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Rate Theory of Multicomponent Adsorption of Organic Species on Silicon Wafer Surface

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For the first time, the time-dependent change in the concentrations of organic species adsorbed on a silicon wafer surface is modeled using numerical calculations based on rate theory. An equation composed of the adsorption rate from the gas phase to the silicon wafer surface and the desorption rate from the silicon wafer surface is developed accounting for competitive processes in a multicomponent system. This equation can describe and predict the actual increase and decrease in the surface concentrations of propionic acid ester, siloxane (D9), and di(2-ethylhexyl)phthalate. It is also indicated that the organic species having a large adsorption rate with a small desorption rate remains in significant abundance on the silicon wafer surface for a very long period after cleaning.

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Advanced electronic devices currently have a serious problem in their fabrication process due to the presence of organic hydrocarbon contamination on the silicon wafer surface.[1-7] To develop the technology to achieve a sufficiently clean silicon wafer surface, many kinds of organic species existing on the silicon wafer surface have been studied, such as propionic (propanic) acid ester,[8] trimethyl silanol,[9] hexamethyl disiloxane,[9] cyclosiloxanes (D3-D11),[8-10] trichloroethyl phosphate,[9,11] and phthalates[8,11] [di(2-ethylhexyl)phthalate (DOP) and dibutyl phthalate]. Some of these organic species are considered to be physisorbed[11] from the clean room air[8,12] to the silicon wafer surface. Chemisorption of organic species, such as 1,4-cyclohexadiene,[13] cyclopentene, and cyclohexene,[14] on a silicon (100) surface also has been reported.

Recently, several studies[8,11,15-18] have reported the existence of a time-dependent change in the concentration of organic species on a silicon wafer surface. Some organic species rapidly show a peak of the surface concentration on the silicon wafer surface; afterward they tend to decrease, indicating gradual replacement by the other organic species. Since organic species seem to compete for the adsorption sites on the silicon wafer surface, this behavior is called the "fruit basket phenomenon."[8] Additionally, their surface concentrations have been reported to depend on the condition of the silicon wafer surface.[8] Although the mechanism of this time-dependent phenomenon has been discussed using the heat of adsorption, the heat of vaporization,[8] the boiling point of the organic contaminant,[18] the polarity of the silicon surface,[18] the sticking probability, and the sticking coefficient,[11,19,20] this phenomenon has not been theoretically expressed in a time-dependent form. Since the analysis and prediction of the fruit basket phenomenon are necessary to advance silicon crystal technology, a theoretical model should be developed.

For the first time, this study clarifies the mechanism of the time-dependent change in the surface concentrations of organic species, developing the model of multicomponent organic species adsorption-induced contamination (MOSAIC), which is an application of rate theory for organic species contamination on a silicon wafer surface. Using the MOSAIC model, this study demonstrates that the adsorption rate and the desorption rate of organic species on the silicon wafer surface are the dominant mechanisms of the fruit basket phenomenon.

Numerical Calculation Model

To describe the fruit basket phenomenon of various organic species on the silicon wafer surface, the rates of the change in the surface concentrations of the organic species are expressed based on the single-component Langmuir-type expression[21-23]

$$\frac{\partial \theta}{\partial t} = (1 - \theta)k_{\mathrm{ad}}c - k_{\mathrm{de}}\theta \qquad [1]$$

where θ is the surface coverage of the silicon wafer surface with the organic species, c is the gas-phase concentration of the organic species, t is time, and k_{ad} and k_{de} are the rate constants of adsorption and desorption of the organic species. In this study, this single-component kinetic model is improved to build the concept of competitive adsorption and desorption in a multicomponent system. The following assumptions are used.

1. There is no interaction between any organic molecule in the gas phase and on the silicon wafer surface, such as a chemical reaction, adduct formation, aggregation, and condensation, in the environment which has a very small concentration of organic species.

2. The organic molecules are physisorbed on the silicon wafer surface due to the forces[24,25] of the van der Waals interaction and the electrostatic charges between the organic molecule and the silicon wafer surface.

Assumption 1 is the description of actual molecular motions of organic species existing in a clean room, where the organic species are so dilute that even the low-vapor-pressure organic species can vaporize to the gas phase. Assumptions 1 and 2 indicate that the adsorption rate of the organic molecules on the organic molecules already adsorbed on the silicon wafer surface is much smaller than the adsorption rate directly on the silicon wafer surface. Since silicon wafers are usually used immediately after their surface cleaning, the adsorption of the organic molecules on the organic molecules does not need to be taken into account to study the actual situation in the industrial silicon process. Therefore, assumptions 1 and 2 indicate that the organic molecules physisorb mainly on the vacant site without organic molecules on the silicon wafer surface. In addition to physisorption, chemisorption of the organic molecules also is considered to be expressed in the MOSAIC model as a quite small desorption rate due to its large activation energy.[26]

3. The physisorption rate of the organic molecule of species i is proportional to its concentration in the gas phase and also is proportional to the difference between the surface concentration of the total physisorbed organic species and its larger limit. When the silicon wafer surface is sufficiently covered with organic molecules, no additional organic molecule can physisorb there.

4. The desorption rate of the organic molecules of species i is proportional to their concentration on the silicon wafer surface.

5. The amount of each organic species and that of the larger limit of the total organic species on the silicon wafer surface are evaluated using weight concentration.

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From these, the time-dependent behavior of the concentration of organic species i on the silicon wafer surface, S_i (kg m^{-2}), is expressed in Eq. 2

$$\frac{\partial S_i}{\partial t} = (S_{max} - S)k_{ad,i}C_i - k_{de,i}S_i \qquad [2]$$

where t is time (s), C_i is the concentration of the organic species i in the gas phase above the silicon wafer surface (kg m^{-3}), $k_{ad,i}$ is the physisorption rate constant of the organic species i to the silicon wafer surface (m^3 kg^{-1} s^{-1}), $k_{de,i}$ is the desorption rate constant of the organic species i from the silicon wafer surface (s^{-1}), and S_{max} is the larger limit of the concentration of the total organic species physisorbed on the silicon wafer surface (kg m^{-2}). The existence of S_{max} is considered to be acceptable, since Takahagi *et al.*[2] reported the steady organic film thickness on the silicon wafer surface.

S is the concentration of the total organic species physisorbed on the silicon wafer surface (kg m^{-2}), which is written as follows

$$S = \sum_i S_i + S_{bg} \qquad [3]$$

S_i and S_j ($j \neq i$) can influence each other via S in Eq. 2 and 3. Although the measurement of the concentrations of all the organic species on the silicon wafer surface and in the gas phase is a current goal[27] to be achieved, the influence of the unidentified organic species must be taken into account in Eq. 2. Therefore, the amount of physisorbed organic species which is not clarified in the present calculation is taken into account as the background concentration, S_{bg} (kg m^{-2}), in Eq. 3. Future development of the measurement technique, which can clarify every surface concentration of organic species, enables no use of S_{bg}.

Using S_{max} and S_{bg}, S_e (kg m^{-2}) is defined as follows

$$S_e = S_{max} - S_{bg} \qquad [4]$$

and is the effective larger limit of the concentration of the organic species physisorbed on the silicon wafer surface, which is calculated in this study.

Throughout this calculation, the value of C_i can be assumed to be constant for each organic species, since most of the fabrication process of silicon devices is considered to be performed under steady concentrations of the organic species, which are transported onto the silicon wafer surface following the forced air flow and diffusion in the clean room. Therefore, this study evaluates the values of the product, $k_{ad,i}C_i$, which allows a discussion of the adsorption rate even when C_i is unknown.

Experimental

To evaluate the validity of the MOSAIC model and to discuss the organic contamination from the multicomponent system, the actual behavior reported by Wakabayashi *et al.*[8] is reproduced using Eq. 2 to determine $k_{ad,i}C_i$, $k_{de,i}$, and S_e. These researchers also reported that the measured surface concentrations of propionic acid ester, siloxane (D9), and DOP depended on the condition of the silicon wafer surfaces. Here, the conditions for the sample preparation and the measurement performed by Wakabayashi *et al.*[8] are briefly reviewed.

The samples used were 150 mm diam polished silicon wafers of p-type 10 Ω cm (100), whose surfaces were (a) covered with a silicon dioxide film prepared using ultraviolet light-ozone (UV-O_3) cleaning, (b) covered with a thermal oxide film and cleaned using 1% hydrogen fluoride aqueous solution, and (c) terminated with hydrogen using wet cleaning with 1% hydrogen fluoride aqueous solution. In this study, the time-dependent changes in the concentration of the organic species on the silicon wafer surface for conditions (a)-(c) are calculated.

The concentration of the organic species on the silicon wafer surface were quantitatively measured using thermal desorption-gas chromatography-mass spectrometry, GL Sciences, Inc.). The calibration curve of the organic species was obtained by measuring a known amount of hexadecane on the silicon wafer surface. The detection limit of this measurement was 6×10^{-10} kg, m^{-2} (conversion from hexadecane). The influence of the matrix on the measurement was assumed to be negligible in this study.

The surfaces of the silicon wafers were exposed to air in a clean room of class 10 with a forced down flow stream using a fan filter unit.

Results

To evaluate the validity of the MOSAIC model, numerical calculations using Eq. 2 are performed to reproduce the measured concentrations[8] of the organic species on the silicon wafer surface.

The circles, squares, and triangles in Fig. 1 show the measured concentrations of the organic species of propionic acid ester, siloxane (D9), and DOP, respectively, on (a) the surface of silicon oxide film cleaned using UV-O_3, (b) the surface of silicon oxide film formed by thermal oxidation, and (c) the hydrogen-terminated silicon surface.

As shown by the circles, squares, and triangles in Fig. 1a-c, the measured surface concentration of each organic species shows the same trend among these three silicon wafer surface conditions. The surface concentration of propionic acid ester rapidly increases after the surface cleaning to a peak and then shows a gradual decrease. Although the surface concentration of siloxane (D9) increases simultaneously, its value remains very small. The surface concentration of DOP begins to increase immediately after the preparation of the silicon wafer surface and continues to increase even 69 h (248, 400 s) after the surface cleaning.

The solid lines in Fig. 1a-c show the surface concentrations of the organic species on the silicon wafer surfaces, calculated using the

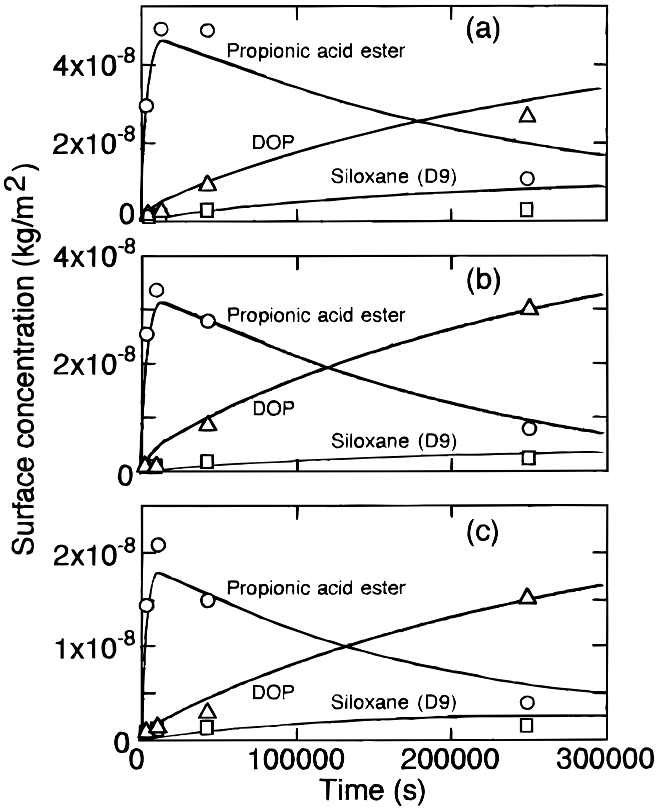


Figure 1. Measured and calculated surface concentrations of the organic species of propionic acid ester, siloxane, and di(2-ethylhexyl)phthalate (DOP), on (a) the surface of silicon oxide film cleaned using ozone under ultraviolet light, (b) the surface of silicon oxide film formed by thermal oxidation, and (c) the silicon wafer surface cleaned using dilute hydrogen fluoride aqueous solution. The circles, squares, and triangles are measured values[8] of propionic acid ester, siloxane (D9), and DOP, respectively. The solid lines are those calculated in this study.

Table I. Rate parameters for organic species obtained in this study.

Silicon water surface	Organic species	$k_{ad,i}C_i$ (s^{-1})	$k_{de,i}$ (s^{-1})	S_e (kg m^{-2})
(a) UV-O_3 oxide	Propionic acid ester	3×10^{-4}	6×10^{-5}	6×10^{-8}
	Siloxane (D9)	4×10^{-6}	0	
	DOP	2×10^{-5}	0	
(b) Thermal oxide	Propionic acid ester	3×10^{-4}	9×10^{-5}	4.6×10^{-8}
	Siloxane (D9)	2×10^{-6}	0	
	DOP	2×10^{-5}	0	
(c) H-terminated by dilute HF	Propionic acid ester	4×10^{-4}	1×10^{-4}	2.5×10^{-8}
	Siloxane (D9)	7×10^{-6}	4×10^{-6}	
	DOP	2×10^{-5}	0	

MOSAIC model. The values of $k_{ad,i}C_i$, $k_{de,i}$, and S_e are obtained as listed in Table I, using the least-squares method. Figure 1 shows that the calculated time-dependent change in the concentrations of the organic species agrees with the measurement and also indicates that the numerical calculations of very small values of organic species concentrations are possible.

Discussion

Rate parameters and surface concentration.—First, from the results obtained in the previous section, it can be considered that the fruit basket phenomenon[8,11,15,16] of organic species occurs following the balance between the physisorption rate and the desorption rate expressed in Eq. 2.

Next, the dominant mechanism of the fruit basket phenomenon is discussed in this section, focusing on the role of the rate parameters of the organic species. Since the behavior of the single-component physisorption gives the fundamental information for discussing the multicomponent system, the physisorption of the organic species on the silicon wafer surface assuming a single-component system is calculated and shown in Fig. 2, using the rate parameters of surface (b) in Table I. The behavior shown in Fig. 2 is expected to occur in an environment having only a single organic species. Therefore, the time-dependent change in the concentration of the organic species on the silicon wafer surface is discussed using Fig. 1 and 2.

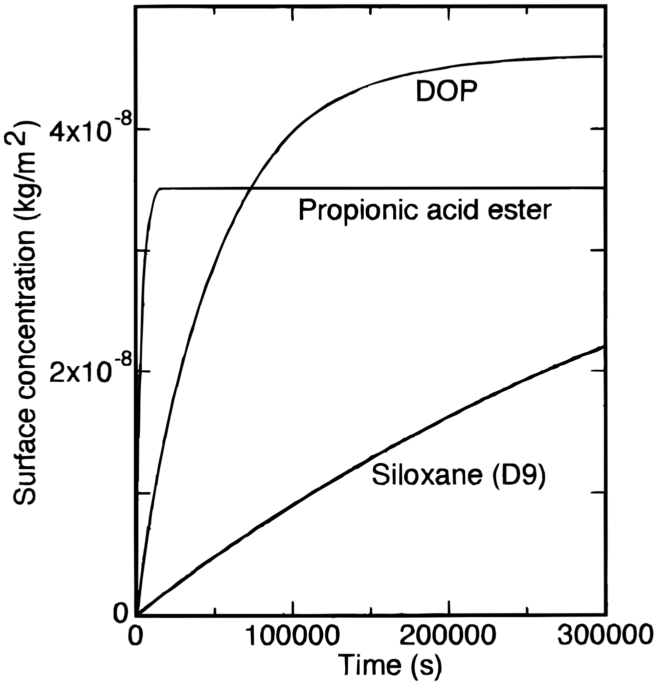


Figure 2. Calculated concentration of propionic acid ester, siloxane (D9), and di(2-ethylhexyl)phthalate (DOP) on the surface of silicon oxide film formed by thermal oxidation, following single-component physisorption and desorption using the rate parameters of surface (b) in Table I.

The rate parameters $k_{ad,i}C_i$, and $k_{de,i}$ listed in Table I show the following relationships

$$\text{propionic acid ester} > \text{DOP} > \text{siloxane (D9), for } k_{ad,i}\, C_i \quad [5]$$

$$\text{propionic acid ester} > \text{siloxane (D9)} \simeq \text{DOP, for } k_{de,i} \quad [6]$$

$$k_{ad,i}\, C_i > k_{de,i}, \text{ for each organic species} \quad [7]$$

Propionic acid ester has greater values of both $k_{ad,i}\, C_i$, and $k_{de,i}$ than those of siloxane (D9) and DOP. The surface concentration of propionic acid ester increases immediately due to its large $k_{ad,i}\, C_i$; the large $k_{de,i}$ quickly causes the surface concentration to reach the steady state, as shown in Fig. 2. This behavior is considered to be fundamentally the same as that in the multicomponent system shown in Fig. 1. Propionic acid ester physisorbs on the silicon wafer surface faster than the other organic species after cleaning of the silicon wafer surface. However, since the large desorption rate of propionic acid ester supplies the vacant site of the silicon wafer surface in order to share competitively with all of the organic species existing in the gas phase following their physisorption rates, propionic acid ester shows a gradual decrease in its surface concentration after reaching its peak.

In contrast to propionic acid ester, siloxane (D9) has small values of both $k_{ad,i}C_i$ and $k_{de,i}$, which indicates that a very long period is necessary to reach the steady state of the surface concentration, as shown in Fig. 2. The surface concentration of siloxane (D9) in the multicomponent system shown in Fig. 1 continues to increase to occupy a small area of the silicon wafer surface, as also shown in Fig. 2.

The value of $k_{ad,i}C_i$ of DOP is smaller than that of propionic acid ester and is greater than that of siloxane (D9). The $k_{de,i}$ of DOP is as small as that of siloxane (D9). These parameters predict that the amount of physisorbed DOP on the silicon wafer surface increases to be greater than that of propionic acid ester. As shown in Fig. 1 and 2, the surface concentration of DOP increases more slowly than that of propionic acid ester and more rapidly than that of siloxane (D9) to occupy a very large area of the silicon wafer surface. Since the decrease in the surface concentration of propionic acid ester is considered to be induced by the large $k_{de,i}$ both a large $k_{ad,i}C_i$ and small $k_{de,i}$ are the dominant conditions for an organic species to finally occupy the largest area of the silicon wafer surface.

Next, the dependence of the rate parameters on the condition of the silicon wafer surface is discussed. Table I shows that $k_{ad,i}C_i$ and $k_{de,i}$ for each organic species change slightly with the condition of the silicon wafer surface. The values of $k_{ad,i}C_i$ for each organic species are nearly constant for the surfaces of (a)-(c), that is, 3-4 $\times$ 10^{-4} s^{-1} for propionic acid ester, 2-7 $\times$ 10^{-6} s^{-1} for siloxane (D9), and 2×10^{-5} s^{-1} for DOP. Therefore, $k_{ad,i}C_i$ changes very little with the condition of the silicon wafer surface. The variation of the values of $k_{de,i}$ for the surfaces of (a)-(c) is also very small, that is, 6×10^{-5} to 1 $\times$

10^{-4} s^{-1} for propionic acid ester, less than 4×10^{-6} s^{-1} for siloxane (D9), and 0 s^{-1} for DOP. From the discussion in this section, it is concluded that $k_{ad,i}C_i$ and $k_{de,i}$ reflect mainly the property of the organic species rather than the condition of the silicon wafer surface. It would be interesting to discuss $k_{ad,i}C_i$ and $k_{de,i}$ in relation to the heat of adsorption, the heat of vaporization,[8] the boiling point, the polarity of the organic species,[18] and the steric hindrance between the organic species; however, this is left for future study.

The rate parameter of S_e is also discussed here. The values of S_e obtained in this study show the relationship as follows

$$(a) > (b) > (c) \qquad [8]$$

The difference between the values of S_e listed in Table I seems to be as small as that of $k_{ad,i}C_i$ and $k_{de,i}$. However, since the relationship of S_e in Eq. 8 is similar to that of the peak height of the surface concentration of the organic species in Fig. 1, and since S_e directly influenced the surface concentration of each organic species in the calculations, S_e is a very sensitive parameter for indicating the difference in the condition of the silicon wafer surface. Therefore, the MOSAIC model is considered to be effective for discussing the influence of the silicon surface condition.

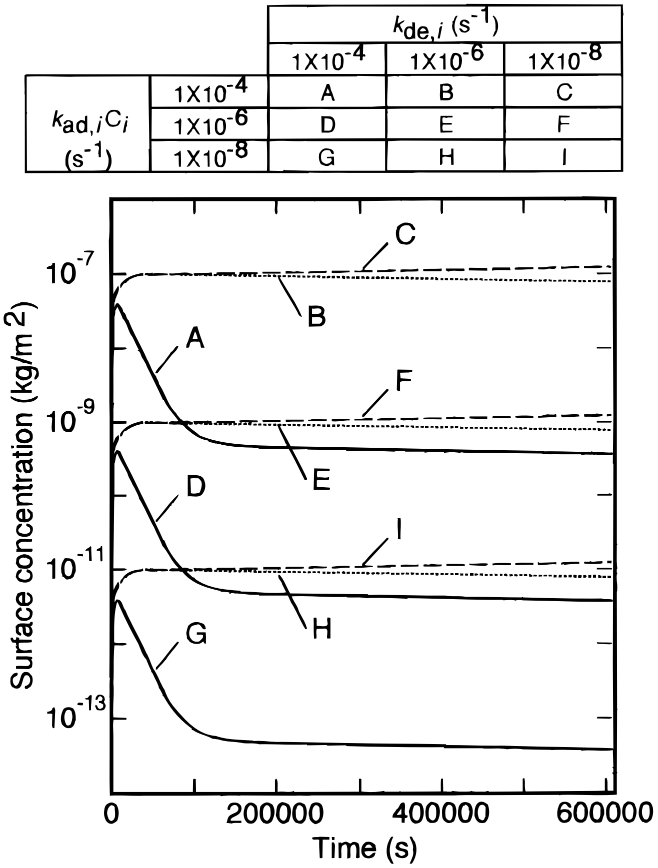
For discussing S_e in detail, note here that the change in both S_{max} and S_{bg} can affect S_e, as defined in Eq. 4. The change in S_{max} indicates the increase or the decrease in the concentration of organic species capable of being physisorbed on the silicon wafer surface. S_{max} can be influenced by factors[24,25] such as (*i*) the surface area effective for the physisorption of organic species, (*ii*) the electrostatic condition of the silicon wafer surface, and (*iii*) properties of the organic species, such as their polarity[18] and their steric hindrance. A further study of these factors will be effective for understanding the physisorption and the desorption of the organic species on the silicon wafer surface. Just like S_{max}, S_e is influenced by S_{bg} due to factors (*i*), (*ii*), and (*iii*). When the amount of an organic species other than those accounted for in this study increases, S_{bg} increases to give a small S_e. Therefore, the measurement of all the organic species on the silicon wafer surface and in the gas phase is necessary to clarify the trend of S_{max} and S_e caused by the condition of the silicon wafer surface. Additionally, knowing C_i will enable the comparison of the rate parameters of $k_{ad,i}C_i$ and $k_{de,i}$ obtained in this study with the sticking probabilities reported in previous studies,[11,20] since $S_e k_{ad,i}$ is considered to be based on the same physical concept as the sticking probability.

In addition to the exposure of the silicon wafer surface to the forced air flow in a clean room evaluated in this study, the MOSAIC model is considered to be capable of evaluating closed ambients, such as that in a wafer box, where the organic species are transported predominantly by diffusion.

Physisorption and desorption in multicomponent system.—To further discuss the properties of various organic species for the fruit basket phenomenon, the influences of $k_{ad,i}C_i$ and $k_{de,i}$ on the trend of the concentration of the organic species on the silicon wafer surface are evaluated using the MOSAIC model. Here, nine organic species, A-I, having various values of $k_{ad,i}C_i$ and $k_{de,i}$, and with a fixed value of S_e, are assumed to exist in the multicomponent system. The calculated surface concentrations are shown in Fig. 3.

The organic species with large $k_{ad,i}C_i$ have large surface concentrations as shown by species A-C in Fig. 3. The organic species having small $k_{ad,i}C_i$, species G-I, always have small surface concentrations as also shown in the Fig. 3. Therefore, the abundance of the organic species on the silicon wafer surface is mainly determined by $k_{ad,i}C_i$.

The organic species A, D, and G, which have large $k_{de,i}$, exhibit the steep peak of their surface concentration, similar to propionic acid ester in Fig. 1; even their $k_{ad,i}C_i$ values distribute from very large to small. Species B, E, and H, with a medium value of $k_{de,i}$, have surface concentrations that decrease very gradually after showing their peak. Species C, F, and I have the smallest values of $k_{de,i}$ in Fig. 3. Species C, F, and I increase immediately like species B, E, and H; however, their surface concentrations continue to increase very gradually. Although species C, F, and I have various $k_{ad,i}C_i$, from very large to small values, this increasing trend is fundamentally the same among these species.



		$k_{de,i}$ (s^{-1})		
		1X10^{-4}	1X10^{-6}	1X10^{-8}
$k_{ad,i}C_i$ (s^{-1})	1X10^{-4}	A	B	C
	1X10^{-6}	D	E	F
	1X10^{-8}	G	H	I

Figure 3. Prediction of surface concentrations of nine organic species, A-I, having various $k_{ad,i}C_i$ and $k_{de,i}$ values, with a fixed value of S_e, 2×10^{-7} kg m^{-2}. Solid lines are organic species A, D, and G; dotted lines are B, E, and H; broken lines are C, F, and I.

Note here from Fig. 3 that the surface concentration of species A finally became lower than those of species E and F. Species D also seems to be replaced by species H and I on the silicon wafer surface, a situation similar to the fruit basket phenomenon in Fig. 1.

From the calculations of the competitive physisorption and the desorption shown in Fig. 3, the condition of the organic species which results in their greatest abundance on the silicon wafer surface is concluded to be dependent on both the largest adsorption rate and the smallest desorption rate. Therefore, the rate parameters of the organic species are expected to be effective for designing the preparation method of the silicon wafer surface throughout the fabrication of silicon devices.

Conclusions

To clarify the time-dependent change in the concentration of organic species on a silicon wafer surface, called the fruit basket phenomenon,[8] a rate theory of physisorption and desorption is applied to build a theoretical model of multicomponent organic species adsorption-induced contamination (MOSAIC). The actual increase and decrease in the surface concentrations of propionic acid ester, siloxane (D9), and DOP are described and predicted using an equation of the MOSAIC model composed of the adsorption rate from the gas phase to the silicon wafer surface and the desorption rate from the silicon wafer surface. Although the surface concentrations of the organic species initially increase following their physisorption rate, the organic species having small desorption rates finally achieve a large concentration on the silicon wafer surface.

Since the MOSAIC model can reproduce the measured behavior of the concentrations of the organic species, the rate parameters of the physisorption and the desorption obtained using the MOSAIC model are expected to be effective for systematic study and practical design of a preparation method of a silicon wafer surface.

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