	0.59	-1.20	-0.71	3.03	-0.18	-0.55	1.54	-0.40	-0.62
$S_1 + r = 1.0$	0.20	-3.30	-0.67	0.48	-1.60	-0.76	2.13	-0.32	-0.68	1.25	-0.55	-0.69
$S_1 + r = 3.0$	0.18	-3.90	-0.70	0.39	-2.05	-0.79	1.67	-0.45	-0.75	0.91	-0.85	-0.77
$S_1 + S_2$	0.17	-4.30	-0.72	0.33	-2.40	-0.80	1.25	-0.65	-0.81	0.77	-1.05	-0.81
$S_1 + S_3$	0.13	-4.40	-0.57	0.17	-3.20	-0.54	0.22	-1.60	-0.36	0.16	-3.20	-0.50
$S_1 + r = 1/3$	0.13	-4.50	-0.58	0.16	-3.60	-0.57	0.21	-1.90	-0.40	0.15	-3.40	-0.52
$S_1 + r = 1.0$	0.13	-4.70	-0.59	0.15	-4.10	-0.60	0.19	-2.30	-0.44	0.14	-4.10	-0.55
$S_1 + r = 3.0$	0.13	-4.70	-0.59	0.14	-4.30	-0.61	0.19	-2.40	-0.45	0.14	-4.30	-0.58
$S_1 + S_2$	0.13	-4.70	-0.59	0.14	-4.40	-0.62	0.19	-2.50	-0.46	0.14	-4.30	-0.58
$S_1 + S_3$	0.04	-19.9	-0.88	0.05	-18.0	-0.88	0.06	-14.2	-0.84	0.06	-14.5	-0.86
$S_1 + r = 1/3$	0.04	-19.9	-0.88	0.05	-17.5	-0.88	0.06	-14.1	-0.84	0.06	-14.8	-0.86
$S_1 + r = 1.0$	0.05	-19.3	-0.88	0.05	-16.4	-0.87	0.06	-13.6	-0.84	0.06	-14.9	-0.86
$S_1 + r = 3.0$	0.05	-16.3	-0.86	0.06	-15.3	-0.86	0.07	-12.4	-0.84	0.06	-14.5	-0.87
$S_1 + S_2$	0.05	-16.3	-0.86	0.06	-15.3	-0.86	0.07	-12.2	-0.84	0.06	-14.2	-0.87

^a S_1 : trichloroethylene, acetone, and *n*-propanol, for top, middle and bottom group, respectively; S_2 : benzene; S_3 : carbon tetrachloride ($r = x_{S_2}/x_{S_3}$).

In the second part of our investigations chromatographic systems with ternary mobile phases of the composition acetone-*n*-heptane-toluene were used. Also, four chromatographed substances were used. The measurements were made in the same way as in the first part of our experiments. In ternary mobile phases the ratios of mole fractions ($r = x_{C_6H_5CH_3}/x_{C_7H_{16}}$) were constant and equal to 1/3, 1 and 3. Fig. 4 presents the results obtained for all compositions of a mobile phase, Table III lists the numerical values of eqn. 3 parameters for all substances and Fig. 5 illustrates the changes of these parameters with the changes in r values. Positive or negative values of β were obtained in the systems investigated. These values depend on the properties of the chromatographed substances and vary with changes of ternary mobile phase composition. Thus, according to the proposed model of the chromatographic process it can be stated that the results obtained for such systems indicate a very complex and differentiated effect of molecular interactions on the chromatographic process. This is due to significant differences in the properties of the individual mobile phase components: strong polar acetone, aromatic weakly polar toluene and non-polar saturated hydrocarbon *n*-heptane.

The values of $1/\alpha$ depend strongly on the composition of this ternary mobile phase. Weak interactions of *n*-heptane molecules with the silica gel surface mean that K_{s1} values in the acetone-*n*-heptane system are greatly differentiated depending on the properties of the substances chromatographed. Addition of toluene to such a system causes a rapid decrease of $1/\alpha$ (and thus the K_{s1} constant) because the toluene molecules participate in competitive adsorption on the adsorbent surface. In the acetone-toluene system the $1/\alpha$ values are low for all test substances and differ only slightly one from another (they are practically the same as in the acetone-benzene system).

The analysis of β and β/α values permits us to conclude that the effect of molecular interactions in a mobile phase on chromatographic retention is different

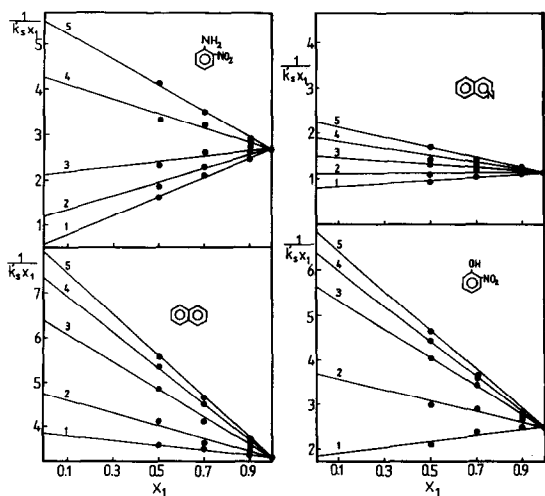


Fig. 4. $1/k'sx_1$ vs. x_1 obtained in *n*-heptane-toluene-acetone.

TABLE III
VALUES OF $1/\alpha$, β , AND $\beta/\alpha = C_1 - 2L_2$ OBTAINED FOR DIFFERENT MOBILE PHASE COMPOSITIONS

Mobile phase composition ^a	Naphthalene			o-Nitrophenol			o-Nitroaniline			Fluorenone		
	$1/\alpha$	β	$C_1 - 2L_2$	$1/\alpha$	β	$C_1 - 2L_2$	$1/\alpha$	β	$C_1 - 2L_2$	$1/\alpha$	β	$C_1 - 2L_2$
$S_1 + S_3$	0.26	-0.55	-0.14	0.54	0.70	0.38	1.82	2.10	3.82	1.25	0.35	0.44
$S_1 + r = 1/3$	0.21	-1.50	-0.32	0.27	-1.20	-0.32	0.83	1.50	1.25	0.91	0.05	0.05
$S_1 + r = 1.0$	0.16	-3.15	-0.49	0.18	-3.20	-0.57	0.48	0.65	0.31	0.67	-0.40	-0.27
$S_1 + r = 3.0$	0.14	-4.00	-0.54	0.16	-3.85	-0.61	0.24	-1.55	-0.36	0.53	-0.80	-0.42
$S_1 + S_2$	0.13	-4.70	-0.59	0.15	-4.45	-0.65	0.18	-2.80	-0.51	0.44	-1.10	-0.49

^a S_1 : acetone; S_2 : toluene; S_3 : n-heptane ($r = x_{S_2}/x_{S_3}$).

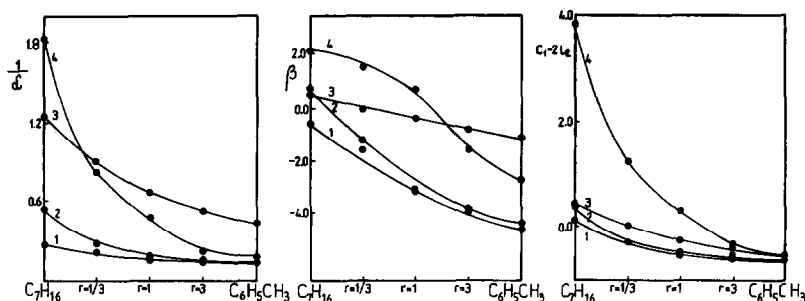


Fig. 5. Dependence of $1/\alpha$ (left), β (centre) and $C_1 - 2L_2$ (right) parameters on different values of r for (1) naphthalene, (2) *o*-nitrophenol, (3) isoquinoline and (4) *o*-nitroaniline in *n*-heptane-toluene-acetone.

from that in the first part of the study. For the acetone-*n*-heptane system and for ternary systems of high concentration of *n*-heptane the β values are positive for all substances except naphthalene. For higher concentrations of toluene the β values are negative. It can be supposed that non-active *n*-heptane molecules facilitate the formation of stable associates between the molecules of acetone and the substance chromatographed (associates of the s-1 type or polymolecular associates). The molecules of relatively active and weakly polar toluene significantly hinder this process and the increase of toluene concentration shifts the $C_1 - 2L_2$ value towards the autoassociation of acetone molecules.

CONCLUSIONS

The study of the mechanism of liquid adsorption chromatography with a ternary mobile phase made on the basis of eqn. 3 produced the following conclusions.

(1) The assumed model of the molecular mechanism of the investigated process significantly broadens the possibility of interpretation of experimental chromatographic data.

(2) The values obtained for the parameters in eqn. 3 allow quantitative evaluation of the molecular interactions determining the retention mechanism.

(3) The quantitative data resulting from this model and from eqn. 3 are realistic and relate to real values of physicochemical parameters characterizing a given chromatographic system.

(4) The widening of these investigations to include other chromatographic systems will produce a better understanding of a complicated retention process.

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