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SIMPLE MODEL BASED ON SOLUBILITY PARAMETERS FOR LIQUID DESORPTION OF ORGANIC SOLVENTS ADSORBED ON ACTIVATED CARBON

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SUMMARY

Liquid desorption recoveries were studied on two carbons (SKC and Merck) commonly used for sampling of solvent vapours in air. For non-polar solvents it was found that carbon disulphide is an effective desorber as long as its surface free energy exceeds that of the adsorbed solvents. For polar solvents the recoveries with carbon disulphide are related to a parameter, ΔE_{DA}, that could be estimated from Hansen's solubility parameters. By using compounds with high ΔE_{DA} values as 5% mixtures in carbon disulphide recoveries could be raised to *ca.* 100% for 2-ethoxyethanol, butanol, acetone, dioxane and propyl acetate.

INTRODUCTION

The collection of organic solvent vapours on activated carbon followed by liquid desorption and gas chromatographic (GC) analysis has been used for a long time as a method to determine workers' exposure to industrial solvents. As early as 1964 Otterson and Guy[1] concluded that the use of carbon disulphide (CS_2) as desorbing agent resulted in almost complete recoveries for vapours of styrene, xylene and perchloroethylene collected on activated carbon. Slightly lower recoveries were obtained with toluene and benzene as desorbing agents, and the use of mesitylene and xylene yielded poorer results. CS_2 was used throughout the extensive study conducted by the National Institute for Occupational Safety and Health (NIOSH)[2] for desorption of non-polar solvents. For most aliphatic, aromatic and halogenated solvents recoveries of *ca.* 100% were obtained: exceptions included styrene, naphthalene, tetrachloroethane and tetrabromoethane.

The NIOSH study clearly demonstrated that for polar solvents, such as alcohols, ketones and ethers, CS_2 was not a good choice. A significant increase could in many cases be obtained by addition of a small amount of an alcohol, *e.g.* methanol. Other desorbing agents, such as dimethylformamide (DMF) in conjuction with a backflush technique[3] or 5% methanol in dichloromethane (photoionization detector)[4], have been found to increase recoveries to *ca.* 100%. However, with these procedures several common non-polar solvents, such as toluene and benzene, are not quantitatively recovered.

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Most suggestions concerning desorbing agents have been made on purely empirical grounds. As far as we know no systematic approach to liquid desorption of solvent vapours from activated carbon has been published. In spite of the difficulty of finding a useful model for an adsorbent that is heterogenous with regard to the chemical nature of the sites as well as the size and shape of the micropores responsible for most of the adsorption, a coarse model is valuable since it limits the number of possible desorbing agents to test. In practice, further limitations are set by the chromatographic conditions since the desorbing mixture has to be chosen so that it does not interfere with peaks emanating from the adsorbed vapours.

The purposes of this paper are to relate adsorption on both the hydrophobic and the hydrophilic parts of the surface to available physical–chemical parameters, and to apply the results to liquid desorption.

THEORY

Briefly, activated carbon consists of basal carbon layers of a "graphite-like" structure, with a hydrophobic surface, and of surface oxides located on the reactive edges of the carbon layers, giving a hydrophilic surface[5]. Usually the hydrophobic surface amounts to more than 90% of the available adsorption area.

The interaction between non-polar surfaces and liquids is predominately due to dispersion forces. The magnitude of the dispersive part of the free energy of interaction, $-W_A^d$, may be estimated[6] from eqn. 1:

$$W_A^d = 2\,(\gamma_l^d \cdot \gamma_s^d)^{\frac{1}{2}} \tag{1}$$

where W_A^d is dispersive part of the work of adhesion and γ_l^d and γ_s^d are the dispersive parts of the surface free energies of the liquid and solid, respectively. For non-polar surfaces, eqn. 1 is a good estimate of the total work of adhesion, W_A. For the sake of simplicity this approximation will be used here.

The solubility parameter, δ, is defined as:

$$\delta^2 = \Delta E_v / V$$

where ΔE_v is the energy of vaporization and V is the molar volume. Hansen and Beerbower[7] have proposed, that on the basis of the different types of intermolecular interaction, the solubility parameter may be further subdivided into three independent components: one due to dispersion forces (δ_D), one due to dipole moments (δ_P) and one due to hydrogen bonding (δ_H). This subdivision is based on the notion that the polar interactions may be divided into contributions from the two phases in the same way as for the dispersion forces (eqn. 1). This assumption can be criticized on theoretical grounds, but the division has been found useful in practice and the parameters are available for almost all industrial solvents. Some examples are given in Tables I and II.

Beerbower[8] has found the following relation between the dispersive component of the surface tension and Hansen's dispersive solubility parameter:

$$\gamma^d = 0.0715 V^{1/3} \cdot \delta_D^2 \tag{2}$$

TABLE I

DATA FOR NON-POLAR COMPOUNDS

The table gives surface free energy, γ (mJ/m^2) (*Handbook of Chemistry and Physics*), dispersive solubility parameter[7], δ_D (cal/cm^3)$^{1/2}$, and molar volume, V (cm^3), for the non-polar organic solvents used in this study.

No.	*Compound*	γ	δ_D	V
1	Hexane	18.4	7.3	131.6
2	Octane	21.8	7.6	163.5
3	Cyclohexane	25.5	8.2	108.7
4	Dichloromethane	26.5	8.9	63.9
5	Carbon tetrachloride	27.0	8.7	97.1
6	Toluene	28.5	8.8	106.8
7	Benzene	28.9	9.0	89.4
8	*m*-Xylene	28.9	8.7*	121.2*
9	1,1,2,2-Tetrachloroethane	36.0	9.2	105.2
10	Tetrachloroethylene	31.7	9.3	101.1
11	Chlorobenzene	33.6	9.3	102.1
12	Styrene	32.1	9.1	115.6
13	Ethylbenzene	29.2	8.7	123.1
14	Bromoform	41.5	10.5	87.5
15	Nitrobenzene	43.9	9.8	102.7
16	1,1,2,2-Tetrabromoethane	49.7	11.1	116.8
17	Trichloroethylene	32.0	8.8	90.2
	Carbon disulphide	32.3	10.0	60.0

* Value for *o*-xylene.

where V is the molar volume and δ_D is Hansen's dispersive solubility parameter. This relation can be used to estimate the dispersive component for polar liquids.

If γ_s^d is assumed to be constant, the dispersive part of the work of adhesion (and indirectly the adsorption strength) should be a function of the surface free energy for the liquid only, or in terms of Beerbower's relation a function of $V^{1/3} \cdot \delta_D^2$. For the use of CS_2 as a desorbing agent for non-polar solvents, the recovery would be expected to be dependent on the surface free energy of the adsorbate, which could be considered to be in the liquid state owing to condensation in the micropores. Compounds with low surface free energy, *e.g.* hexane, are not expected to work well as desorbing agents, in contrast to benzene or CS_2.

If we now consider the hydrophilic part of the surface, "acid–base" interactions (electron donor–acceptor interactions) are generally considered to be the most important forces for adsorption of polar compounds[9,10]. Adsorption of "acidic and basic" polymers is well correlated with the donor–acceptor properties of the solvents used. According to Jensen[11] and others, the donor (basic) and acceptor (acidic) properties of a liquid can be expressed by two numbers, donor number (DN) and acceptor number (AN). These numbers are determined experimentally by measuring the strength of interaction with a standard acid (gives DN) and a standard base (gives AN). It is recognized that DN and AN give a description with a closer relation to molecular parameters of the polar interactions than δ_P and δ_H. Unfortunately DN and AN are known for relatively few solvents: some examples are given in Table II.

TABLE II

DATA FOR POLAR COMPOUNDS

The table gives donor numbers (DN) and acceptor numbers (AN)[11], Hansen's solubility parameters[7], δ (cal/cm^3)$^{1/2}$, ΔE_{DA} values (kcal/mol, calculated with eqn. 3) and molar volume, V (cm^3), for the polar solvents used in this study.

No.	*Compound*	*DN*	*AN*	*Solubility parameter*			ΔE_{DA}	V
				δ_D	δ_P	δ_H		
1	Ethyl formate	–	–	7.6	4.1	4.1	2.7	80.2
2	Methyl acetate	16.5	–	7.6	3.5	3.7	2.1	79.7
3	Ethyl acetate	17.1	–	7.7	2.6	3.5	1.9	98.5
4	Butyl acetate	–	–	7.7	1.8	3.1	1.7	132.5
5	2-Ethoxyethyl acetate	–	–	7.8	2.3	5.2	4.4	136.2
6	Tetrahydrofuran	20.0	8.0	8.2	2.8	3.9	1.9	81.7
7	*p*-Dioxane	14.8	10.8	9.3	0.9	3.6	1.2	85.7
8	Acetone	17.0	12.5	7.6	5.1	3.4	2.8	74.0
9	Butanol	–	–	7.8	2.8	7.7	6.1	91.5
10	Propanol	–	–	7.8	3.3	8.5	6.3	75.2
11	Ethanol	20.5	37.1	7.7	4.3	9.5	6.4	58.5
12	Cyclohexanol	–	–	8.5	2.0	6.6	5.0	106.0
13	Diacetone alcohol	–	–	7.7	4.0	5.3	5.5	124.2
14	2-Ethoxyethanol	–	–	7.9	4.5	7.0	6.7	97.8
15	Acetonitrile	14.1	18.9	7.5	8.8	3.0	4.5	52.6
16	Dimethylformamide	26.6	16.0	8.5	6.7	5.5	5.8	77.0
	2-Butoxyethanol	–	–	7.8	2.5	6.0	5.6	131.6
	Diethylene glycol monoethyl ether	–	–	7.9	4.5	6.0	7.4	130.9
	Nitrobenzene	4.4	14.8	9.8	4.2	2.0	2.2	102.7
	Benzyl alcohol	–	–	9.0	3.1	6.7	5.6	103.6
	Acetophenone	–	–	9.6	4.2	1.8	2.4	117.4

Ideally a good correlation of adsorption on the polar surface of activated carbon with the properties of the adsorbate should be based on an estimation of the DN and AN of the surface. This could possibly be done provided that we have a knowledge of the chemical nature of the adsorption sites. Attempts to reveal these structures were made by the use of electron spectroscopy for chemical analysis (ESCA) but these did not give the desired result.

However, another approach could be tried. It has been found that in many cases there is a good correlation between Hansen's polar solubility parameters (see Table II) and ΔE_{DA}[10], where $\Delta E_{\mathrm{DA}} = \mathrm{DN} \cdot \mathrm{AN}/100$ (kcal/mol):

$$\Delta E_{\mathrm{DA}} = V \cdot (\delta_{\mathrm{P}}^2 + \delta_{\mathrm{H}}^2)/1000 \qquad (3)$$

If it is assumed that all hydrophilic sites as an average have about the same donor and acceptor strength, the interaction energy as a geometric average of the DN and AN values (ΔE_{DA}) of the adsorbate would be expected to show some correlation with the observed adsorption in a solvent with high dispersive interactions like CS_2 (*i.e.* the polar interactions of the solvent with the surface are relatively small and only

CS_2 is adsorbed on the hydrophobic surface). For amphoteric solvents with about equal donor and acceptor strengths (alcohols) an overestimation of adsorption would probably be the result, and for solvents that are typically donors or acceptors an underprediction could be expected.

For a highly microporous adsorbent like activated carbon, there are phenomena related to the pore structure that could be of importance. In small pores (micropores less than 20 Å) the molecular diameter is of the same magnitude as the pore diameter. If the ratio of these two diameters is close to unity, the force field of the opposite pore wall contributes significantly to the adsorption energy[12]. Very small pores (or constrictions in the pore system) could be inaccessible to larger molecules. This pore size discrimination could be of importance when a small desorber molecule is substituted for a larger one. The micropore diffusion rate decreases considerably when the molecular diameter increases, and a long equilibration time would be the result.

The surface energy is given against air or the liquid's own vapour. The orientation of the molecule towards the surface is important[13]. A planar molecule such as benzene "fits the graphite-like" surface very well compared with branched aromatics such as butylbenzene. Hexane could be expected to interact more closely with the surface than cyclohexane, where the average distance to the surface of the atoms may be dependent on the "chair" or "boat" configuration of the molecule.

A desorption agent can work in two ways. First, it can displace the adsorbed compounds from the surface and thus decrease the number of available sites. Second, it can interact with the analyte in the solution thereby reducing the affinity of the adsorption sites. A good desorption agent usually combines these two properties. A compound that is strongly adsorbed should be a good displacing agent for compounds more weakly adsorbed, provided that adsorption takes place on the same type of sites.

EXPERIMENTAL

Two carbons SKC 120 (SKC, Eightyfour, PA, U.S.A.) and Merck (Nr. 9624) were used in this study: their characteristics are shown in Table III. Methods used for characterization have been described earlier[5].

Dried carbon (100 mg) was placed in a 3-ml screw-cap vial and the solvents (p.a. quality or better) were added from a syringe. After a delay of at least 24 h, samples were desorbed with 1.0 ml of desorbing agent. The equilibration time was

TABLE III

CHARACTERISTICS OF ACTIVATED CARBONS USED

	SKC	*Merck*
BET-surface area (m^2/g)	1120	1150
Pore volume (cm^3/g)	0.59	0.60
Micropore volume (cm^3/g)	0.43	0.37
Oxygen (% by weight)	*ca.* 8.7	*ca.* 10
pH in aqueous solution	9.7	5.7

ca. 24 h (with CS 2 h). Analysis was performed on a Varian 3400 gas chromatograph equipped with a flame ionization detector using non-polar or medium polar capillary columns (25 m × 0.2 mm I.D.). Samples were injected with a split injector and peak areas recorded with an Altex Chromatopak integrator.

RESULTS AND DISCUSSION

Recovery as a function of the surface free energy for a number of non-polar compounds (1 μl on 100 mg of carbon) desorbed with 1 ml of CS_2 is shown in Fig. 1. Surface free energy values have been obtained from *Handbook of Chemistry and Physics*. The same pattern was obtained for other loadings in the range 0.2–10 μl on 100 mg of carbon.

Aliphatic hydrocarbons, chlorinated aliphatics and aromatic hydrocarbons with surface free energy lower than that of CS_2 are easy to desorb, and recoveries are *ca.* 100%. Chlorobenzene and styrene, which have higher surface free energy, yield recoveries significantly less than 100%. As the surface free energy is further increased recoveries steadily fall. When compounds with about the same surface free energies are compared it is noticeable that halogenated aliphatic hydrocarbons have higher recoveries than aromatic compounds. None of these compounds is in a strict sense non-polar: halogenated compounds can act as electron acceptors and the π-electrons of the aromatics could be important. The atoms of the planar aromatic nucleus are also closer to the surface.

Recoveries obtained with hexane, cyclohexane, nitrobenzene, benzyl alcohol and acetophenone as desorbing agents are shown in Table IV. Cyclohexane gives extremely poor recoveries, and, as previously mentioned, this could be explained by the different conformations of the molecule. As anticipated, hexane is not an efficient desorbing agent: the recoveries obtained are also in this case a function of the surface free energy of the adsorbate. Of the three substituted aromatics, acetophenone and

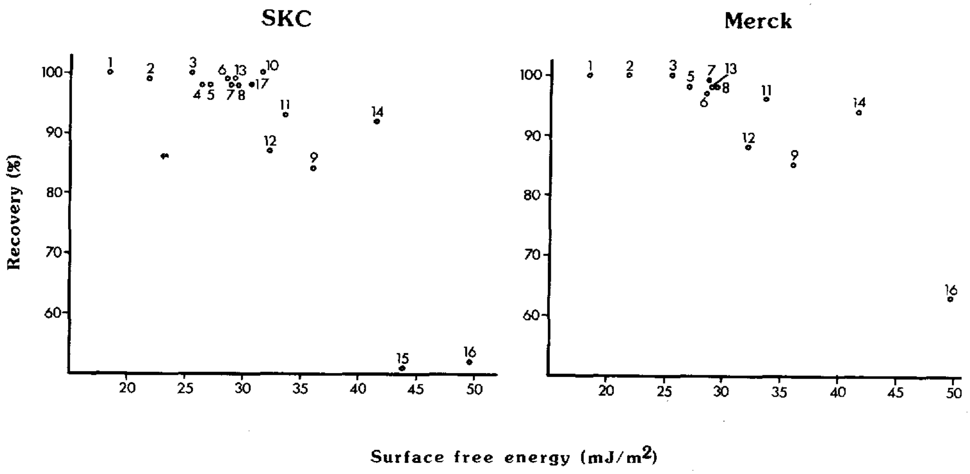

Fig. 1. Recovery as a function of the surface free energy for a number of non-polar compounds (1 μl added to 100 mg of carbon) desorbed with carbon disulphide. For key see Table I.

TABLE IV

EXPERIMENTS WITH DIFFERENT DESORBING AGENTS

The table shows recoveries for 1 μl of *n*-decane, toluene, styrene, trichloroethylene (TCE) and butanol on SKC and Merck (Mck) activated carbons using 1 ml of cyclohexane, hexane, benzyl alcohol, acetophenone and nitrobenzene as desorbing agents.

Desorbing agent	*Recovery (%)*									
	n-Decane		*Toluene*		*Styrene*		*TCE*		*Butanol*	
	SKC	*Mck*	*SKC*	*Mck*	*SKC*	*Mck*	*SKC*	*Mck*	*SKC*	*Mck*
Cyclohexane	8	–	4	–	2	–	–	–	–	–
Hexane	81	96	34	43	10	15	–	56	–	–
Benzyl alcohol	90	94	91	97	88	90	95	91	98	93
Acetophenone	97	97	100	101	96	94	99	102	101	98
Nitrobenzene	94	96	100	100	98	95	96	99	–	–

nitrobenzene yield higher recoveries than benzyl alcohol. The γ^d values calculated with Beerbower's relation are 32.3, 32.1 and 27.2, respectively. However, the interpretation of these numbers should not be stretched too far. A calculation for CS_2 gives $\gamma^d = 28$ mJ/m^2; the experimental value, γ, is 32.3, which shows that this relation is rather approximate. It is also interesting to note that acetophenone is a useful desorbing agent for polar compounds: recoveries for butanol are *ca.* 100%.

From a practical point of view none of these aromatics is especially suitable. Autosampler injections are unreliable (high viscosity?) and the long retention time is a problem. By using a backflush technique the latter problem might be solved. The results for decane could be an indication that 24 h of equilibration is not sufficient.

A scrutinization of Hansen's tables[7] does not reveal any attractive alternative to CS_2. In addition to its short retention time on most GC columns, CS_2 has another advantage, namely its low response on the flame ionization detector. Possible desorbing agents with higher surface free energy generally have high boiling points and viscosities.

Since the non-polar compounds that are difficult to desorb with CS_2 generally have low vapour pressures, the use of other adsorbents with lower capacity than activated carbon could be suggested. Nitrobenzene, for example, could be collected on porous polymers[14]. It is possible to desorb strongly adsorbed compounds from activated carbon, as has been demonstrated by Gu and Manes[15]. By the addition of naphthalene to the desorbing solvent they were able to desorb chlorinated polyaromatics. However, this procedure makes the analysis complicated.

Results obtained by other workers could also be explained in terms of the surface free energy of the desorbing agent, for dichloromethane $\gamma = 26.2$ and for DMF $\gamma^d = 22.0$ mJ/m^2 (calculated from Beerbower's relation). The latter value is probably too low. With dichloromethane the recovery for toluene was 85%[4]. With DMF the following results were obtained (compounds listed in order of increasing surface free energy): 1,2-dichloroethane ($\gamma = 24.2$ mJ/m^2) 93%, dichloromethane 91%, toluene 75%, benzene 81% and styrene 49%[3]. Thus, there is a good correlation between surface free energy and recovery.

In a study by Mueller and Miller, dichloromethane and trichloroethylene were desorbed with trichlorotrifluoroethane, chloroform and *o*-dichlorobenzene[16]. The first-mentioned solvent was, as could be expected (γ^d = 18.2 mJ/m^2), not efficient (recoveries 67% and 13%). Chloroform (γ = 27.1 mJ/m^2) was capable of quantitatively recovering dichloromethane, although lower recoveries were obtained for trichloroethylene. *o*-Dichlorobenzene yielded recoveries of *ca.* 90%, but the short equilibration time was probably not sufficient.

Recoveries for polar compounds (1 μl on 100 mg of carbon) as a function of ΔE_{DA} are given in Fig. 2. Although there is a fair amount of scatter, as could be expected, a definite relationship is established ($r = -0.87$ for both carbons). It should also be pointed out that for practical reasons, the recoveries were not obtained with equimolar amounts of adsorbate. For most compounds, 2-ethoxyethanol being an exception, only minor changes in the figure are expected if results for equimolar amounts are compared. Generally adsorption is stronger on the Merck carbon; this could be attributed to the higher oxygen content on this carbon (Table III) and to the fact that this carbon contains more acidic surface oxides[5]. In spite of these facts the same pattern is followed on both carbons. Some peculiarities could be stressed: dioxane is rather strongly adsorbed on Merck in contrast to SKC, and the difference between the alcohols and 2-ethoxyethanol is more pronounced on SKC.

If electron donors and acceptors are adsorbed on the same sites it should follow that compounds with high ΔE_{DA} values should be effective displacing agents. To put the matter to test, six solvents with different ΔE_{DA} values were prepared as 1% and 5% (by volume) mixtures in CS_2. Then 0.2–10 μl of butanol, 2-ethoxyethanol, acetone, *p*-dioxane and propyl acetate were added to the carbon and later desorbed with these mixtures. The test series on Merck was of more limited scope and comprised only butanol and 2-ethoxyethanol. The results (1 μl on 100 mg of carbon) are shown in Figs. 3 and 4. No values for dioxane are presented, but the results agreed very well with those for propyl acetate.

The efficiency of a desorbing agent is clearly related to its ΔE_{DA} value, although not as a simple function. As seen in Fig. 3 there is a slight "dip" for DMF, and in

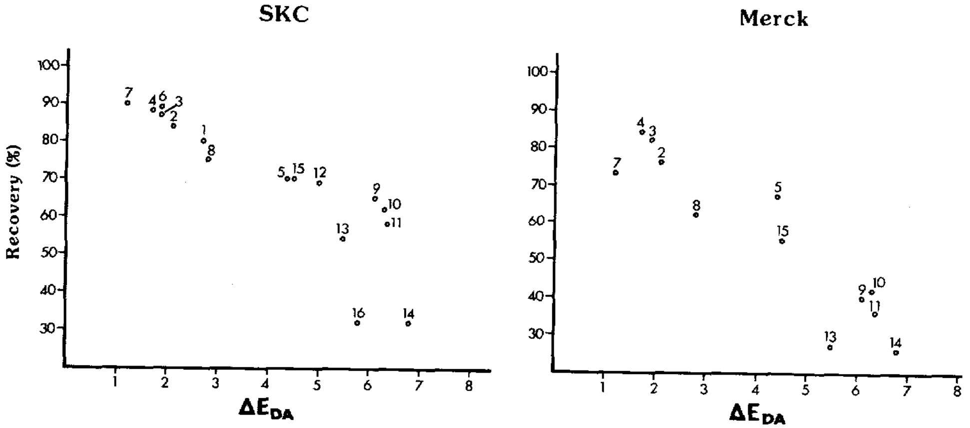

Fig. 2. Recovery as a function of ΔE_{DA} (see Table II) for a number of polar compounds (1 μl added to 100 mg of carbon) desorbed with carbon disulphide. For key see Table II.

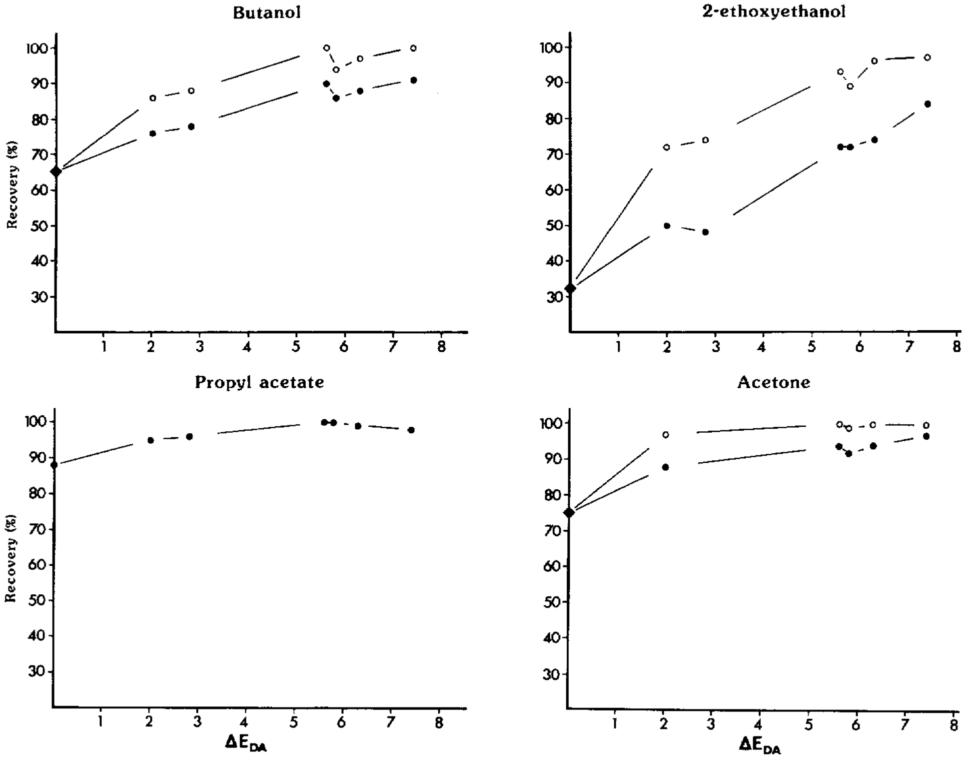

Fig. 3. Recoveries for 1 μl of butanol, 2-ethoxyethanol, acetone and propyl acetate on SKC carbon using 1% (●) and 5% (○) mixtures in carbon disulphide of the following solvents with ΔE_{DA} values given in Table II: methyl acetate, acetone, 2-butoxyethanol, dimethylformamide, ethanol and diethylene glycol monoethyl ether.

Fig. 4 a peak for ethanol (butanol) and DMF (2-ethoxyethanol). Surface heterogeneity and variations in donor–acceptor strength on the two carbons are possible explanations. Acetone (5%) gives about the same recoveries for butanol and 2-ethoxyethanol as 1% of the compounds with higher ΔE_{DA} values. For a solvent with a low ΔE_{DA} value the percentage added has to be increased in order to remove strongly retained adsorbates. Thus 50% acetone in CS_2 yields recoveries of 100% for butanol and 94% for ethyl cellosolve, whereas 50% dioxane is capable of raising recoveries for butanol to only 85%. Given a certain desorbing mixture, recoveries are related to the ΔE_{DA} values of the adsorbate. A strongly adsorbed compound, such as 2-ethoxyethanol, requires a higher ΔE_{DA} value/concentration of an addition compared with acetone for example.

It must be noted that the desorption of polar compounds requires a desorbing agent with sufficiently strong dispersive interactions with the hydrophobic surface, because otherwise the polar compounds would be adsorbed on this part of the surface. Ethanol or methanol would therefore not work well if used as desorbing agents by themselves.

The demonstrated relationships between recovery and surface free en-

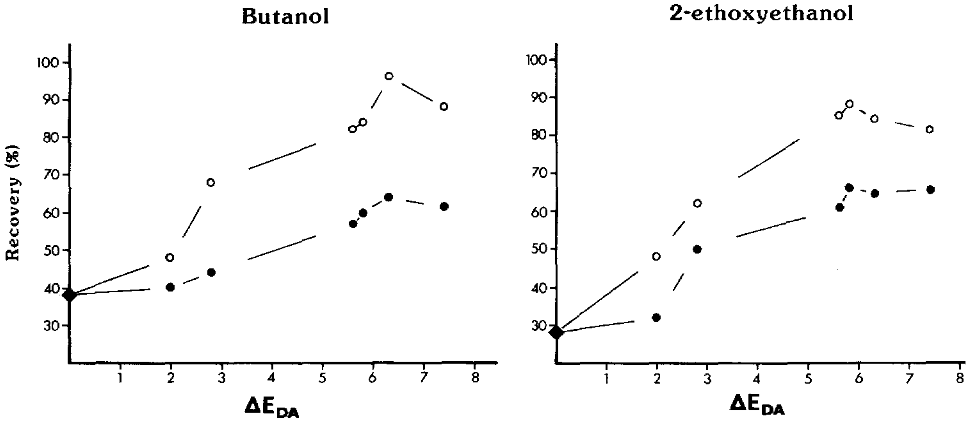

Fig. 4. Recoveries for 1 μl of butanol and 2-ethoxyethanol on Merck carbon using 1% (●) and 5% (○) mixtures in carbon disulphide of the following solvents with ΔE_{DA} values given in Table II: methyl acetate, acetone, 2-butoxyethanol, dimethylformamide, ethanol and diethylene glycol monoethyl ether.

ergy/ΔE_{DA} are rather approximate and cannot be used to predict recoveries for a certain desorbing system. The preferred uses of these relationships are to single out difficult compounds that could be used as touchstones for a desorbing agent, and to select suitable candidates for desorbing mixtures. This approach should increase experimental efficiency by limiting the number of experiments needed as new carbons and desorbing mixtures are tested.

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