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ORIGINAL PAPER

Oxidation of Methyl Linoleate in Oil-in-Water Micro- and Nanoemulsion Systems

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Abstract Oxidation of methyl linoleate in O/W emulsions having droplets of median diameters ranging from 17 nm to 8.0 µm was carried out at 40°C. The oxidation process was analyzed on the basis of a kinetic equation of the autocatalytic type. The induction period was found to be shorter and the oxidation rate constant lower for emulsions with smaller oil droplets. The stoichiometry between methyl linoleate and oxygen was observed to be independent of both the size of oil droplet and the type of the surfactant and was found to be unity during the early stage of the oxidation. However, more oxgen was consumed in the oxidation of the methyl linoleate in the later half of the oxidation process.

Keywords Nanoemulsion · Lipid oxidation · O/W emulsion · Methyl linoleate

Introduction

Lipid oxidation is one of major causes for the deterioration of foods containing lipids, and it has been extensively investigated [1]. An emulsion is a common structure in lipid-containing foods from home cooking to industry. Many factors, such as the addition of an antioxidant [2–5], protein [6–8], metal ion [9] and chelator [5, 10], emulsifier type [10, 11], and homogenization conditions [12, 13], affect the lipid oxidation in the emulsion system. Oil-droplet size of the emulsion is also an important factor affecting the oxidation [11, 13–15]. Although the oil-droplet sizes were in almost the same range (from 0.2 to 30 µm) in the studies, their conclusions were quite conflicting. Lethuaunt et al. [14] reported that the rates of oxygen consumption and conjugated diene formation were higher for the small oil-droplets. On the other hand, Nakaya et al. [15] reported that the hydroperoxide content in the emulsion with a smaller droplet size was significantly lower than that in the emulsion containing large droplets. However, Osborn and Akoh [11] concluded that the oil-droplet size had no effect on the lipid oxidation in structured lipid-based oil-in-water emulsions.

Rapid developments in nanotechnology have allowed us to focus on its application to foods. A nanoemulsion has several interesting properties. The emulsion looks bluish transparent because the droplets are too small to scatter visible light. Droplets in the emulsion have much higher specific surface areas than the common emulsion that has a larger droplet size. The high surface area would elicit an effect at the interface between the oil and aqueous phases on phenomena occurring in the emulsions, including the lipid oxidation.

In this context, we prepared oil-in-water (O/W) emulsions having the oil-droplet sizes of 17 nm to 8.0 µm in the same compositions of oil and surfactant in order to exclude the effects of the composition on the oxidation. Methyl linoleate and decaglyceryl laurate were used as the oil phase and surfactant, respectively. The oxidation processes of methyl linoleate in the emulsions were measured at 40°C in order to examine the effect of the oil-droplet size on the oxidation kinetics over a wider range of oil-droplet size compared to the previous studies [11, 13–15]. The results obtained here will conclude the effect and show superiority of nanoemulsion for suppression of lipid oxidation in an emulsion system.

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Materials and Methods

Materials

Methyl linoleate was purchased from Tokyo Chemical Industry (Tokyo, Japan). Methyl palmitate, which was used as the internal standard for the gas chromatographic analysis of unoxidized methyl linoleate, hexane, chloroform, and methanol were purchased from Wako Pure Chemical Industries (Osaka, Japan). The hydrophilic surfactants, Sunsoft® Q-12Y and SY-Glyster® ML-750, were supplied by Taiyo Kagaku (Yokkaichi, Japan) and Sakamoto Yakuhin Kogyo (Osaka, Japan), respectively. They were both categorized as decaglycerol monolaurate.

Preparation of Emulsions having Different Oil-Droplet Diameters

Table 1 lists the conditions for preparing the O/W emulsions with various oil-droplet diameters using the Q-12Y surfactant. Phosphate buffer (50 mmol/L, pH 7.4) was used as the water phase. The O/W emulsions using the ML-750 surfactant were also prepared under similar conditions. A surfactant was dissolved in purified water at 20 or 30% (w/v). Methyl linoleate was added to the surfactant solution. The mixture was then homogenized using a rotor/stator homogenizer (Polytron PT20SK, Kinematica, Lucern, Switzerland). In order to produce emulsions having small oil droplets, a coarse emulsion was further homogenized twice using a high-pressure homogenizer (PEL-20, Nanomizer, Tokyo, Japan) at 1,500 kg-force/cm^2. The O/W emulsions were diluted with a phosphate buffer. Sodium azide was also added to the emulsion at 1 mmol/L to prevent microbial growth. The concentrations of the surfactant and methyl linoleate were 3% (w/v) and 0.75% (v/v), respectively, for all the emulsions. The diameter of the oil droplets in the emulsions was measured using a DLS-7000 dynamic light scattering spectrophotometer (Otsuka Electronics, Osaka, Japan) or a SALD-2100 laser diffraction particle size analyzer (Shimadzu, Kyoto, Japan) depending on the size of the oil droplets.

Oxidation of Methyl Linoleate in Emulsion

The emulsion (4.5 mL) was placed in 100-mL amber vials and each vial was tightly sealed with a silicone cap. The vials were maintained at 40°C in a DN-400 thermo-controlled oven (Yamato Scientific, Tokyo, Japan). The emulsions in the vials were mixed with a magnetic stirrer at 600 rpm in order to promote the supply of oxygen from gas phase to the emulsion phase and to prevent creaming of the emulsions. At appropriate intervals, one of the vials was removed from the oven to measure the amount of unoxidized methyl linoleate using a gas chromatograph.

Measurement of the Amount of Unoxidized Methyl Linoleate

To a sampled emulsion (400 μL) in a disposable sample tube was added 1,000 μL of chloroform:methanol = 2:1 (v/v). The sample tube was vigorously shaken by hand and then vigorously vortex-mixed. The mixture was centrifuged at 17,600×*g*, where *g* is the acceleration of gravity, for 5 min (MC-150, Tomy Seiko, Tokyo, Japan). The supernatant was discarded, and 600 μL of the lower-phase solution was transferred to another sample tube. The solvent in the solution was completely removed under reduced pressure. The residue was dissolved in 300 μL of hexane containing 112.5 mmol/L methyl palmitate, which was an internal standard for the gas chromatographic determination, by vortex-mixing. The solution was centrifuged at 17,600×*g* for 5 min, and 1.0 μL of the supernatant was analyzed by gas chromatography to determine the unoxidized methyl linoleate. The gas chromatograph was a GC-14BPF (Shimadzu, Kyoto, Japan) equipped with a capillary column (J&W Scientific DB-1; length 30 m, inner diameter

Table 1 Conditions for preparing O/W emulsions with different initial diameters of oil droplets using the Q-12Y surfactant

Initial median high-pressure homogenizer diameter of oil droplets [μm]	Surfactant[a] [% (w/v)]	Methyl linoleate[a] [% (v/v)]	Rotor/stator homogenizer		High-pressure homogenizer	
			Rotational speed [rpm]	Duration time [min]	Pressure [kg-force/cm^2]	Passage number
0.041	30	7.5	2.0×10^4	5	1.5×10^3	2
0.043	30	7.5	2.0×10^4	5	1.5×10^3	2
0.052	30	7.5	2.0×10^4	5	1.5×10^3	2
0.079	30	7.5	2.0×10^4	5	–	–
0.92	30	7.5	1.0×10^4	5	–	–
1.7	30	7.5	7.0×10^3	3	–	–
5.9	20	5.0	3.0×10^3	3	–	–

[a] The final concentrations of surfactant and methyl linoleate were 3% (w/v) and 0.75% (v/v), respectively, in all the emulsions

0.25 mm, film thickness 0.25 μm, Agilent Technologies, Santa Clara, CA, USA) and a flame ionization detector. Temperatures of the column, injector and detector of the gas chromatograph were 220, 250, and 270°C, respectively. The chromatogram was recorded by an integrator (C-R6A, Shimadzu).

Measurement of Partial Pressure of Oxygen During Lipid Oxidation

The emulsion (500 μL) was poured into an amber vial and the vial was tightly sealed with a lid equipped with a UC-12 oxygen sensor (Central Kagaku, Tokyo, Japan). The vial was maintained at 40°C in a thermo-controlled oven. The volume of the gas phase in the vial was 119.7 mL, and the emulsion in the vial was also mixed by a magnetic stirrer at 600 rpm during the measurement. The partial oxygen pressure was recorded using a strip-chart recorder (Type 201, Asahi Kagaku, Tokyo, Japan) during the oxidation.

Results and Discussion

Oxidation of Methyl Linoleate in Emulsions

Figure 1a and b show the oxidation processes of methyl linoleate in the emulsions, which were prepared using Q-12Y and ML-750, respectively, as the surfactant. The emulsions contained oil droplets of different initial sizes. The oil-droplet size significantly affected the oxidation process for the emulsions prepared using the Q-12Y surfactant. There was a tendency that the period of the induction step was longer and the oxidation rate during the propagation step was higher for the emulsion containing the larger droplet. On the other hand, the effect of the initial oil-droplet size on the oxidation process was not remarkable for the emulsions prepared using the ML-750 surfactant.

The oxidation process of methyl linoleate in the emulsion having any oil-droplet size was analyzed on the basis of the rate equation of the autocatalytic type expressed by Eq. 1 [16, 17]:

$$\frac{dY}{dt} = -kY(1 - Y) \tag{1}$$

where Y is the fraction of unoxidized methyl linoleate, t is the time, and k is the rate constant. The integration of Eq. 1 under the initial condition of $Y = Y_0$ at $t = 0$ gives Eq. 2:

$$\ln\frac{1 - Y}{Y} = kt + \ln\frac{1 - Y_0}{Y_0} \tag{2}$$

where Y_0 is the initial fraction of unoxidized methyl linoleate, which is conveniently introduced to solve Eq. 1 and reflects the initial state of the methyl linoleate being used.

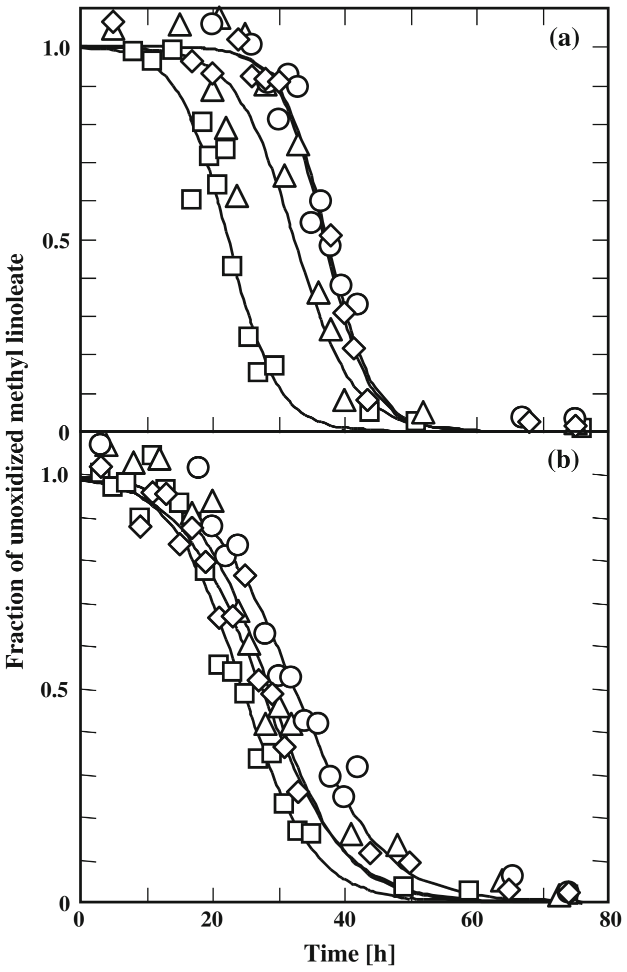

Fig. 1 Oxidation processes of methyl linoleate in O/W emulsions prepared using **a** Sunsoft® Q-12Y and **b** SY-Glyster® ML-750 at 40°C. The initial median diameters of oil droplets in the emulsions were **a** 43 nm (*open square*), 79 nm (*open triangle*), 0.92 μm (*open circle*), and 5.9 μm (*open diamond*), and **b** 17 nm (*open square*), 73 nm (*open triangle*), 0.66 μm (*open circle*), and 7.2 μm (*open diamond*)

Figure 2a and b show a plot of ln[(1−Y)/Y] versus time t for the oxidation processes shown in Fig. 1a and b, respectively. For any emulsion, the plots of ln[(1−Y)/Y] versus t lie on a straight line during the early stage of oxidation of methyl linoleate. The rate constant k was estimated from the slope of the line. The Y_0 value was also estimated from the intercept of the line. The solid curves in Fig. 1 were drawn using the estimated k and Y_0 values.

Effect of the Initial Oil-Droplet Size of Emulsion on the Oxidation Behavior

The induction period t_{ind} for the methyl linoleate was defined as the time at wich the tangent at $Y = 0.5$ crossed the line of $Y = 1$, that can be calculated from the k and Y_0 values using Eq. 3:

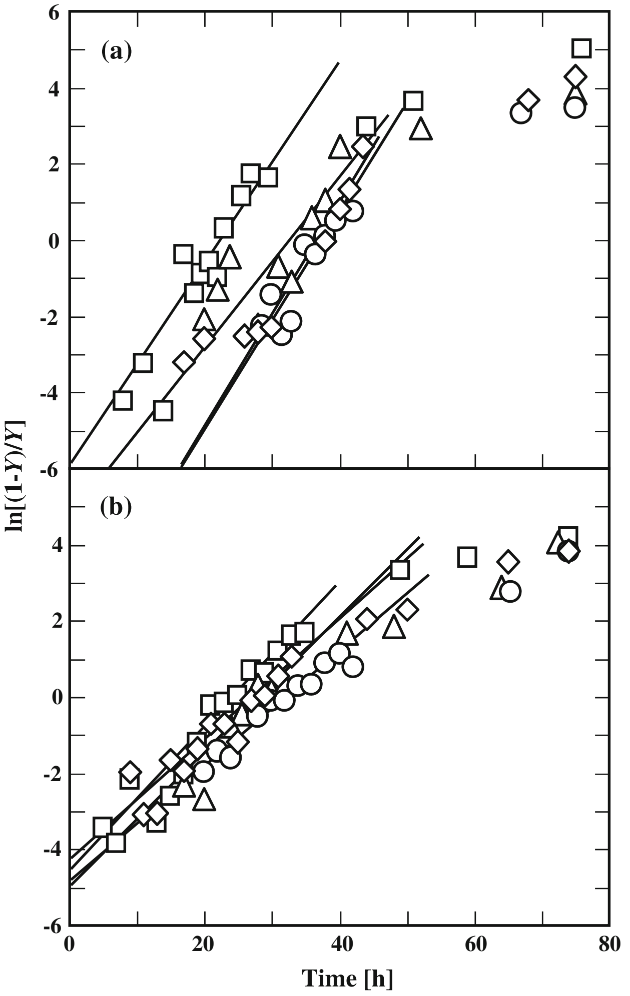

Fig. 2 A plot of ln[(1−*Y*)/*Y*] versus time based on Eq. 2 for the oxidation processes of methyl linoleate in O/W emulsions prepared using **a** Sunsoft® Q-12Y and **b** SY-Glyster® ML-750. The symbols are the same as those in Fig. 1

$$t_{\text{ind}} = \frac{1}{k}\left(\ln\frac{Y_0}{1-Y_0} - 2\right) \tag{3}$$

Figure 3 shows the dependencies of the k and t_{ind} values on the initial diameter of the oil droplets in the emulsions. For the emulsions prepared using the Q-12Y surfactant, both the k and t_{ind} values were smaller for the emulsions having smaller oil droplets. On the other hand, there were no significant dependencies of the k and t_{ind} values on the initial oil-droplet size of the emulsions prepared using the ML-750 surfactant. As shown later, the oil-droplet size hardly changed for the emulsions prepared using the Q-12Y surfactant, but it significantly changed for those prepared using the ML-750 surfactant. The difference in the stability between the emulsions prepared using the Q-12Y and ML-750 surfactants seemed to result in the different dependencies of the k and t_{ind} values on the initial oil-droplet size.

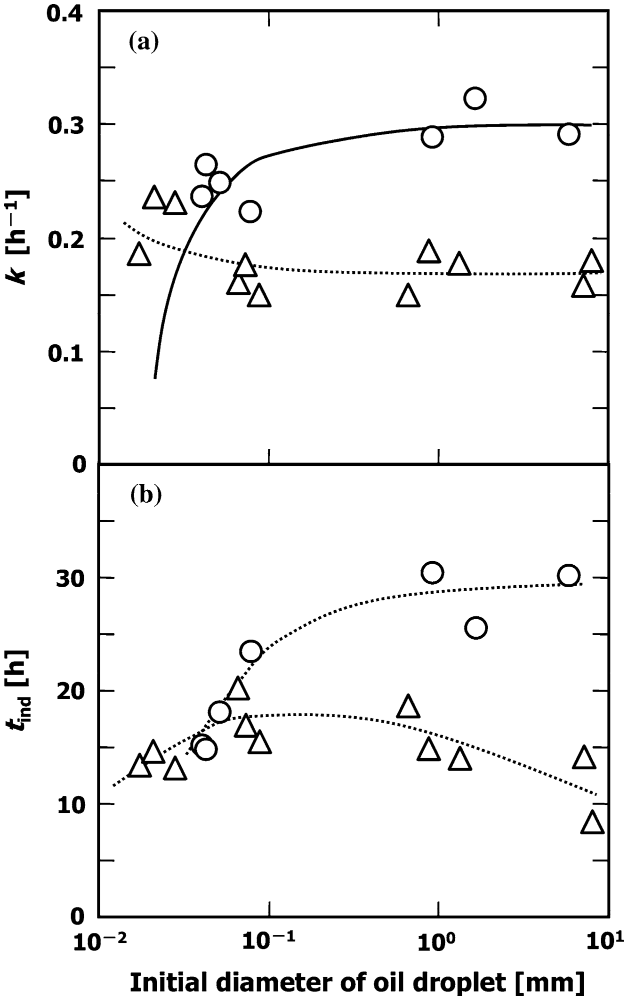

Fig. 3 Dependencies of the kinetic parameters, k (*open circle*, *open triangle*) and t_{ind} (*open circle*, *open triangle*), for the oxidation of methyl linoleate on the initial oil-droplet size in O/W emulsions prepared using (*circle*) Sunsoft® Q-12Y and (*triangle*) SY-Glyster® ML-750. The *solid curve* for Sunsoft® Q-12Y was drawn based on the proposed model, while *dotted curves* were drawn by smoothly connecting the experimental points

A Dilution Effect of Substrate by Hydrophobic Moiety of Surfactant

The specific surface area of the oil-droplet is higher for the smaller oil-droplets. The increase in the specific surface area seems to promote the oxidation of methyl linoleate when the diffusion of oxygen through the interface is rate-limiting or to maintain the k value constant when the rate-limiting step is the oxidation reaction itself. However, the k value was smaller for the emulsion containing smaller oil-droplets.

As previously described, Nakaya et al. [15] explained the slower oxidation of the lipid for the O/W emulsions with smaller oil-droplet sizes by the wedge effect of the hydrophobic acyl residues of the surfactant. The wedge was supposed to suppress the mobility of the acylglycerol molecules migrating in the oil phase. The supposition was based on ^{1}H NMR measurements.

We now propose another model which describes the dependence of the *k* value on the oil-droplet size of the emulsion. This model is based on the property of the autocatalytic-type rate expression and dilution of methyl linoleate by a hydrophobic moiety of the surfactant. The applicability of the model will be examined for the emulsions prepared using the Q-12Y surfactant because they were stable without any significant change in the oil-droplet size during the oxidation.

The rate equation of the autocatalytic type (Eq. 1) is derived under the assumption that the oxidation rate is proportional to the product of the concentration of the remaining substrate and that of the reacted one:

$$\frac{\mathrm{d}C_\mathrm{S}}{\mathrm{d}t} = -k'C_\mathrm{S}(C_\mathrm{S0} - C_\mathrm{S}) \tag{4}$$

where C_S is the substrate concentration at time *t*, C_S0 is the initial concentration of the substrate, and k' is a rate constant. The normalization of C_S by C_S0 gives Eq. 1, and the rate constants, *k* and k', are related to each other by the following equation [18]:

$$k = k'C_\mathrm{S0} \tag{5}$$

Because the k' value is intrinsic at a specific temperature, the *k* value is proportional to the initial substrate concentration, C_S0.

Surfactant molecules cover the surface of the oil droplets, and the surfactant molecule penetrates its hydrophobic moiety into the oil phase. The hydrophobic moiety of the Q-12Y surfactant is the lauroyl residue, which is a saturated acyl chain. The penetration of the saturated lauroyl residue into the methyl linoleate phase should dilute the substrate, which would result in the lowering of the *k* value based on Eq. 5.

Assuming that an oil-droplet having a diameter D_O is fully covered with surfactant molecules to form the hydrophilic layer, the thickness of which is D_H, that the volume of a hydrophobic moiety is V_L, and that the residual area per surfactant molecule is *a*. Based on these assumptions, the *k* value for an oil-droplet covered with surfactant is given by Eq. 6:

$$\frac{k}{k_0} = 1 - \frac{6V_\mathrm{L}}{a(D_\mathrm{O} - 2D_\mathrm{H})} \tag{6}$$

where k_0 is the rate constant for methyl linoleate which is not covered by the surfactant.

The hydrophilic and hydrophobic moieties of the Q-12Y surfactant are the decaglyceryl and lauroyl residues, respectively. Based on Kopp's rule [19], their volumes were estimated to be 1.53 and 0.41 nm^3, respectively. The cross-sectional area of the HCOH group in the hydrophilic moiety was also estimated to be 0.3 nm^2 based on the rule. The cross-sectional area is assumed to be the same as the *a* value, and the D_H value was evaluated to be 5 nm by dividing the volume of the hydrophilic moiety by its cross-sectional area. The k_0 value was roughly estimated to be 0.3 h^{-1} from the *k* values for the microemulsions. The *k* values for the emulsions having various oil-droplet sizes were calculated by substituting the estimated V_L, *a*, D_O, D_H, and k_0 values into Eq. 6, and are shown in Fig. 3a by the solid curve. This curve expressed the experimental results well. This fact indicates that the dilution of the substrate oil by the saturated hydrophobic moiety of the surfactant plays an important role in the smaller *k* value for the emulsions having smaller oil droplets.

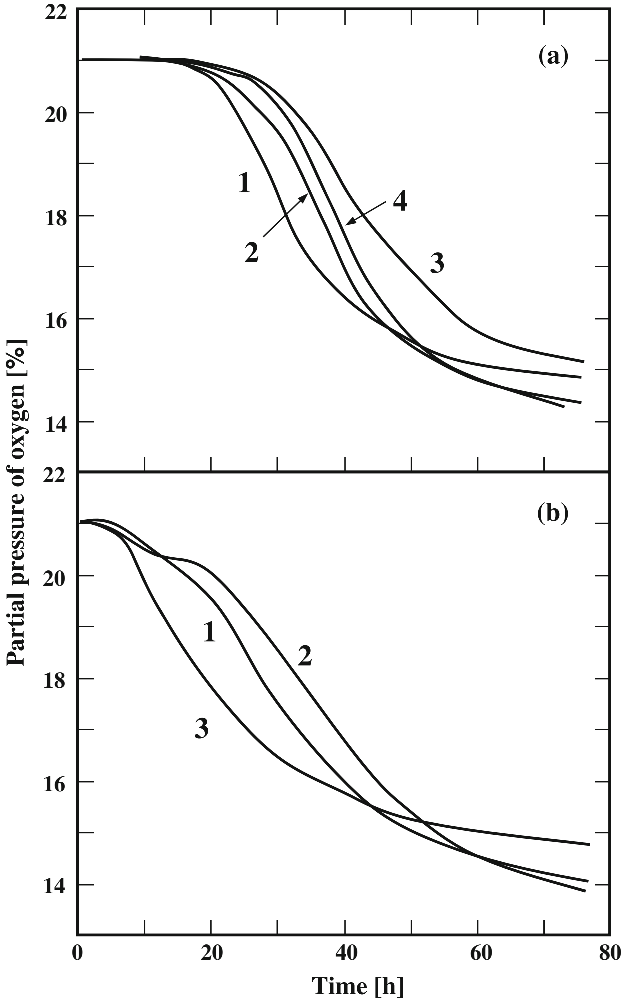

Fig. 4 Changes in the partial pressure of oxygen during the oxidation of methyl linoleate in O/W emulsions prepared using **a** Sunsoft® Q-12Y and **b** SY-Glyster® ML-750 at 40°C. The initial median diameters of the oil droplets in the emulsions were **a** 52 nm (*label 1*), 79 nm (*2*), 0.92 µm (*3*), and 1.7 µm (*4*), and **b** 17 nm (*1*), 88 nm (*2*), 1.3 µm (*3*)

Consumption of Oxygen During Oxidation of Methyl Linoleate

Figure 4a and b show the changes in the partial pressure of oxygen during the oxidation of methyl linoleate in the emulsions prepared using the Q-12Y and ML-750 surfactants, respectively. The oxygen consumption behavior was similar to the decrease in the fraction of the remaining methyl linoleate.

The molar amount of the consumed oxygen was estimated on the basis of the ideal gas law, and the molar amount of the consumed methyl linoleate was calculated from the fraction of the remaining substrate. Figure 5 shows the stoichiometric relationship between the oxidized substrate and the consumed oxygen for the emulsions with various initial oil-droplet diameters. The emulsions were prepared using the Q-12Y and ML-750 surfactants. All the plots lie on a curve. The plots during the early stage of the oxidation lie on a straight line with the slope of unity, indicating that the stoichiometric coefficient between methyl linoleate and oxygen was unity. This result agreed with our previous one for the oxidation of methyl linoleate in a bulk system [20]. During the later stage of oxidation, more oxygen was consumed in order to oxidize the methyl linoleate. Both the oil-droplet size and the surfactant type did not affect the stoichiometric relationship.

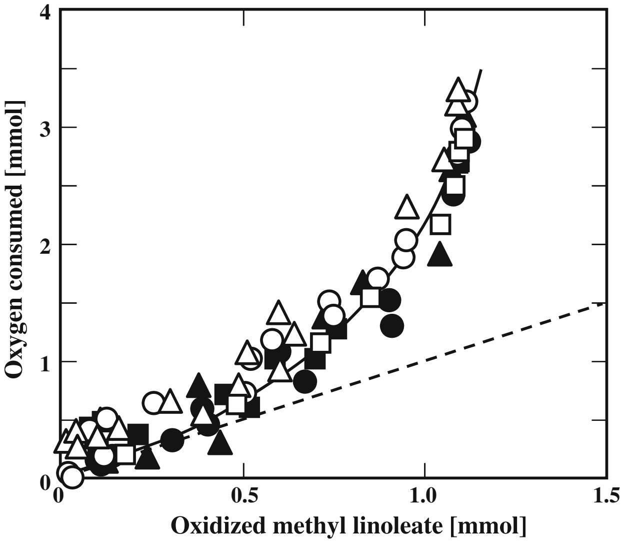

Fig. 5 Relationship between the amount of consumed oxygen and that of oxidized methyl linoleate during the oxidation of methyl linoleate on the initial oil-droplet size in O/W emulsions prepared using (*closed symbols*) Sunsoft® Q-12Y and (*open symbols*) SY-Glyster® ML-750. The initial median diameters of the oil droplets in the emulsions were 52 nm (*filled circle*), 79 nm (*filled circle*), 0.92 µm (*filled square*), 17 nm (*open circle*), 88 nm (*open triangle*), and 1.3 µm (*open square*)

Change in Oil-Droplet Size During Oxidation of Methyl Linoleate

Figure 6a and b show the changes in the oil-droplet size of the emulsions prepared using the Q-12Y and ML-750 surfactants. The emulsions had various initial oil-droplet sizes. Irrespective of the initial oil-droplet size, the oil-droplet sizes of the emulsions prepared using the Q-12Y surfactant hardly changed except during the later stage of the storage in which the methyl linoleate was almost completely oxidized. On the other hand, the oil-droplet size of the emulsions prepared using the ML-750 surfactant significantly changed during the storage. The size of the small oil-droplets gradually increased and leveled off at about ten times the initial size. The large oil-droplets quickly became small droplets, then they became larger, similar to the oil-droplets having the small initial size.

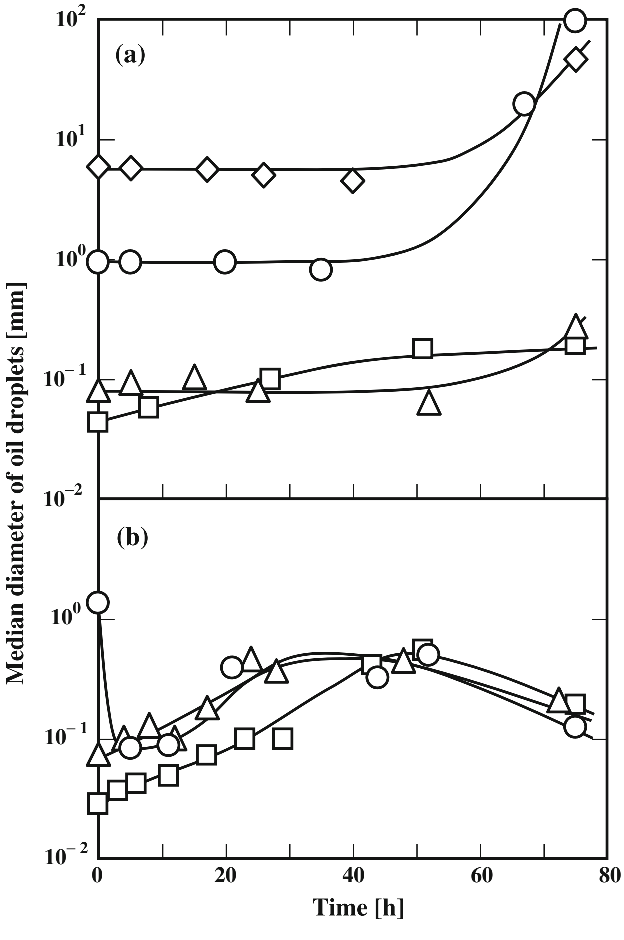

Fig. 6 Changes in the median diameter of oil droplets of the O/W emulsions prepared using **a** Sunsoft® Q-12Y and **b** SY-Glyster® ML-750 during storage at 40°C. The initial median diameters of oil droplets in the emulsions were **a** 43 nm (*open square*), 79 nm (*open triangle*), 0.92 µm (*open circle*), and 5.9 µm (*open diamond*), and **b** 28 nm (*open square*), 73 nm (*open triangle*), and 1.3 µm (*open circle*)

Both the Q-12Y and ML-750 surfactants are chemically decaglyceryl laurate. However, the hydrophilic moiety is not strictly the decaglyceryl residue, but there is a distribution in the degree of polymerization of the glyceryl residues for both surfactants. Although we cannot obtain the precise distributions for the surfactants because they have not been provided by the manufacturers and depend on the lot number of a surfactant, the distribution of the Q-12Y surfactant would be different from that of the ML-750 surfactant. This difference seemed to be a reason for the difference in the stability between the emulsions prepared using the surfactants.

The oxidation rate of methyl linoleate in the emulsion containing smaller sized oil-droplets was slower. A model for explaining the decrease in the oxidation rate for the emulsions having the smaller oil-droplet sizes was proposed on the basis of the dilution of the substrate by the hydrophobic moiety of the surfactant which wedges into the oil phase.

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