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The Model of Volatile Hydrocarbons Removal from Their Emulsions in the Flotation Process

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Abstract

A mathematical model for the removal of volatile hydrocarbons from their O/W-type emulsions is presented. Two simultaneous processes are discussed: the mass transport of the dissolved hydrocarbon molecules from water into the bubble as a consequence of evaporation, and the interception of hydrocarbon droplets by a rising bubble based on a hydrodynamic model. A mesitylene (1,3,5-trimethylbenzene) results are used to test a mathematical model for both processes. Fairly good agreement is obtained.

INTRODUCTION

Air flotation is widely used for the separation of oily components from their emulsions (*1–7*). The oil droplets in O/W-type emulsions are usually very fine (a few μm in diameter). Many authors have discussed the model of fine particle flotation (*8–15*). It has been stated that the flotation depends on the collision efficiency (E_c) of the particle and the bubble. It is generally stated that collision occurs on the basis of the inertional impact, by interception due to hydrodynamic forces, Brownian, and thermal diffusion, or by electrical interactions (*13*, *15*, *16*). The forces which influence the resulting collision efficiency depend on the bubble and particle diameters as well as on the hydrodynamic parameters of the fluid flow. The collision efficiency is dependent on the Stokes number, the

Reynolds number, and the terminal settling velocity of the particle (Stk, Re, u_p) (*9–12*).

The collision efficiency for coarse particles (characterized by a Stokes number greater than ~1.0) depends strongly upon inertial forces. For the case of fine particles (Stokes number less than ~0.1) and small bubbles, the inertia of the particles may be neglected when computing their trajectories relative to the approaching bubbles (*11*).

According to Reay's model, as the Stokes number becomes very small, particles will attach to bubbles as a result of hydrodynamic forces alone (for small particles and large bubbles). In effect, the vacuum induced in the wake of a rising bubble can trap particles in spite of interfacial repulsion due to electrostatic effects. This model suggests that for the flotation of oily water, another mechanism (hydrodynamic capture) in addition to collision may contribute to the overall removal rate.

Sylwester showed (*3*) that hydrodynamic capture is an operative mechanism for oil drop flotation if the bubbles have diameters of 0.2–0.7 mm.

It was found that the collision efficiency is proportional to the d_p/d_b value (*6*, *13*, *15*). According to Reay's empirical relation, the collision efficiency may be calculated by $E_c = \alpha(d_p/d_b)^\beta$, where α and β depend mainly on the kind of particles and the fluid.

Sato's experiments on oil particle flotation confirmed the theoretical model based on interception, which was used for the calculation of the collision efficiency (*6*).

This study was initiated to gain an understanding of the flotation effect differences observed for different hydrocarbons. The removal of different hydrocarbons from their emulsions by flotation was investigated earlier (*7*, *17–19*). It was found that the removal of aromatic hydrocarbons (mesitylene, cumene, pseudocumene) is much better (*7*, *18*) than the removal of aliphatic hexadecane (*17*). The hydrocarbon mass balance calculation shows that the hydrocarbon losses are about 0 to 20% in the case of hexadecane flotation. However, in aromatics flotation the losses can reach 95%. Those observations suggest different mechanisms of the removal process. It is assumed that hexadecane droplets are removed from the emulsion due to their capture by a bubble. For aromatic hydrocarbons the process is accompanied by evaporation into the bubble of the molecules dissolved in water (*20*).

The purpose of this study was to analyze the processes which take place in a batch-type flotation system during aeration of mesitylene emulsion.

CHARACTERISTICS OF THE SYSTEM

Consider the mesitylene emulsion. The drop size distribution of mesitylene droplets in emulsion was investigated by a microscopic method. It has been stated that droplets with a diameter of less than 4 μm exist in a prepared emulsion to the extent of 90–95% (*21*). The mean droplet diameter (so-called Sauter diameter) was calculated by

$$d_{32} = \frac{\sum n_i d_i^3}{\sum n_i d_i^2} \tag{1}$$

The terminal settling velocity of mesitylene droplets is about 2.9×10^{-5} and 1.8×10^{-4} cm/s for droplets of 2 and 5 μm diameter, respectively.

Flotation process carried out in the column described earlier (*7*) involve bubbles between 0.2 and 1.2 mm diameter. Such bubbles have terminal Reynolds numbers in water between 3 and 200. This is a flow range below Newton's law region and above Stokes' law region. The rise velocity of the bubble in this flow range may be calculated from the following equation (*22, 23*):

$$u = \frac{2g\rho r_b^2}{9\eta}\left[1 + \frac{1}{4}\left(\frac{\rho r_b u}{2\eta}\right)^{1/2} + \frac{0.34\rho r_b u}{12\eta}\right]^{-1} \tag{2}$$

The rise velocity calculated from this equation is ~1.7 and ~15.0 cm/s for bubbles of 0.2 and 1.2 mm diameter, respectively.

As may be seen, the rise velocity of bubbles is about 10^3 to 10^5 times greater than the settling velocity of mesitylene droplets, so the gravity settling of droplets may be neglected.

The mean bubble diameter was calculated according to

$$d_{30} = \sqrt[3]{\frac{\sum n_i d_i^3}{\sum n_i}} \tag{3}$$

(this is a mean arithmetical diameter) or according to Eq. (1).

The Stokes number calculated for bubbles of 0.2 to 4 mm diameter and droplets of 1 to 4 μm diameter was in 10^{-5} to 10^{-4} range; for droplets of 5–10 μm diameter, it was 10^{-4} to 10^{-3}.

The solubility of mesitylene in water as determined by Clayton (*24*) equals 57 mg/dm³. The mesitylene vapor pressure at 20°C equals 1.95 mmHg (*25*).

THEORY

Two simultaneous processes are discussed.

I. Droplets Capture by a Rising Bubble

On the basis of the calculated Stk values there is no possibility of an inertial impact of the droplet and the bubble. The critical value of Stk corresponding to the largest particle diameter which will not impact the bubble equals 0.1. In our system Stk $\ll$ 0.1 for droplets of the sizes found in the emulsion. Thus the inertial effects of droplets may be neglected, and the collision efficiency may be predicted by a simple trajectory theory based on interception. If Stk $\ll$ Stk$_{crit}$, then the particle may also be entrapped in the wake by a diffusion process.

The collision efficiency for a boundary layer flow may be calculated according to Sato (*6*):

$$E_c = 0.995\left[\frac{r_p}{r_b}\right]^2\sqrt{\mathrm{Re}} \tag{4}$$

The flotation effect may be expressed as the mesitylene concentration change in the emulsion. The concentration of emulsified component after a given time τ from the beginning of aeration may be calculated from

$$C = C_0 \exp\left[-\frac{3E_cV_g\tau}{r_bD}\right] \tag{5}$$

The hydrocarbon mass removed by the interception process (M_i) is therefore

$$M_i = V_sc_0\left[1 - \exp\left(-\frac{3E_cV_g\tau}{r_bD}\right)\right] \tag{6}$$

II. Solute Mass Transport from Water into the Bubble by Evaporation

This process is connected with the fact that mesitylene is slightly soluble in water. In addition, its vapor pressure is large enough for it to be considered a volatile compound. Solute transport by rising bubbles was examined by Wilson et al., especially in relation to the solvent sublation process (*23*, *26*, *27*). Thank to the volatility (even very low), a few hydrophobic substances (for example, naphtalene) were satisfactorily removed by aeration of their solutions (*27*).

The transport rate of mesitylene from solution into the interior of a bubble is proportional to the difference between the actual mass of hydrocarbon inside a bubble and the mass which would be in the vapor phase if the bubbles were at equilibrium with the surrounding solution. The rate of mass transfer to one bubble is

$$dm/dt = 4\pi r_b^2 k(\tfrac{4}{3}\pi r_b^3 K_{\mathrm{H}} C_{aq} - m)/\tfrac{4}{3}\pi r_b^3 \tag{7}$$

where m is the actual mass of mesitylene inside the bubble and t is the residence time of a bubble in the aqeuous layer. We can make the approximation that C_{aq} changes negligibly during the time required for the bubble to rise through the aqueous layer. Rearrangement and integration of Eq. (7) leads to Eq. (8) which allows calculation of the mass of mesitylene carried out of the water by one bubble:

$$m = \frac{4}{3}\pi r_b K_{\mathrm{H}} C_{aq}\left[1 - \exp\left(-\frac{3kh}{r_b u}\right)\right] \tag{8}$$

The change of solute mass transported into the vapor phase from the solution may be generally expressed as:

$$dM_e/d\tau = nm \tag{9}$$

After substitution:

$$\begin{aligned}\frac{dM_e}{d\tau} &= \frac{3V_g}{4\pi r_b^3}\frac{4}{3}\pi r_b^3 K_{\mathrm{H}} C_{aq}\left[1 - \exp\left(-\frac{3kh}{r_b u}\right)\right] \\ &= V_g K_{\mathrm{H}} C_{aq}\left[1 - \exp\left(-\frac{3kh}{r_b u}\right)\right] \end{aligned} \tag{10}$$

By applying Eq. (10) and because

$$\frac{dM_e}{d\tau} = -\frac{dC}{d\tau}V_s \tag{11}$$

the concentration change in the solution during the aeration time may be calculated:

$$C = C_{aq} \exp\left[-\frac{V_g K_H \tau}{V_s}\left(1 - \exp\left(-\frac{3kh}{r_b u}\right)\right)\right] \tag{12}$$

From Eqs. (11) and (12), the formula for calculating the total mass of solute carried out of the aqueous solution is found:

$$M_e = V_s C_{aq}\left\{1 - \exp\left[-\frac{V_g K_H}{V_s}\left(1 - \exp\left(-\frac{3kh}{r_b u}\right)\right)\right]\right\} \tag{13}$$

During the aeration of mesitylene emulsion, the concentration of soluble hydrocarbon diminishes according to Eq. (12). Mesitylene is slightly soluble in water, but vigorous homogenization during preparation of the emulsion and a long (about 12 h) storage before investigation assures obtaining a mesitylene concentration in water equal to its solubility. If we consider that the rate of mesitylene dissolving from emulsion droplets during flotation is very slow, and that only evaporation influences the changes of C values, then the total mass carried out of the water by evaporation may be calculated from Eq. (13).

However, if the rate of mesitylene molecules transfer from the emulsion droplets into the aqueous solution is so fast that the mass transfer coefficient for oil-water transfer of mesitylene is much more greater than the diffusion coefficient of mesitylene in water, then the concentration of mesitylene in the aqueous solution may be treated as a constant during the aeration process. In such cases the total mass of mesitylene carried out of the solution on the basis of evaporation may be calculated from

$$M_e = nm\tau \tag{14}$$

and, after substitution,

$$M_e = V_g C_{aq} K_H \tau\left[1 - \exp\left(-\frac{3kh}{r_b u}\right)\right] \tag{15}$$

Equation (15) may be used only until all droplets disappear (dissolved or captured by a bubble due to hydrodynamic forces). From that moment, Eq. (13) is valid.

THEORETICAL RESULTS

The collision efficiency dependence on bubble diameter for different sizes of mesitylene droplets are shown in Fig. 1. E_c values were calculated

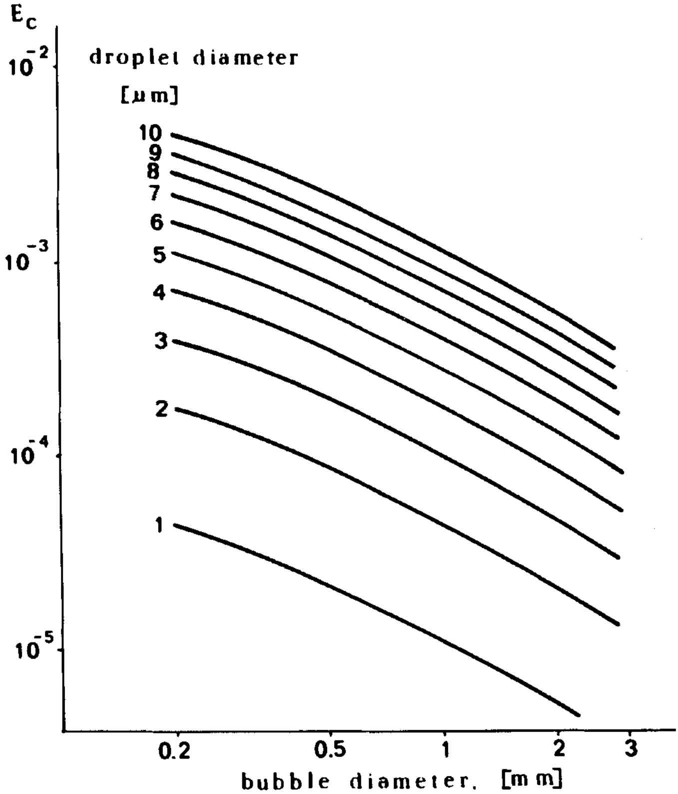

FIG. 1. Dependence of collision efficiency E_c on bubble diameter for different mesitylene droplet sizes.

accordingly to Eq. (4). As can be seen, the smaller the bubble and the larger the droplet, the greater the collision efficiency value, as would be expected. For droplets of 1 to 5 μm diameter and for bubbles of 0.6 mm (most often found in our system), the E_c value varies from 1.9×10^{-5} to 4.7×10^{-4}, respectively. Figure 2 presents the theoretical flotation efficiency of mesitylene (expressed as its concentration in raffinate) from its emulsion for different emulsion droplet diameters. The concentration in raffinate was calculated by Eq. (5) with the assumption that the collection efficiency equals the collision efficiency. Note that fast removal of mesitylene droplets occurs when they are larger than ~5 μm diameter; the smaller the bubble diameter, the better the removal.

Figures 3, 4, and 5 present the evaporation process effect calculated on the basis of Eq. (12). The mass transfer rate coefficient values were chosen on the basis of Lionel's work (*26*) and were 5×10^{-4} to 5×10^{-2} cm/s. It is seen that the evaporation effect depends on the bubble diameter and the mass transfer rate coefficient. It was found that a mass transport

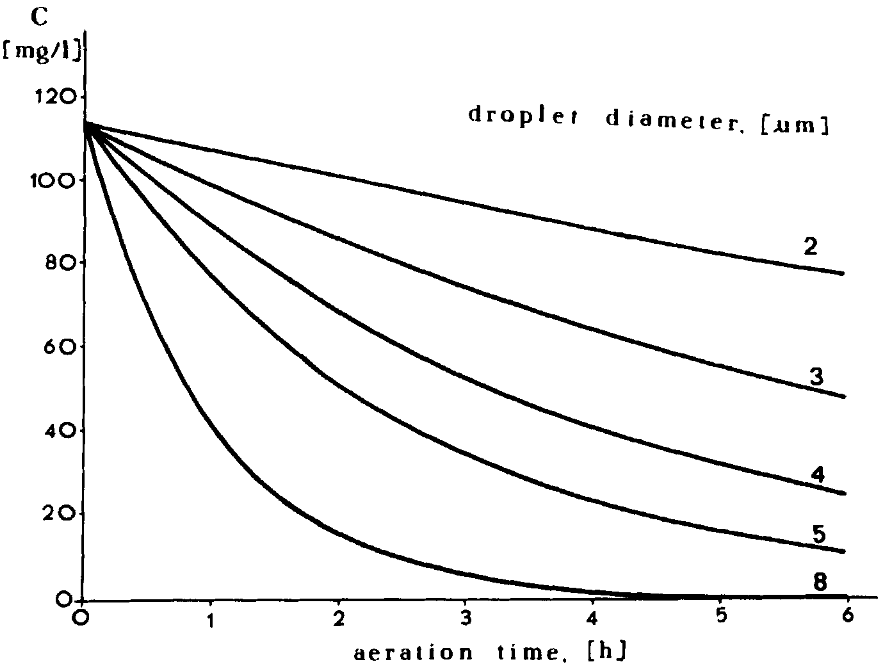

FIG. 2. The effect of droplet diameter on the removal of mesitylene droplets by interception mechanisms. Bubble diameter d_b = 0.6 mm, air flow rate V_g = 1.508 cm^3/s.

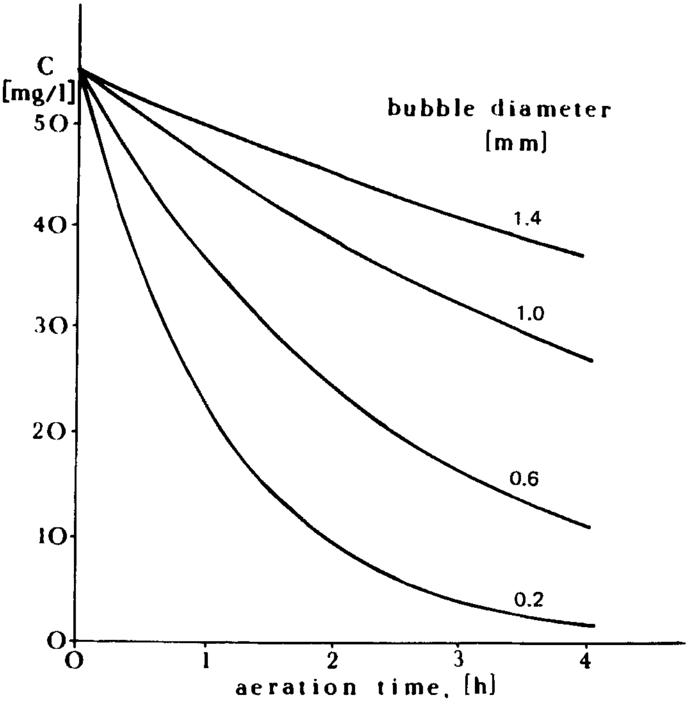

FIG. 3. Removal of mesitylene from the solution into the bubble due to the evaporation process; the effect of bubble diameter. $K_H = 0.265$, $k = 0.0005$ cm/s.

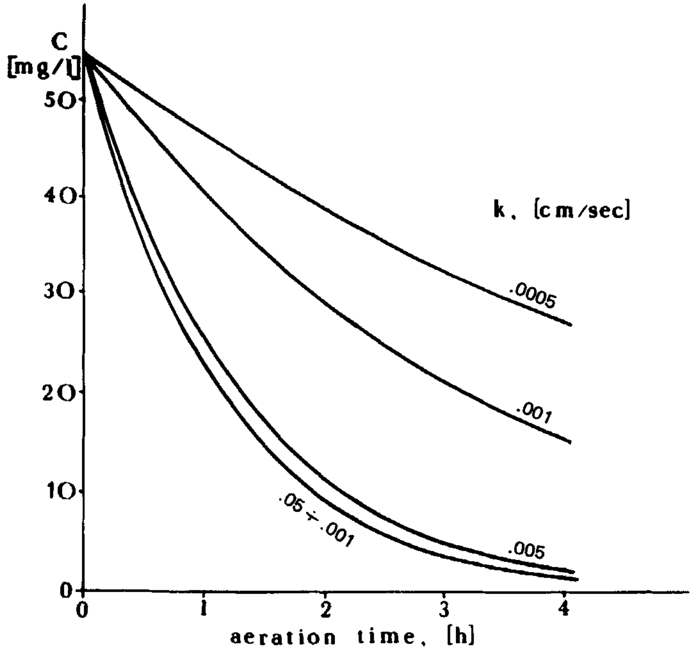

FIG. 4. Removal of mesitylene due to the evaporation process; the effect of the mass transfer rate coefficient value. Bubble diameter d_b = 1 mm, K_H = 0.265.

coefficient of 0.05–0.0005 cm/s does not influence the mesitylene removal rate in the case of fine bubbles ($d_b < 0.2$ mm).

In the case of a large mass transport coefficient ($k = 0.05$ cm/s), bubble size does not influence the removal rate. The terminal evaporation efficiency was observed for all sizes of bubbles if $k = 0.05$ cm/s or more, and also for all values of k if the bubble diameter equals 0.2 mm or less. Further improvement of the evaporation efficiency is possible by increasing the volume gas flow rate.

Figure 6 presents the total mass of mesitylene carried out of the solution by evaporation. Calculations were made on the basis of Eq. (13) (solid lines) and Eq. (15) (dashed lines) for bubbles of 1 mm diameter. The solid lines correspond to the processes in which the solute concentration in water diminishes due to the evaporation process, and the dissolution of emulsion droplets is so slow that it does not influence the solute concentration. The dashed lines correspond to the processes in which solute dissolution is much faster than evaporation into the bubble, thus the solute concentration in water may be treated as a constant

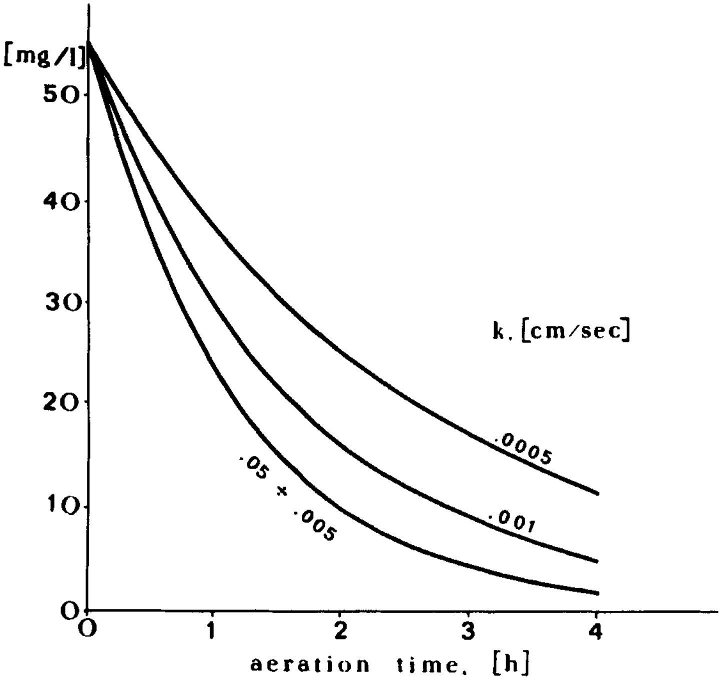

FIG. 5. Removal of mesitylene due to the evaporation process; the effect of the mass transfer rate coefficient value. Bubble diameter d_b = 0.6 mm, K_H = 0.265.

during the run. Lines corresponding to the real processes probably lie in between, for which it would be necessary to take into account the above-mentioned dissolution rate constant.

The results of mesitylene removal calculated for both the simultaneous processes (hydrodynamic capture and evaporation) may be summarized as follows. Figure 7 presents mesitylene removal from an emulsion of 114 mg/dm^3 during its aeration. The summary curve is drawn as Line 3 [the sum of interception (Line 1) and evaporation (Line 2)], or as Line 5 [the sum of interception (line 1) and evaporation with simultaneous droplet dissolution (Line 4, then Line 2)]. We have assumed that from the moment identified as "X," when the total concentration of mesitylene reaches 57 mg/dm^3 and all droplets of hydrocarbon disappeared (dissolved or captured by a bubble), the mesitylene is removed only by evaporation. From that moment the resulting curve (5B) is computed only according to Eq. (13) because interception and dissolution do not occur.

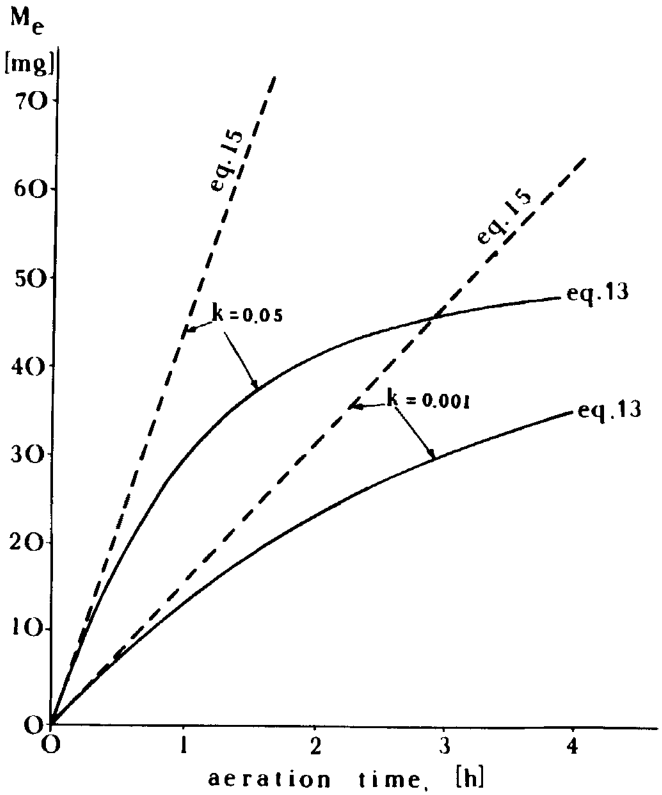

FIG. 6. Total mass of mesitylene removed due to the evaporation process calculated according to Eq. (13) (solid lines) or Eq. (15) (dashed lines). Bubble diameter d_b = 1.0 mm, K_H = 0.265.

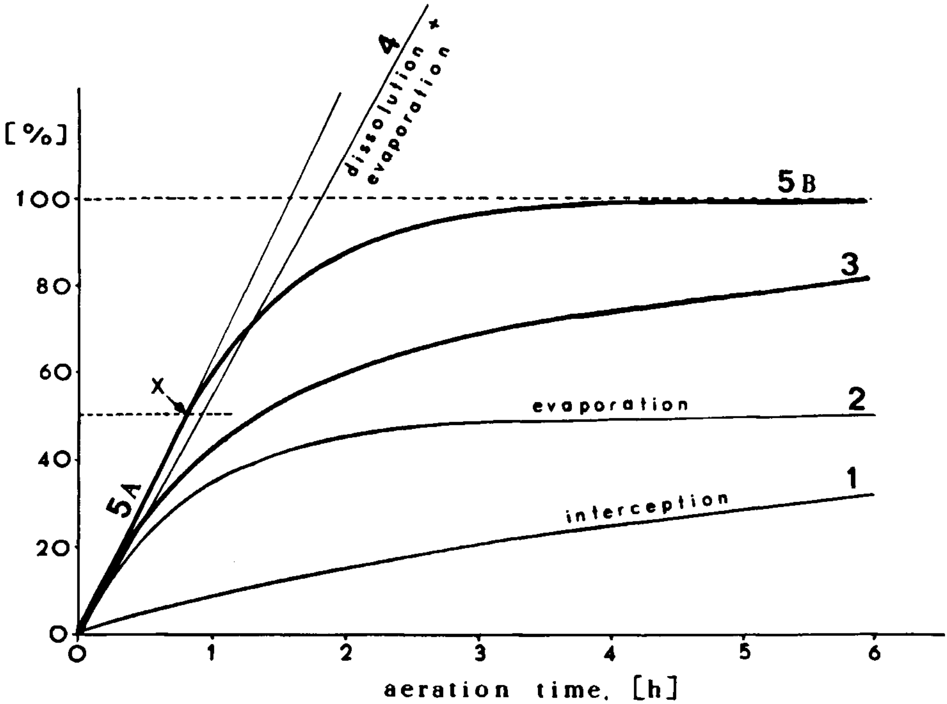

FIG. 7. Mesitylene removal efficiency calculated for interception and evaporation processes. Initial content of mesitylene in emulsion: 114 mg/dm^3 (57 mg/dm^3 as droplets and 57 mg/dm^3 as dissolved). Bubble diameter d_b = 0.616 mm, particle diameter d_p = 3.34 μm, K_H = 0.444, k = 0.01 cm/s, V_g = 1.508 cm^3/s. (1) Interception (Eq. 6). (2) Evaporation (Eq. 13). (3) Sum of (1) and (2). (4) Dissolution + evaporation (Eq. 15). (5) Sum of (1) and (4) (below point X), or (2) (above point X).

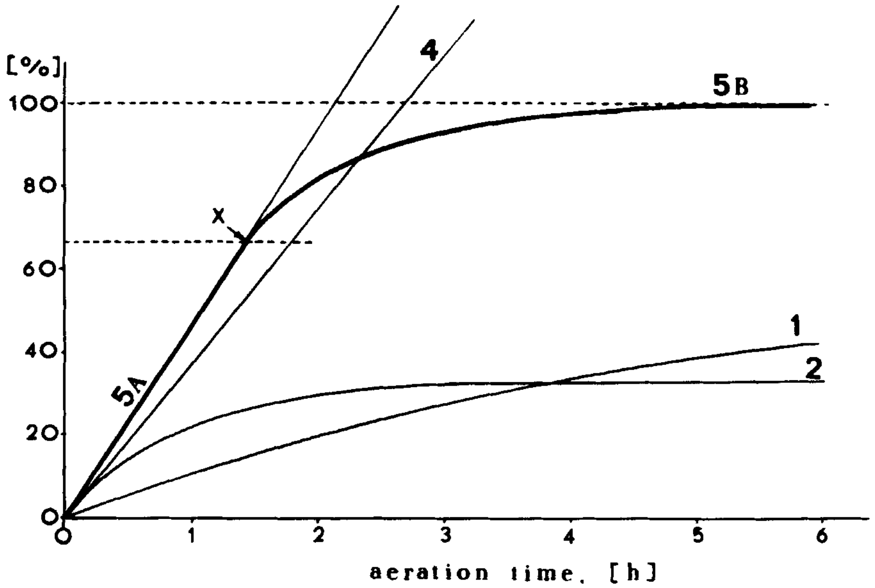

FIG. 8. Mesitylene removal efficiency calculated for interception and evaporation processes. Initial content of mesitylene in emulsion: 171 mg/dm^3 (114 mg/dm^3 as droplets and 57 mg/dm^3 as dissolved). Remarks and computer parameters as in Fig. 7.

Figure 8 presents the curves of partial processes proceeding during the aeration of a mesitylene emulsion of concentration 171 mg/dm^3.

Note that interception of hydrocarbon droplets by a bubble has an insignificant influence on the efficiency of mesitylene removal. This hydrocarbon is removed mainly by evaporation. This is especially true for less concentrated emulsions. For example, after 1 h of aeration of an emulsion containing 171 mg/dm^3 mesitylene, about 22% is removed by evaporation and 10% by interception (Fig. 8), while from an emulsion containing 114 mg/dm^3 mesitylene, 33% is removed by evaporation and only 8% by interception (Fig. 7). For an emulsion containing 70 mg/dm^3, the relative values are 47 and 3%.

EXPERIMENTAL

Batch runs of the flotation were carried out in the glass column described previously (*7*). The liquid head was 95 cm, the inner column diameter was 45 mm, and the gas flow rate were as noted in the

TABLE 1
Parameters Characterizing Flotation Runs

Run	Emulsion characteristic: Mesitylene content: Total (mg/dm^3)	As droplets (%)	Dissolved (%)	Mean droplet diameter (μm)	Volume gas flow rate V_g (cm^3/s)	Mean bubble diameter d_{32} (mm)	d_{30} (mm)
I	70	18.6	81.4	2.52	1.508	0.616	0.510
II	89	36.0	64.0	3.93	1.508	0.518	0.440
III	103	44.7	55.3	4.41	1.111	0.587	0.480

particulate runs of Table 1. The mesitylene emulsions were stabilized mechanically. The mesitylene concentration was determined by the GLC method described earlier (7). The lowest hydrocarbon concentration determined by this method was about 0.8 mg/dm^3. The droplet sizes in the emulsion were determined by microscopic measurements. The bubble sizes were measured photographically. The mean bubble diameter used in the theoretical evaporation computations was calculated on the basis of Eq. (1) (so-called Sauter diameter, Table 1). The use of this diameter allows the condition of constant bubble volume and surface to be fulfilled in both systems: in the real system and in the system containing bubbles of mean diameter. This is especially important in the evaporation process where the bubble surface is the most important parameter influencing the removal efficiency. The mean bubble diameter calculated according to Eq. (3) was applied in the theoretical computation of the interception process.

The mean mesitylene droplet diameter was calculated according to Eq. (1).

Figures 9, 10, and 11 present the results of experimental flotation runs and the theoretical results computed for related parameters.

CONCLUSIONS

The flotation of aromatic hydrocarbons (for example, mesitylene) from their O/W-type emulsions proceeds according to a mixed model. This model includes interception of oil droplets by a rising bubble as well as evaporation of dissolved hydrocarbon from the bulk into a bubble. The removal effect depends on the initial hydrocarbon content in the

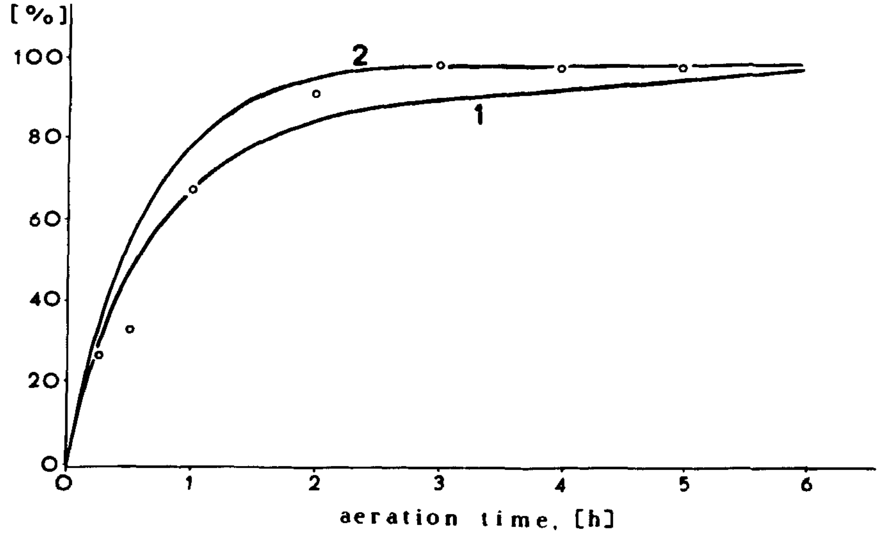

FIG. 9. Comparison of experimental data (circles) and computed results (solid lines) for Run I. (1) Results computed according to the interception and evaporation model. (2) Results computed according to interception and evaporation with the dissolution model. Parameters: $k = 0.01$ cm/s, $K_H = 0.444$, other parameters as in Table 1.

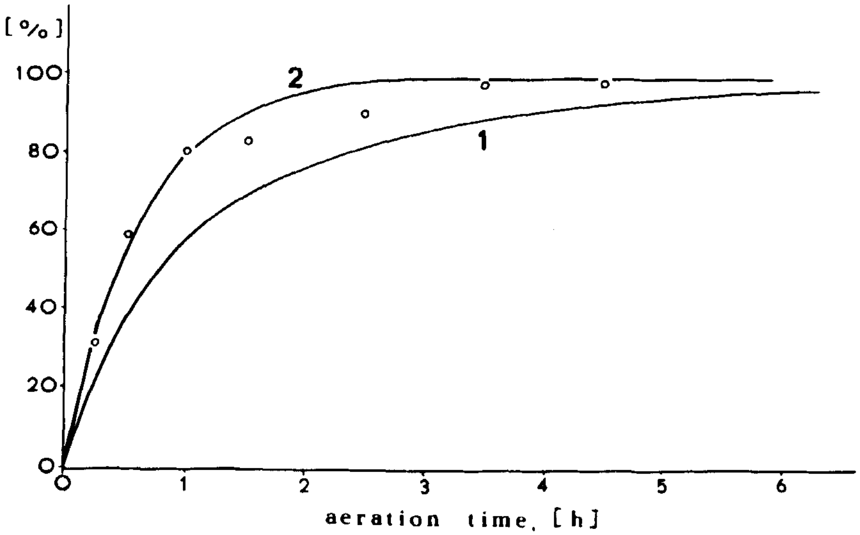

FIG. 10. Comparison of experimental data (circles) and computed results (solid lines) for Run II. Remarks and parameters as in Fig. 9.

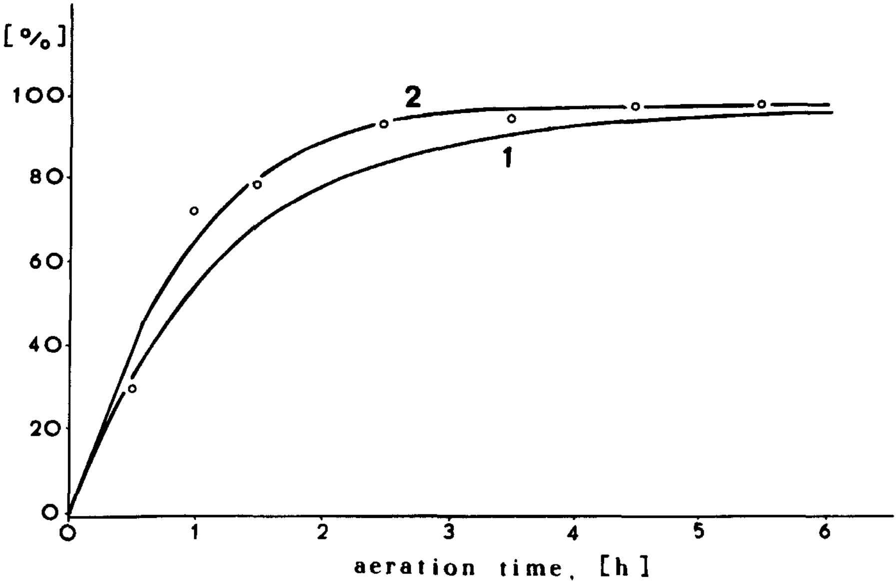

FIG. 11. Comparison of experimental data (circles) and computed results (solid lines) for Run III. Remarks and parameters as in Fig. 9.

emulsion and also on the water solubility and saturated vapor pressure of the hydrocarbon.

Good agreement was found between experimental mesitylene removal and theoretically computed results, especially when the dissolution process of the experimental data was considered (Figs. 9–11, Curves 2). The differences observed at the beginning of the runs are probably connected with two assumptions. 1) The dissolution process proceeds much faster than the evaporation process, thus the mass transport is a consequence of the evaporation as calculated with the use of constant solute concentration equal to its solubility (Curves 2). 2) The dissolution process is so slow that the bulk solute concentration change caused by this process is negligible, and this concentration depends on the evaporation rate only (Curves 1).

In reality, the dissolution rate influences the solute concentration in bulk, and during the flotation run this concentration is probably lower than the extreme solubility used in the computed model results and presented by Curves 2 (Figs. 9–11).

We conclude that the dissolution rate constant should be taken into account in the simulation of experimental results.

SYMBOLS

r_b, r_p	radius of bubble and particle, respectively
d_{32}	Sauter mean diameter
d_{30}	arithmetic mean diameter
u	bubble rise velocity
n	number of bubbles generated in a unit of time = $3V_g/4\pi r_b^3$
n_i	number of bubbles or droplets of diameter d_i
g	gravitational constant
η	aqueous layer viscosity
ρ	aqueous layer density
ρ_p	particle density
h	height of the liquid head
V_g	volumetric gas flow rate
V_s	volume of the solution
D	flotation column diameter
t	residence time of a bubble in the aqueous layer = h/u
τ	time from the beginning of the run
C, C_0	residual and initial hydrocarbon concentration in emulsion, respectively
C_{aq}	concentration of dissolved hydrocarbon
k	mass transfer rate coefficient
K_H	Henry's law constant for solute in water
Re	Reynolds number = $2ur_b\rho/\eta$
Stk	Stokes number = $2\rho_p r_p^2/9\eta r_b$
M_i	hydrocarbon mass removed by interception
M_e	hydrocarbon mass removed by evaporation
M	global hydrocarbon mass removed = $M_i + M_e$

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