Adsorption Isotherms of Sesame Oil in a Concentrated Miscella System

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The adsorption of peroxides, unsaturated carbonyls, free fatty acids and carotenoids from unrefined sesame oil on vegetable carbon (5%, w/w) in a miscella system was studied. Three different solvent conditions (hexane/ethanol, 100:0, 95:5 and 75:25, vol/vol), combined in a factorial design with five levels of solvent (0, 10, 20, 30 and 40%, w/w), were used to develop the miscella. Equilibrium adsorption was not achieved during the 100 min of adsorption, mainly because the oil components were involved in oxidation reactions during the adsorption process. However, for a given solvent concentration, adsorption of the oil components showed a significant linear regression on their respective initial concentration in the miscella (C_i). Peroxides and carbonyls showed, at all solvent levels investigated, an affinity for the carbon more independent of their C_i than free fatty acids and carotenoids. In general, at the same C_i, a higher adsorption was achieved as solvent concentration increased. The results indicated that free fatty acid adsorption may depend on competitive adsorption based on molecule hydrophobicity. However, in spite of the higher hydrophobicity of carotenoids, compared with free fatty acids, they might not be competing for the same adsorbing sites. Ethanol showed a prooxidant effect that increased peroxide production during adsorption but did not affect the reaction involved in carbonyl production.

KEY WORDS: Activated carbon, adsorption isotherms, carbonyl, carotenoid, concentrated miscella, covariance analysis, fatty acids, partition coefficient, peroxide, sesame oil.

Crude vegetable oils are generally processed by degumming, alkali refining, bleaching and deodorizing to obtain an odorless, bland and stable oil. The effects of different processing stages on chemical and physical properties of several crude vegetable oils have been studied (1,2). The bleaching process, mainly designed to perform pigment removal, is the only adsorption process in vegetable oil refining (3,4).

Adsorption involves separation of a substance (*i.e.,* adsorbate) from one phase, accompanied by its accumulation or concentration on the surface of the adsorbing phase (*i.e.,* adsorbent). An equilibrium between the adsorbate concentration in each phase is defined for each combination of specific adsorbate, adsorbent and conditions of temperature, pH and ionic strength (5). The study of selective adsorption of undesirable components from unrefined oils could lead to increased efficiency of refining methods, a better understanding of bleaching procedures, as well as a better control of the physicochemical/functional properties of refined oils (6,7). Investigations regarding the adsorption effect on the different oil components that should be eliminated during oil refining are needed.

Studies on adsorption of oil components have investigated the effectiveness of different adsorbents to eliminate pigments, as well as the fundamental properties of the adsorbent that are responsible for pigment adsorption efficiency (3,8–12). Thus, it has been shown that carotenoids and pheophytin adsorption from soybean oil can be described by a simple model that includes adsorbent surface acidity and pore volume (3). Additionally, it has been suggested that competitive adsorption based on the polarity of the oil components might play an important role in the binding of lutein, a soybean oil carotenoid, by silicic acid (8) and silica derived from rice hull ashes (10).

In other studies with dilute miscellas (*i.e.,* <40% oil in solvent), Freundlich-type isotherms have been observed for different components of vegetable oils (6,8,10–13). These investigations might be useful in the control of bleaching operations where the viscosity effect on adsorption is negligible (*i.e.,* continuous-flow oil refining) (11). However, studies in miscella systems at higher oil concentrations, where the viscosity effect on adsorption might be considerable, are lacking. Oil extraction processes that use up to 40% less solvent than conventional processes have been developed. The use of concentrated miscellas in adsorption processes would decrease the cost of energy necessary for solvent evaporation.

The purpose of this study was to evaluate the adsorption pattern of several oil components with vegetable carbon as adsorbent and unrefined sesame oil in a concentrated miscella system. The adsorption conditions investigated include three different proportions of hexane/ethanol (100:0, 95:5 and 75:25, vol/vol) combined in a factorial design with five percentages of solvent (0, 10, 20, 30 and 40%, w/w). Under such conditions, the miscella viscosity should affect the adsorption performance, and therefore its effect was considered. Although activated carbon is one of the most used adsorbents in the industry, there is little information regarding its adsorption behavior in vegetable oils. Previous research has shown that peroxide and carotenoid adsorption on vegetable carbon might be described through regression models (4).

EXPERIMENTAL PROCEDURES

Adsorption system. Unrefined sesame oil was obtained from a local industry (Aceitera San Juan, Salvatierra, Gto., México). The oil was distributed in amber bottles (250 mL) and stored at 4°C under nitrogen atmosphere.

To eliminate secondary effects, such as particle size or moisture content, the same lot of activated carbon (Clarificantes Mexicanos, Santa Clara, Estado de México, Mexico) was utilized in all the experiments after sieving (U.S. standard, −14/+24), washing (mixing for 24 h in 2 vol of deionized water), filtering and drying (100°C for 24 h).

Adsorption variables investigated included three different solvent conditions (hexane/ethanol, 100:0, 95:5 and 75:25, vol/vol) combined in a completely balanced factorial design (14) with five levels of solvent (0, 10, 20, 30 and

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40%, w/w). The treatments were distributed among the oil bottles in a randomized design with two replicates.

Adsorption was performed at 50°C in a closed-system mixer consisting of a three-neck distilling flask (500 mL) assembled with a 300-mL Graham condenser (with running tap water at ≈20°C), a thermometer and a glass mixing shaft with Teflon mixing blades (3″). The mixing shaft was coupled to a variable-speed mixer (Model GT 21-18; G.K. Heller Corp., Bellerose, NY). Temperature control was achieved with a water bath (Büchi 461; Büchi Laboratoriums-Technik AG, Flawil, Switzerland).

All adsorption experiments were carried out as follows. The oil, hexane and ethanol were combined in the proper ratio, and this miscella was analyzed for free fatty acids (15), peroxides (15), carotenoids (16) and unsaturated carbonyls (17). The miscella (380 g) was weighed into the distilling flask, the mixing system was assembled and the miscella was mixed gently until it reached a temperature of 50°C (≈5–10 min). Without stopping the mixing, the thermometer was momentarily removed from the flask, 20 g of adsorbent (5%, w/w) was added directly to the miscella, and the mixing speed was increased to just avoid vortex formation. Preliminary experiments with miscellas (*e.g.*, 30 and 40% hexane in sesame oil) showed that after 90 min of adsorption with 5% (w/w) carbon, the concentration of the oil components did not change significantly. Therefore, after 100 min of constant agitation with the adsorbent, the mixture was centrifuged (5000 rpm/15 min), and the miscellar phase was again analyzed to determine the adsorbed amount per mass of carbon for each of the oil components. Solvent evaporation occurred during centrifugation; therefore, appropriate corrections were made in the calculations.

Viscosity measurements. The apparent viscosity (in centipoises) of the miscellas was determined at 30°C with a Brookfield DV-II viscosimeter (Brookfield, Co., Stoughton, MA) at 12 rpm with spindle #1 of the LV type. A 600-mL beaker containing 400 mL of miscella was used in all determinations. Temperature control was achieved with a water bath.

Statistical analysis. The statistical analysis included covariable methodology (18,19) combined with factorial analysis. For each of the oil components investigated (*i.e.*, adsorbates), the analysis utilized the respective initial adsorbate concentration in the miscella as covariable. After testing the significance of the covariable ($P < 0.10$), the adsorption data were evaluated in a factorial design consisting of the five levels of solvent and three levels of hexane/ethanol ratio investigated. The effects of these variables and their interactions were evaluated in single degrees of freedom; when the effect was not significant ($P > 0.10$), its associated sum of squares was pooled into the error term. The statistical analysis was performed with the STATISTICA program (StatSoft, Tulsa, OK) (20).

RESULTS

The descriptive statistics of the variables investigated are shown in Table 1. Assuming that adsorption equilibrium was achieved during the 100 min of adsorption, the data were expressed according to the isotherm models most utilized (*i.e.*, Langmuir, BET, Freundlich). However, appropriate fits were not obtained. As an example, a Freundlich isotherm for free fatty acids is shown in Figure 1.

During adsorption, in addition to the adsorbate-ad-

TABLE 1

Descriptive Statistics of the Variables Investigated

	n[a]	Mean	Standard deviation	Minimum	Maximum
Solvent (%)	5	20.00	15.81	0.00	40.00
Ethanol (%) (in the solvent)	3	10.00	13.22	0.00	25.00
Initial:					
Peroxides (μEq/g miscella)	30	2.63	0.52	1.62	3.67
Free fatty acids (mg[b]/g miscella)	30	3.83	0.65	2.69	5.20
Unsat. carbonyls (μMole/g miscella)	30	1.38	0.41	0.26	1.97
Carotenoids (μg/g miscella)	30	3.17	0.64	2.24	4.67
Adsorbed:					
Peroxides (μEq/g carbon)	30	18.07	8.14	−1.44[c]	34.10
Free fatty acids (mg[b]/g carbon)	30	17.63	9.77	−8.37[c]	45.47
Unsat. carbonyls (μMole/g carbon)	30	3.00	4.61	−9.53[c]	14.91
Carotenoids (μg/g carbon)	30	21.63	7.09	5.82	33.72

[a]Number of replicates. Each replicate done by duplicate. [b]As oleic acid. [c]A negative figure indicates that production of that substance rather than adsorption occurred during the process. Unsat., unsaturated.

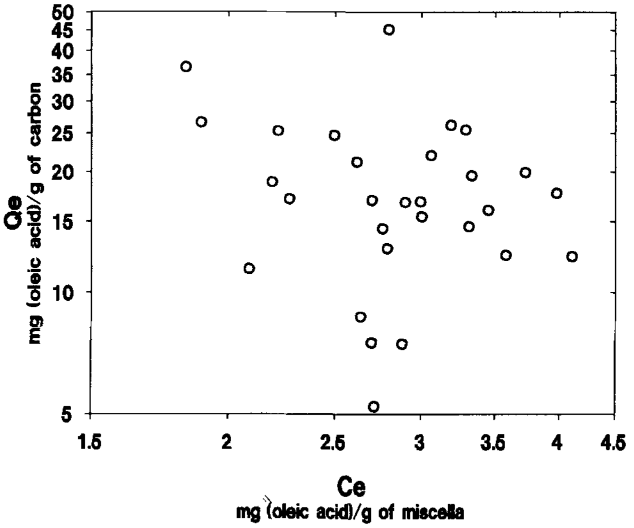

FIG. 1. Freundlich isotherm for free fatty acids. Qe is the adsorbed amount after 100 min of adsorption with 5% carbon at 50°C. Ce is the concentration of free fatty acids remaining in the miscella after adsorption.

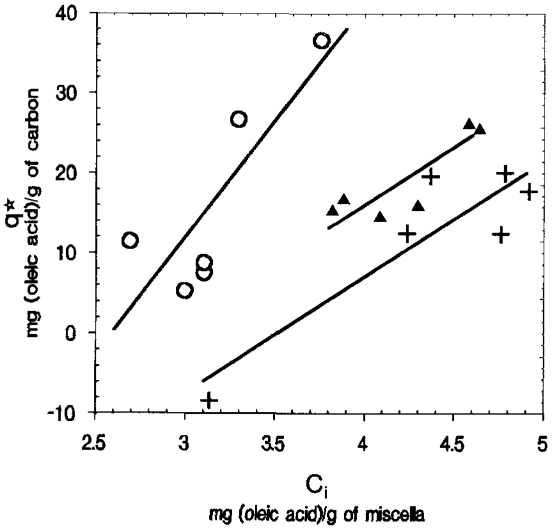

FIG. 2. Regression lines at three solvent levels (0, 10 and 40%) for pseudo-equilibrium adsorption (q*) of free fatty acids on the respective initial free fatty acids concentration in the miscellar phase (C_i). The regression coefficients were at least 0.854. Solvent: +, 0%; ▲, 10%; ○, 40%.

sorbent interaction, oxidation reactions are also taking place in the oil phase as well as on the adsorbent surface (1,2,4,7). Thus, after the adsorption process, the concentrations of the oil components involved in the oxidation reactions are the result of a balance between the events of adsorption and oxidation (4,7,21). The predominance of any of these events depends upon the interactions among several variables, mainly the adsorbent concentration, the stage of oxidation of the oil, the susceptibility of the fatty acids to react with oxygen and the concentration of compounds with antioxidant (*i.e.*, carotenoids) or prooxidant (*i.e.*, free fatty acids) activity (7). The negative figures for peroxides, unsaturated carbonyls and free fatty acids shown in Table 1 indicate when oxidation reactions predominated over the process of adsorption (*i.e.*, after adsorption, the result was an increase in the concentration of the compound in the oil phase); these circumstances were always found at the lowest levels of solvent in the miscella (*e.g.*, 0 and 10%). At 30 and 40% of solvent in the miscella, the process of adsorption predominated over oxidation reactions, producing a decrease in the concentration of the adsorbate in the oil phase and reaching an apparent adsorption equilibrium. Therefore, for these oil components, the thermodynamic concept of equilibrium adsorption was not appropriate, mainly because the equilibrium conditions (*e.g.*, equilibrium concentrations) were not just dependent upon the factors involved in the adsorption phenomenon. The adsorbed amount obtained under such conditions was defined as the pseudo-equilibrium adsorption (q* = the adsorbed amount after 100 min). Several studies with vegetable oils have observed Freundlich-type isotherms for different oil components (6,10,13). However, the adsorption systems consisted of oil concentrations lower (<40%) than the ones used in the present study, as well as shorter adsorption times. In such systems, the effect of the oxidation reactions on the achievement of equilibrium adsorption should be limited, and equilibrium conditions might be assumed.

When the q* for each of the oil components (*i.e.*, the response variable) was plotted against their respective initial concentrations in the miscella (C_i), it was observed that, at each of the solvent levels investigated, a significant part of the variability of the oil components' adsorption was explained by the linear regression of q* on C_i. As an example, Figure 2 shows this relationship for free fatty acids at three different solvent levels. This relationship allowed the possibility of describing the adsorption for each of the oil components in terms of a single partition coefficient, K_p (*i.e.*, the slope of the positive linear regression of q* on C_i). For a given oil component, K_p was statistically the same ($P > 0.25$) for the different solvent levels utilized. These results showed that, after considering the effect of the solvent, the main driving force to achieve q* was C_i. Therefore, if a treatment was randomly assigned to a larger (or smaller) initial adsorbate concentration in the miscella, then a significant part of the adsorption was due to the higher (or lower) value of C_i. Consequently, to assess the actual effect of the variables investigated, a covariance analysis was performed. This statistical methodology combines the features of analysis of variance and regression (18,19) and, therefore, allowed the mathematic adjusting of q* obtained under different solvent conditions to a common value of C_i. Through this technique, the variation in q* due to differences in the initial adsorbate concentration (*e.g.*, the covariable) was removed.

The equations derived from the covariance analysis and utilized in the mathematic adjusting of the adsorbed amount are shown in Table 2. For each of the oil components investigated, the common value utilized to adjust q* was the respective mean of the initial adsorbate concentration in the miscella (values shown in Table 1 under the "Mean" column).

With exception of the unsaturated carbonyls, just the initial adsorbate concentration in the miscellar phase was

TABLE 2

Equations[a] Derived from the Covariance Analysis and Utilized in the Mathematical Adjustment of the Pseudo-Equilibrium Adsorption for the Different Oil Components Investigated

$$P_{\text{adjusted}} = P_{\text{unadjusted}} - 7.46\ (P_{\text{initial}} - 2.63)$$

$$FFA_{\text{adjusted}} = FFA_{\text{unadjusted}} - 14.45\ (FFA_{\text{ininital}} - 3.83)$$

$$UNC_{\text{adjusted}} = UNC_{\text{unadjusted}} - 10.40\ (UNC_{\text{initial}} - 1.38) - 4.0\ (P_{\text{initial}} - 2.63)$$

$$CAR_{\text{adjusted}} = CAR_{\text{unadjusted}} - 13.01\ (CAR_{\text{initial}} - 3.17)$$

[a] Legend: P, peroxide; FFA, free fatty acids; UNC, unsaturated carbonyls; CAR, carotenoids. The subscripts "unadjusted" and "adjusted" indicate the pseudo-equilibrium adsorption unadjusted or adjusted for the effect of the initial adsorbate concentration. The subscript "initial" indicates the initial concentration of the oil component in the miscella. The units are the same as indicated in Table 1.

significant as covariable (Table 2), *i.e.*, after considering the effect of the solvent, the driving force for adsorption, C_i, had a linear effect on q*. For the unsaturated carbonyls, an additional amount of variability in carbonyl adsorption was explained by the peroxide concentration in the miscella (Table 2). Carbonyls originate from a bimolecular peroxide decomposition, and the magnitude of the rate constant for this reaction has a linear dependence on the initial peroxide concentration (21). Consequently, carbonyl concentration, and therefore carbonyl adsorption, was an indirect function of the initial peroxide concentration in the system. The correlation coefficients for the effect of the covariable (*i.e.*, C_1) on the q* of peroxides (data not shown), free fatty acids (Fig. 2) and carotenoids (data not shown) were at least 0.818, 0.854 and 0.754, respectively ($P < 0.001$). For the unsaturated carbonyls, the multiple regression coefficient was 0.819 ($P < 0.001$) (data not shown).

The effects of the solvent concentration and the hexane/ethanol ratio on oil components adsorption were evaluated in a factorial analysis by using as the response variable the q* values obtained with the equations shown in Table 2. For each of the oil components investigated, the experimental error associated with the q* achieved under constant initial adsorbate concentration (*i.e.*, adjusted adsorption) was at least three times smaller than the one associated with the unadjusted adsorption. Thus, more precise comparisons among the treatments investigated were obtained. Figures 3–6 show just the significant effects associated with the treatments evaluated.

In general, as solvent concentration increased in the system, higher adsorption was achieved. The apparent viscosity of the miscella showed an inverse relationship with the solvent concentration in the miscella according to the following equation ($R^2 = 0.98$, $P < 0.0001$):

$$1/\cap_{ap} = 10.4014 \times 10^{-3} + (4.283 \times 10^{-3}) \cdot (\%\ \text{solvent}) \quad [1]$$

where $\cap_{ap}$ is the miscella apparent viscosity (in cps) determined at 30°C and a shear rate of 12 rpm, and % solvent represents the percentage (w/w) of hexane in the miscella. This decrease in viscosity reached a plateau at 30% of solvent concentration in the miscella. As the viscosity decreased in the system, the diffusion of the adsorbates toward the surface and through the pores of the carbon was promoted, thus increasing the access of the molecules to the adsorbing sites.

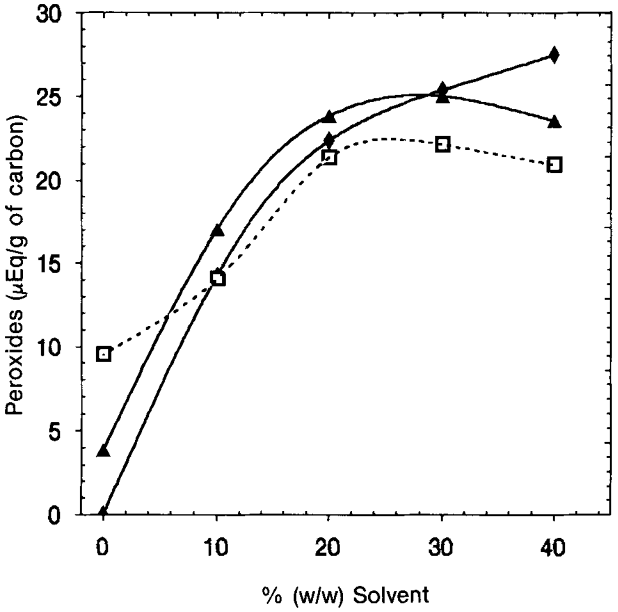

FIG. 3. Peroxide isotherms obtained with 5% carbon at 50°C in unrefined sesame oil miscellas. The proportion of ethanol (0, 5 and 25%) in the solvent was different for each isotherm. The standard error of the estimates was 2.85 µEq/g carbon. Ethanol: -□-, 25%; -▲-, 5%; -◆-, 0%.

For oil components, of which the concentration depended on oxidation reactions occurring during the adsorption process (*e.g.*, peroxides and carbonyls), the additional effect of oil dilution should also be considered. Previous results (7) have suggested that peroxide behavior during the adsorption process is the result of a balance between the events of peroxide formation, peroxide decomposition and peroxide adsorption. Thus, as the solvent diluted the compounds involved in the reactions of oxidation, peroxide production and decomposition (*i.e.*, carbonyl production) were limited. As a result, as solvent percentage increased, peroxide and carbonyl adsorption predominated over peroxide and carbonyl production (Figs.

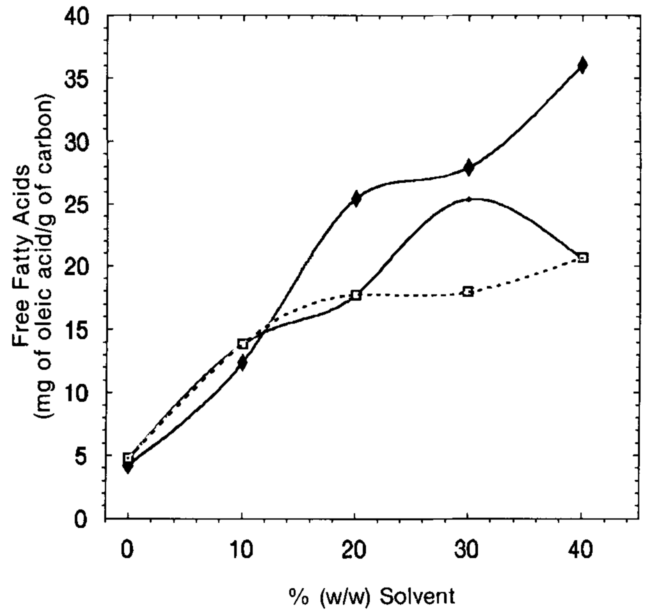

FIG. 4. Free fatty acid isotherms obtained with 5% carbon at 50°C in unrefined sesame oil miscellas. The proportion of ethanol (0, 5 and 25%) in the solvent was different for each isotherm. The standard error of the estimates was 3.83 mg/g carbon. Ethanol: -□-, 25%; -●-, 5%; -◆-, 0%.

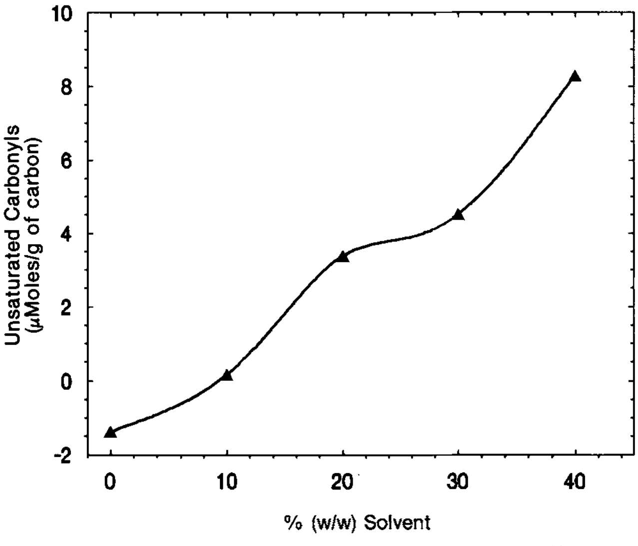

FIG. 5. Unsaturated carbonyl isotherms obtained with 5% carbon at 50°C in unrefined sesame oil miscellas. The proportion of ethanol (0, 5 and 25%) in the solvent was different for each isotherm. The standard error of the estimates was 2.81 μMole/g carbon.

3 and 5). In the specific case of peroxides, at solvent levels of 30 and 40%, the presence of ethanol significantly decreased peroxide adsorption. This effect was more pronounced at the highest proportion of ethanol in the solvent (Fig. 3). This reduction in peroxide adsorption was associated with a prooxidant effect on ethanol. Yanishlieva and Kortenska (22) reported that, when C14 to C20 alcohols were added to acylglycerides, peroxide formation was accelerated. Apparently, this prooxidant effect depends on the type and degree of unsaturation of the lipid system, its peroxide content and the chainlength and concentration of the alcohol (22). Thus, although in general peroxide adsorption predominated over peroxide formation, its prevalence was less significant as the effective ethanol concentration increased in the miscella. Carbonyl adsorption was not affected by the presence of ethanol (Fig. 5), probably because ethanol did not affect the reactions involved in peroxide decomposition.

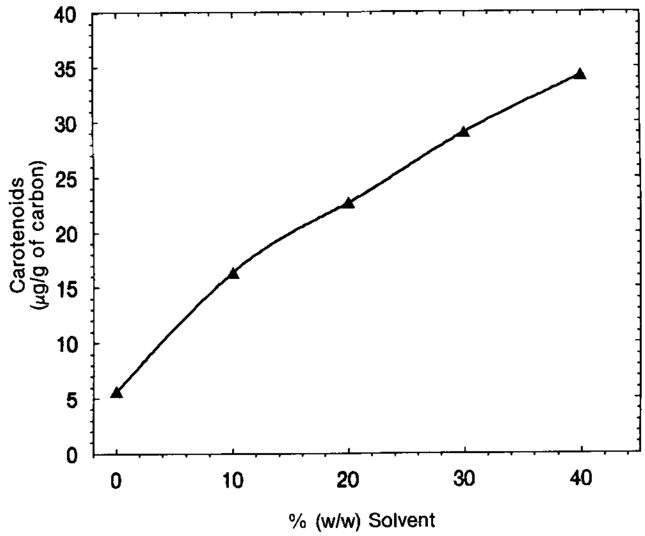

FIG. 6. Carotenoid isotherms obtained with 5% carbon at 50°C in unrefined sesame oil miscellas. The proportion of ethanol (0, 5 and 25%) in the solvent was different for each isotherm. The standard error of the estimates was 4.34 μg/g carbon.

In both free fatty acid and carotenoid adsorption (Figs. 4 and 6), q* increased as a linear function of solvent concentration in the system ($P < 0.001$). Free fatty acid adsorption decreased significantly when ethanol was present in the solvent ($P < 0.05$). However, no difference in adsorption was observed between the two proportions of ethanol investigated ($P > 0.25$). On the other side, carotenoid adsorption was not significantly affected by the presence of ethanol ($P > 0.35$). Studies made with a silica-type adsorbent indicated that the addition of 1% isopropanol to soybean oil-hexane miscellas promoted free fatty acid adsorption while it decreased adsorption of the carotenoid lutein (10,13). The discrepancy observed with these results might be explained by considering that ethanol and isopropanol have different conformational structures. Furthermore, silica and carbon have different mechanisms to accomplish adsorption. Whereas hydrogen bond formation between the adsorbate and the silanol groups plays a major role in adsorption on silica (23), hydrophobic interactions are the main forces involved in adsorption on carbon (24). Fatty acids from vegetable oils (*i.e.*, mainly C12 to C18) are more hydrophobic than ethanol, because of their longer carbon chainlength as well as the less polar character of the -COOH functional group in contrast with the -OH group present in ethanol. However, ethanol molecules should diffuse more efficiently through the carbon pores than fatty acids, and therefore, ethanol should reach the adsorption sites more rapidly. Nevertheless, and in spite of the high ethanol concentration utilized in the solvent miscella, fatty acid adsorption was not completely eliminated. Apparently, in the presence of ethanol, fatty acid adsorption depended upon a phenomenon of competitive adsorption, probably based on molecule hydrophobicity. In contrast, aromatic compounds, such as carotenoids, have a high adsorption affinity in carbon-type adsorbents, due not only to their high hydrophobicity but also because of their π-bonding ability with the ring structures present along the carbon surface (25). However, carotenoids might be in a colloidal state in the miscella, thus needing pore diameters in the 50–100 Å region to be adsorbed (3,26). Unpublished results (Toro-Vazquez, J.F.) indicated that vegetal carbon, similar to the one used in this investigation, had at most 12–13% of the total surface area ($\approx$1231 m^2/g of carbon) distributed in pores with diameter > 50 Å (surface area and pore volume were determined by the BET method with nitrogen as adsorbate). These results suggested that carotenoids did not compete for the same adsorbing sites to which ethanol and fatty acids molecules were adsorbed. Probably, most of the pores of the carbon utilized had diameters readily accessible for ethanol and free fatty acid molecules but not equally accessible for carotenoids.

For all oil components, higher adsorption was obtained in the absence of ethanol and at 40% solvent concentration. However, additional factors should be considered. The K_p, obtained from the slope of the equations derived from the covariance analysis (Table 2), indicated the relative sensitivity of the oil component q* to its initial concentration in the miscella. Thus, peroxides and unsaturated carbonyls (with K_ps of 7.46 and 10.40, respectively) showed, at all the solvent levels investigated, an affinity for the carbon more independent of their initial concentrations than free fatty acids and carotenoids (with K_ps of 14.45 and 13.01, respectively). From a practical standpoint and without consideration of the solvent level, the removal of peroxides and unsaturated carbonyls might be achieved either in a batch or a flow-through system (27). However, under the conditions investigated, a batch treatment for free fatty acid and carotenoid removal might not be suitable, mainly because their adsorption capacities showed a high dependence on their initial concentration. In this case, some type of countercurrent system may be necessary to improve efficiency (27).

Successful utilization of concentrated miscellas in an adsorption process for oil refining would depend upon the adsorption efficiency obtained in terms of the removal of undesirable oil components as compared to that achieved with diluted miscellas. Additionally, the costs associated with the energy required for solvent evaporation should be considered.

Although the mean of the initial adsorbate concentration was the common value used for mathematical adjusting of the q* of the different oil components, the covariate equations (Table 2) might use as common adjusting value any initial adsorbate concentration within the intervals shown in Table 1. Although a different magnitude of the equilibrium adsorption would be obtained, the conclusions of the analysis should be identical, mainly because for a given oil component, K_p was statistically the same ($P > 0.25$) for the different solvent levels utilized. Ongoing research is now considering a wider interval of initial adsorbate concentrations to validate and improve the isotherm model.

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