In Situ Method for Determining Combination of Organic Compounds Interacting with Each Other on Silicon Oxide Surface

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An experimental process for determining a combination of organic compounds, interacting with each other on a silicon oxide surface, was developed using a quartz crystal microbalance. The surface concentration of the four organic compounds, such as L-menthone (Mth), decylacetate (DA), diethylphthalate (DEP) and octanol (OCT), on a silicon oxide surface was evaluated in multi-component systems. The increase and decrease in the surface concentrations were not reversible with the changing gas phase concentrations in the two-component system consisting of Mth and DEP. Additionally, the small droplet shape of Mth placed on the DEP-coated glass plate surface was significantly different from that of DEP on the Mth-coated surface. Based on these results, Mth and DEP molecules were considered to have a significant interaction with each other.

For this purpose, the molecular behaviors in single-component and multi-component systems have been studied by taking into account the physisorption and desorption rates using the quartz crystal microbalance (QCM). In order to achieve a desirable surface condition, the adsorption behavior of various compounds should be clarified. Particularly, the combination of compounds interacting with each other should be found by means of a simple and easy method.

Figure 1 shows the QCM system used to measure the physisorption from the QCM. Organic compounds was evaluated for verifying the results obtained from the QCM.

Experimental

Measurement.— This study mainly used decylacetate (DA), L-menthone (Mth) and diethylphthalate (DEP). Octanol (OCT) was additionally used as the reference material. The molecular structures and Fig. III shows the molecular shapes. DA has a long carbon chain including two oxygen atoms, as shown in Figs. 1a–II. One of the oxygen atoms is considered to be oriented to the silicon oxide surface, as shown in Figs. 1a–III. Figs. 1b–II shows that Mth contains cyclohexane to which one oxygen atom is bonded. When Mth is physisorbed on the silicon oxide surface, the oxygen atom is expected to be oriented to the silicon oxide surface. As shown in Figs. 1c–II and III, DEP has four oxygen atoms, which are effective for the physisorption on the silicon oxide surface.

Figure 2 shows the QCM system used to measure the physisorption and desorption of organic compounds on a silicon surface in a dry ambient atmosphere. The QCM sensor (Halloran Electronics Co., Ltd., Tokyo) consisted of a circular-shaped quartz plate, AT-cut,18

(a) Decylacetate (DA): C₁₉H₂₆O₂
b.p.: 272 °C
MW: 200.32
Oxygen: 2
Long chain

(b) L-Menthone (Mth): C₁₇H₂₆O
b.p.: 208 °C
MW: 154.25
Oxygen: 1
Bulky

(c) Diethylphthalate (DEP): C₁₄H₁₀O₄
b.p.: 295 °C
MW: 222.24
Oxygen: 4
Bulky with many oxygen

Figure 1. Organic compounds mainly used in this study. (a) Decylacetate (DA), (b) L-menthone (Mth) and diethylphthalate (DEP). (I): molecular structure and (II): molecular shape oriented to silicon oxide surface.
Figure 2. Quartz crystal microbalance system used to measure the physisorption and desorption of molecules of decylacetate (DA), L-menthone (Mth) and diethylphthalate (DEP) on a silicon oxide surface in ambient nitrogen.

Figure 3. Process and surface condition. (a) Gas phase concentrations of compounds Org1 and Org2, (b) molecular condition at surface, when shared with compounds Org1 and Org2, for Conditions A-E, (c) molecular condition at surface, when compounds Org1 and Org2 are stacked, for Conditions A-E, and (d) macroscopic surface condition consisting of droplet and surface film, for Conditions B and D.
When Org1 and Org2 have no significant interaction with each other and share the silicon oxide surface, the surface condition becomes those for Conditions B and D in Fig. 3b. Because Conditions B and D are equivalent, the surface concentration for Condition B is the same as that for Condition D. The measured surface concentration during the process shown in Fig. 3a has a reversible increase and decrease behavior, that is, symmetrical shape, as shown in Fig. 3b.

Next, Fig. 3c shows a different possibility. When the oxygen atoms of an organic molecule, shown in Fig. 1, are oriented to the silicon oxide surface, the upper surface has a non-polar nature. The molecules on such a surface may have a non-polar molecular interaction. When Org1 and Org2 have a different type of interaction with each other, such as the mixing or adhesion relationship on the silicon oxide surface, the surface condition is expected to become different, for example, as those shown for Conditions B and D in Fig. 3c. Condition B in Fig. 3c shows the same as Condition B in Fig. 3b. Org1 and Org2 are individually physisorbed on silicon oxide surface without interacting with each other. In contrast, Condition D in Fig. 3c shows that Org1 and Org2 are mixed or combined with each other, utilizing an interaction with each other. Because the surface conditions depicted in Condition B in Fig. 3c are not equivalent to that of Condition D in Fig. 3c, the surface concentration for Condition B can be different from that for Condition D. Thus, the measured surface concentration during the process shown in Fig. 3c may have an irreversible increase and decrease, that is, asymmetrical shape. Thus, the surface concentration plots have symmetrical and asymmetrical appearances for the non-interacting and interacting compounds, as shown in Figs. 3b and 3c, respectively.

For the QCM measurement, the increasing and decreasing trend between the previous and next conditions is empirically quite well reproduced, even when the values are often slightly shifted. Thus, the symmetrical or asymmetrical shape of the surface concentration plot from Condition A to E was considered to be important information for determining the existence of the molecular interaction.

When the molecular interaction between Org1 and Org2 exists, it may appear as a macroscopic behavior, like a liquid-liquid contact. Figure 3d shows a possible example. Comparing the droplet shape of Org1 placed on the Org2 thin layer with the opposite combination, shown as Conditions B and D in Figs. 3b and 3d, respectively, it is expected to be a very simple and easy method. Although the liquid/liquid interface and gas/liquid interface is not the same, both of them can have the same molecule-molecule combinations and interactions at the interfaces. Thus, the obtained result by the liquid/liquid interface was expected to include the same behavior by the gas/liquid interface.

When the droplet shapes in the two cases are significantly different from each other, a considerable molecular interaction may exist. When the Org1 droplet is physically placed on the Org2 liquid layer surface without any special interaction, the Org1 droplet does not spread over the Org2 liquid layer surface as shown for Condition B in Fig. 3d. In contrast, when the Org2 droplet has a very good contact with the Org1 liquid layer beneath it, the Org2 droplet widely spreads over the Org1 liquid layer surface as shown for Condition D in Fig. 3d. Such a difference shown in Figs. 3b and 3d is expected to be important information, when it is consistent with the results obtained from the QCM.

In order to avoid the liquid droplet formation by condensation, the surface temperature should be higher than room temperature.

**Rate theory.—Rate parameters.—** The physisorption and desorption behavior of airborne organic compounds on a silicon oxide surface is described taking into account the following assumptions, for the MOSAIC model.9–16

Assumption 1: There is quite small interaction between organic molecules in gas phase and on silicon surface.

Assumption 2: Organic molecules are physisorbed on silicon surface due to very weak interactions between organic molecule and the silicon surface.

Assumption 3: The physisorption rate of organic molecule of compound $i$ is proportional to its concentration in gas phase and is also proportional to the difference between the surface concentration of the total physisorbed organic compounds and its larger limit.

Assumption 4: The desorption rate of organic molecules of compound $i$ is proportional to its concentration on silicon surface.

By these assumptions, Eqs. 1–3 are obtained.

$$\frac{dS_i}{dt} = (S_{\text{MAX}} - S_{\text{all}}) k_{\text{ad},i} C_i - k_{\text{de},i} S_i,$$

$$S_{\text{all}} = \sum_j S_j,$$  \hspace{1cm} (2)

in a steady state,

$$S_i = \frac{(S_{\text{MAX}} - S_{\text{all}}) k_{\text{ad},i} C_i}{k_{\text{de},i}}.$$  \hspace{1cm} (3)

where $i$ and $j$ indicate the organic compounds, $S_i$ is the surface concentration of organic compound $i$ (ng cm$^{-2}$), $C_i$ is the concentration of organic compound $i$ in the gas phase (ng cm$^{-3}$), $k_{\text{ad},i}$ is the physisorption rate constant of organic compound $i$ on a silicon oxide surface (cm$^2$ ng$^{-1}$ s$^{-1}$), $k_{\text{de},i}$ is the desorption rate constant of organic compound $i$ from the silicon oxide surface (s$^{-1}$), and $S_{\text{MAX}}$ is the higher limit of the concentration of the total organic compounds physisorbed on a silicon oxide surface (ng cm$^{-2}$). $S_{\text{all}}$ is the summation of all the organic compounds physisorbed on a silicon oxide surface (ng cm$^{-2}$).

In a single-component system and in the steady state, the inverse values of $S_i$ and $C_i$ show a linear relationship as described by Eq. 4.12–14

$$\frac{1}{S_i} = \frac{k_{\text{de},i}}{S_{\text{MAX}} k_{\text{ad},i}} \frac{1}{C_i} + \frac{1}{S_{\text{MAX}}}.$$  \hspace{1cm} (4)

The values of $k_{\text{de},i}/k_{\text{ad},i}$ and $S_{\text{MAX}}$ are obtained using the plot of $1/S_i$ versus $1/C_i$.

The occupied molecule area, $A_i$ (cm$^{-2}$), and the molecular radius of compound $i$, $r_i$ (cm), are calculated using the $S_{\text{MAX}}$ value.

$$A_i = r_i^2 \pi = \frac{MW_i}{S_{\text{MAX}} \times N_A}$$  \hspace{1cm} (5)

where $MW_i$ is the molecular weight of organic compound $i$ (g mol$^{-1}$). $N_A$ is Avogadro’s number.

The desorption rate constant is obtained using Eq. 6 along with the desorption process of an organic compound.9–14

$$\frac{dS_i}{dt} = -k_{\text{de},i} S_i.$$  \hspace{1cm} (6)

$S_{\text{MAX}}$ and molecular interaction radii in a multicomponent system.—The $S_{\text{MAX}}$ value is used for evaluating the molecular interaction radii of the organic compounds in a multicomponent system.13 When the unit area, 1 cm$^2$, is fully occupied by an organic compound $i$ ($i = 1, 2, ..$), its situation can be described using the maximum population of the organic compound $i$, $N_{\text{MAX},i}$ (molecules/cm$^2$) and $r_i$. Thus, the summation of the occupation area by the organic molecules, $A_{\text{all}}$ (cm$^{-2}$), is expressed as follows:

$$A_{\text{all}} = 1 = \sum_i N_{\text{MAX},i} r_i^2 \pi,$$  \hspace{1cm} (7)

where $N_{\text{MAX},i}$ is assumed to have a relationship with the $S_{\text{MAX}}$ of the organic compound $i$, $S_{\text{MAX},i}$ (ng cm$^{-2}$).

$$N_{\text{MAX},i} = \frac{S_{\text{MAX},i}}{MW_i \times 10^9} \times N_A,$$  \hspace{1cm} (8)

$$S_{\text{MAX}} = \sum_i S_{\text{MAX},i}. \hspace{1cm} (9)$$
Thus, Eq. 12 is obtained for evaluating the and kinetic one between organic compounds in the gas phase and equal to 1.0. However, because any significant attraction and/or repulsion between the molecules is expected to cause shrinking and expansion, respectively, of the molecular radii, Eq. 10 will not be equal to 1.0. When the molecular radii in the multicomponent system are the same as those in the single-component system, the right side of Eq. 10 can produce values near 1.0. From Eq. 1 in the steady state, the ratio between the $S_{\text{max,i}}$ values is assumed to be the same as that of the $S_i$ values and also as that of the $k_{\text{ad,i}} / k_{\text{de,i}}$ values as follows:

$$S_1 : S_2 : \ldots : S_i : \ldots = \frac{k_{\text{ad,i}} C_i}{k_{\text{de,i}}} : \frac{k_{\text{ad,2}} C_i}{k_{\text{de,2}}} : \ldots : \frac{k_{\text{ad,i}}}{k_{\text{de,i}}} : \ldots$$

$$\equiv S_{\text{max,1}} : S_{\text{max,2}} : \ldots : S_{\text{max,i}} : \ldots$$

Thus, Eq. 12 is obtained for evaluating the $S_{\text{max,i}}$ value.

$$S_{\text{max,i}} = S_{\text{MAX}} \left( \frac{k_{\text{ad,i}} C_i}{k_{\text{de,i}}} \right)$$

When the molecular radii in the multicomponent system are the same as those in the single-component system, the right side of Eq. 10 can produce values near 1.0. However, because any significant attraction and/or repulsion between the molecules is expected to cause shrinking and expansion, respectively, of the molecular radii, Eq. 10 will not be equal to 1.0.

For further extension, the interactions, such as attraction, repulsion and kinetic one between organic compounds in the gas phase and at the surface should be described and added to Eq. 1. However, for a simple approach, the difference of the surface concentration behavior between the measurement and that from the MOSAIC model was found and evaluated, in this study.

**Results and Discussion**

**Two-component system.**— The two-component system of various compound combinations was studied. Figures 4a and 4b show the gas phase concentration and the surface concentration, respectively, in a two-component system consisting of Mth and DA. As shown in Fig. 4a, the gas phase concentration of DA decreased and increased, while that of Mth increased and decreased. Figure 4b shows that the surface concentration of the total physisorbed amount of Mth and DA increased and decreased. The surface concentration for Condition B is the same as that for Condition D. Although there was a slight difference between the surface concentrations for Conditions A and E, the entire shape of the plots from A to C is recognized to be the reverse shape of that from C to E. Thus, the two-component system of Mth and DA was classified as a symmetrical or reversible behavior shown in Fig. 3b, that is, the system without a considerable molecular interaction.

Additionally, Figs. 5a and 5b show the gas phase concentration and the surface concentration, respectively, in a two-component system consisting of DA and DEP. As shown in Fig. 5a, the gas phase concentration of DEP decreased and increased, while that of DA increased and decreased. Figure 5b shows that the surface concentration of the total physisorbed amount of DA and DEP increased and decreased. The surface concentration for Condition B of about 16 ng cm$^{-2}$ was similar to that for Condition D. The surface concentration for Condition A was the same as that for Condition E. The entire shape of the plots from A to C is recognized to be the reverse shape of that from C to E. Thus, the two-component system of DA and DEP was classified as symmetrical or reversible behavior shown in Fig. 3b, that is, the system without a considerable molecular interaction.

Next, Figs. 6a and 6b show the gas phase concentration and the surface concentration, respectively, in a two-component system consisting of Mth and DEP. As shown in Fig. 6a, the gas phase concentration of DEP increased and decreased, while that of Mth decreased and increased. Figure 6b shows the surface concentration of the total physisorbed amount of Mth and DEP. The surface concentration for Condition B was similar to that for Condition A. Next, the surface concentration for Condition C became lower than that for Condition
The surface concentrations for Conditions D and E were similar to that for Condition C. Although the trends from Condition A to C and from Condition C to E in Figure 6b were entirely unclear, it might show an asymmetrical or irreversible shape, unlike Figs. 4 and 5. Because the surface concentration at Condition E tended to be slightly less than that for Condition D, this is recognized to be different from that expected by the change going from Condition A to B. The characteristic behavior was that the surface concentration did not increase with the increasing gas phase concentration of Mth after being exposed to DEP at the high gas phase concentration.

**Three-component system.**— By means of one sequential operation using the three-component system, the behavior obtained in Figs. 4, 5 and 6 was expected to be easily determined, as shown in Fig. 7. Figure 7 shows the gas phase concentration and surface concentration in a three-component system consisting of Mth, DA and DEP. Overall, nine conditions, Conditions A-I, were set using the various combinations of the gas phase concentrations.

In Conditions A, B and C, the gas phase concentration of Mth remained constant. That of DA decreased and increased, while that of DEP increased and decreased. As shown for Conditions A, B and C in Fig. 7c, the surface concentration decreased for Condition B, while that for Condition C again increased to the same value as that for Condition A.

A similar behavior could be observed for Conditions G, H and I. Through G, H and I, the gas phase concentration of DEP remained constant. The gas phase concentration of Mth increased and decreased, while that of DA decreased and increased. Under these conditions, the surface concentrations decreased for Condition H. That for Condition I again increased to the same value as that for Condition G. The surface concentration behavior observed for Conditions A-C and G-I was recognized to be symmetric or reversible.

In contrast, the surface concentration behavior for Conditions D, E and F was different from those for Conditions A-C and G-I. Through Conditions D, E and F, the gas phase concentration of DA remained constant. That of Mth decreased at Condition E. Next, it increased at Condition F. Simultaneously, that of DEP increased at Condition E. Finally, it then decreased at Condition F. Although the surface concentration increased at Condition E, that at Condition F was maintained and did not return to that at D.

In Fig. 7, the surface concentration behavior for Conditions D-F was clearly different from that for Conditions A-C and G-I. This indicated that the Mth physisorbed on DEP had a different behavior from that by the combinations of Mth-DA and DA-DEP. Similar to Fig. 6, the characteristic behavior was that the surface concentration had not increased with the increasing gas phase concentration of Mth after being exposed to DEP at the high gas phase concentration.

The molecular conditions are speculated and schematically shown in Fig. 8, in which Conditions A-I correspond to those in Fig. 7. In this figure, each bar on silicon oxide surface indicates the organic compound. The bar height qualitatively shows the effective maximum surface concentration of the organic compound, obtained in the single-component system. The bar width qualitatively indicates the ratio of surface concentration of the organic compound. The hatched bar is the condition having interaction between DEP and Mth.

As shown for Conditions A, B and C, the surface concentration of DA were considered to decrease and increase, while that of DEP were considered to increase and decrease, corresponding to the gas phase concentration changes in Fig. 7. In these processes, the amount of interacted condition was considered to be still quite small. At Condition D, the surface condition was expected to be similar to that before Condition C. At Condition E, the surface concentration of DEP increased, while that of Mth decreased. Additionally, the $S_{all}$ value increased. During the change from Condition D to E, the amount having the interaction was considered to increase and remain.

As shown for Conditions E-I in Fig. 7, the $S_{all}$ value entirely shifted to higher. The surface was expected to have and keep an additional amount of DEP and Mth, having the interaction between them, indicated as the hatched region in Fig. 8. Thus, the total amount on the silicon oxide surface was expected to be greater than that calculated without the interaction. This situation is depicted for Conditions E-I in Fig. 8. The total organic compound is greater than that originally on the silicon oxide surface.

In order to confirm that the interacting combination included DEP, the three-component behavior was measured using Mth, DA and OCT, as shown in Fig. 9. Conditions A-C, D-F and G-I have the constant gas phase concentration of DA, Mth and OCT, respectively. The molecular conditions are schematically shown in Fig. 10, in which Conditions A-I correspond to those in Fig. 9. In Fig. 10, the bar height relatively indicates the effective maximum surface concentration of the organic compound, obtained in the single-component system. The bar width quickly indicates the ratio of surface concentration.

Because the surface concentration symmetrically increased and again decreased at Conditions A-C, D-F and G-I, the surface concentrations of Mth, DA and OCT are considered to simply change corresponding to the gas phase concentrations, as shown in Fig. 10. Thus, the molecular interaction between Mth, DA and OCT was recognized to be small. Comparing the behavior in Fig. 9 with that in Fig. 7, the irreversible increase and decrease result could be obtained only for Conditions D, E and F in Fig. 7. Thus, the combination of Mth...
Figure 9. Gas phase concentration and surface concentration in a three-component system consisting of Mth, DA and OCT. (a): gas phase concentration of Mth, (b) gas phase concentration of DA and OCT, (c): surface concentration and (d): $A_{\text{all}}/A_{\text{SiO}_2}$ ratio.

Figure 10. Molecular conditions of OCT, Mth and DA at silicon dioxide surface. Conditions A-I correspond to those in Fig. 9. Bar height indicates the effective maximum surface concentration of the organic compound, obtained in the single-component system. Bar width indicates the ratio of surface concentration.

and DEP was considered to have a considerable interaction between them.

Rate parameters.— In order to numerically evaluate the existence of the molecular interaction, the ratio, $A_{\text{all}}/A_{\text{SiO}_2}$, was evaluated using Eq. 10 and the rate parameters listed in Table I. These values were evaluated using Eqs. 1–12 in this study and previous studies.13–16

When the $A_{\text{all}}/A_{\text{SiO}_2}$ ratio is greater than one, the organic molecules in a multi-component system were more densely arranged than those in a single-component system. Thus, the molecules are considered to be attracted. In contrast, when the $A_{\text{all}}/A_{\text{SiO}_2}$ ratio is less than one, the organic molecules in a multi-component system have large distances between them than those in a single-component system. Thus, the molecules are considered to undergo repulsion. Taking into account that the measurement error is roughly 10%, the $A_{\text{all}}/A_{\text{SiO}_2}$ ratio less than 0.9 and greater than 1.1 may show a considerable molecular interaction.16

As shown in Fig. 7d, before Condition D, the $A_{\text{all}}/A_{\text{SiO}_2}$ ratio was near 1.1. However, the $A_{\text{all}}/A_{\text{SiO}_2}$ ratio became greater than 1.2 after Condition E till Condition I. This indicated that the molecular interaction between Mth and DEP occurred at Condition E. In contrast, in Fig. 9d, the $A_{\text{all}}/A_{\text{SiO}_2}$ ratio was near 1.1 from Condition A to I. This suggested that the molecular interaction among Mth, DA and OCT was not significant. This behavior is classified as the reversible or symmetric one shown in Fig. 3b.

It is noted here that the gas phase concentration of DEP was still high for Conditions G–I in Fig. 7. Additionally, DA and Mth have no obvious interaction as shown in Figs. 4 and 9. Taking into account that the measurement was sequentially performed from Conditions A to I, the influence of the intense interaction caused during Condition E might remain after Condition F in Figs. 7 and 8 in order to maintain the high $A_{\text{all}}/A_{\text{SiO}_2}$ ratio value.

Droplet contact.— Because the surface concentration behavior for the combination of Mth and DEP might be different from the other combinations, the droplet shapes of Mth on DEP and that of DEP on Mth were observed, as shown in Fig. 11. The droplet had a volume of 0.025 cm$^3$.

Fig. 11a shows the Mth and DEP droplet placed on a bare glass plate surface, which had been carefully wiped with ethanol. The diameter of the Mth droplet and DEP droplet on the glass plate was greater than 8 mm and 6 mm, respectively. However, as shown in Fig. 11b, the diameter of the Mth droplet on the DEP-coated surface was greater than 20 mm, while that of DEP on the Mth-coated surface was nearly 7 mm, as shown in Fig. 11c. Thus, the DEP droplet on

Table I. Rate parameters of organic compounds obtained in a single-component system.

| Octanol (Oct) | L-Menthone (Mth) | Decylacetate (DA) | Diethylphthalate (DEP) |
|--------------|------------------|------------------|------------------------|
| Physisorption rate constant (ng s cm$^{-1}$) | $5.5 \times 10^{-3}$ | $1.1 \times 10^{-3}$ | $2.4 \times 10^{-3}$ | $6.5 \times 10^{-3}$ |
| Desorption rate constant (s$^{-1}$) | 0.29 | 0.26 | 0.11 | 0.046 |
| Effective maximum surface concentration (ng cm$^{-3}$) | 38 | 26 | 47 | 27 |
| Occupation area of molecule (cm$^2$) | $5.7 \times 10^{-15}$ | $9.9 \times 10^{-15}$ | $7.1 \times 10^{-15}$ | $1.4 \times 10^{-14}$ |
| Relative occupation area of molecule (-) | 0.5 | 1 | 0.7 | 1.4 |
| Occupied diameter of molecule (cm) | $7.5 \times 10^{-8}$ | $9.9 \times 10^{-8}$ | $8.4 \times 10^{-8}$ | $1.2 \times 10^{-7}$ |
| Relative occupied diameter of molecule (-) | 0.8 | 1 | 0.8 | 1.2 |
the Mth-coated surface had a shape like that on the bare glass plate surface; the condition of Mth on the DEP-coated surface was quite different from those shown in Figs. 11a and 11c. Thus, the DEP-coated surface provided an interaction which produced a sufficiently stable contact with Mth.

In order to totally understand the liquid-liquid contact behavior, the droplets of four organic compounds used in this study were placed on the surfaces coated with the four organic compounds. The droplet sizes are listed in Table II. Small, medium, large, and very large are the diameters of less than 10 mm, 10–15 mm, 15–20 mm, and greater than 20 mm, respectively.

Both the Mth droplet on the OCT-coated surface and the OCT droplet on the Mth-coated surface showed a large size. However, the DA droplet put on the Mth-coated surface had a large size, while the Mth droplet on the DA-coated surface showed a medium size. Overall, when the relationship of the droplet and the coated material became opposite, the droplet size tended to be unchanged or shift to the next level, such as small to medium, or medium to large. Being consistent with Fig. 9, the Mth droplet on the DEP-coated surface had the highest level, that is, very large, although the DEP droplet on the Mth-coated surface showed the smallest level. The combination showing such a significant difference was only DEP and Mth. This result might indicate the existence of a considerable interaction, consistent with those obtained in Figs. 6 and 7.

## Conclusions

An experimental process for determining a combination of organic compounds interacting with each other on silicon oxide surface was developed using a quartz crystal microbalance and a liquid-liquid contact shape. The adsorption and desorption behaviors of four organic compounds, such as L-menthone (Mth), decylacetate (DA), diethylphthalate (DEP) and octanol (OCT), on a silicon oxide surface was evaluated in a multi-compound system. When the increase and decrease of the surface concentrations with the increasing and decreasing gas phase concentrations, respectively, of various compounds were irreversible, the combination of the organic compounds was assumed to have a considerable interaction with each other. For the combination of Mth and DEP, the surface concentration behavior was irreversible. Additionally, while the small Mth droplet size placed on the DEP-coated glass plate surface widely spread, it was different from the size of the small DEP droplet placed on the Mth-coated surface. Based on these results, the Mth and DEP molecules were considered to have a considerable interaction with each other.

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