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# Oil droplet coalescence does not necessarily affect the flavor release from oil-in-water emulsions



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#### HIGHLIGHTS

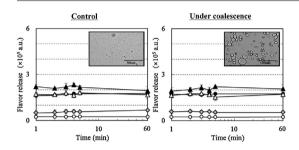
- Impact of oil droplet coalescence on flavor release from emulsions was studied.
- Flavors used in this work were various fruity ethyl esters.
- Flavor release was directly measured using a gas tight syringe.
- Coalescence was able to be successfully controlled by an emulsifier or enzyme.
- Oil droplet coalescence did not affect the flavor release from emulsions.

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#### GRAPHICAL ABSTRACT



### ABSTRACT

Effects of oil droplet coalescence on the release of fruity aroma compounds from milk proteins or a modified starch-based solutions and emulsions were studied by static headspace-gas chromatography. While a coalescence promoter, diglycerol ester of oleic acid decreased the release of relatively hydrophobic ethyl pentanoate and ethyl hexanoate from sodium caseinate solution due to incorporation of the flavor compounds into the vesicles formed in the solution, the other coalescence promoter, an amylase did not significantly affect the flavor release from modified starch solution. For emulsions, the release of the hydrophobic flavor compounds was generally decreased as compared to the solutions, indicating that flavor compounds used in this research tend to dissolve into the dispersed oil phase. Coalescence of oil droplets was successfully induced by the coalescence promoters under static conditions. Evaluation of the flavor release from the emulsions exhibiting coalescence revealed that oil droplet coalescence does not necessarily affect the flavor release from oil-in-water emulsions.

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## 1. Introduction

Flavor is a sensory property that affects food quality and of decisive importance for consumers to choose food products. To meet consumers' requirements, when food researchers and technologists develop new commercial products, they often make much effort not only to add and keep food flavors during processing

and subsequent storage in shelves [1] but also to control flavor release from food stuffs at a desired site and time and a specific rate [2]. Many researches were conducted on factors affecting the flavor release from food products from the following two major viewpoints; that is, physicochemical properties of food ingredients including flavor compounds such as concentration and molecular interactions [3,4] and thermodynamic or mechanical changes of food matrix systems like phase transition, particle fracture and so on [4,5].

Flavor delivery and perception mainly depend on availability of flavor compounds in the gas phase and, therefore, affinity of the

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**Table 1**Physicochemical properties of ethyl esters used in this research.

	Abbreviation	CAS no.	Purity	Log P	Structural formula
Ethyl acetate	EE4	141-78-6	>99.5%	0.642	<u> </u>
Ethyl propanoate	EE5	105-37-3	>97.0%	1.24	<b>^</b> ₀ <b>/</b>
Ethyl butyrate	EE6	105-54-7	>97.0%	1.80	\ <u>\</u>
Ethyl pentanoate	EE7	539-82-2	>98.0%	2.32	\ <u>\</u>
Ethyl hexanoate	EE8	123-66-0	>95.0%	2.80	

flavor compounds for food matrix [6]. Among the major food components, i.e., lipids, proteins and carbohydrates, non-specific trap of flavors in molecules and specific binding of flavor compounds to hydrophobic sites were found for various carbohydrates and proteins [3,6]. On the other hand, lipids often play the key role in the flavor release from food products as they generally have the strongest impact on gas/food product partitioning of flavor compounds consisting of carbon chains [6,7]. Flavor release of lipophilic aroma compounds usually decreases with an increase of lipid content in food products [8,9], while less lipophilic compounds do not exactly relate to the lipid concentration in the products [10].

Oils and fats referred to as lipids normally exist in natural or processed food products as an oil-in-water or water-in-oil emulsion in which one phase is dispersed as small droplets in the other immiscible phase; e.g., milk beverages, fresh fruits juice and ice cream [11]. Emulsions are kinetically stable but thermodynamically unstable so that they usually undergo various types of periodical destabilization such as creaming, flocculation, aggregation and coalescence even under static conditions on a shelf [12]. These kinds of changes, in addition, are well-known to rapidly and intensively occur during consumption of food emulsions in the mouth; for example, flocculation, aggregation and coalescence of oil droplets are induced by saliva and shear stresses depending on the saliva concentration, or ionic strength, emulsifier type and pH of the emulsions [13,14].

Recently, there have been several attempts to relate the colloidal destabilization of emulsions in the oral environment to the sensory properties regarding mouth feels such as creaminess/fattiness and aroma perception [15–17]. Although understanding of the behavior of emulsions post consumption and the subsequent destabilizing effects on the sensory properties are both currently still limited, Sarkar and Singh [18] described a possibility in their literature based on the recent works that oil droplet coalescence affects the release of aroma compounds as most of them are fat soluble. Dresselhuis et al. [19] also clearly pointed out in their work that occurrence of oil droplet coalescence enhances the flavor release from emulsions.

Oil droplet coalescence in the mouth, however, is caused by a series of physicochemical oral processing, that is, temperature change, mechanical shear and dilution [18] and even in a test tube it should be triggered by high-shear stresses or centrifugal treatments [20]. Since all these factors are also expected to strongly affect aroma behavior released from a food system, it has been difficult to accurately estimate the impact of oil droplet coalescence on the flavor release from emulsions.

In our previous research, the authors found that coalescence of oil droplets stabilized by sodium caseinate (NaCAS) can be promoted by bacteriostatic emulsifiers, diglycerol esters of fatty acids in short times under static conditions [21]. Dresselhuis et al. [19] utilized emulsions prepared with octenylsuccinic starch (OSA) in their experiments which are sensitive to an amylase to induce rapid oil droplet coalescence. In the present work, we employ such combinations of materials as a model emulsion where oil droplet coalescence can be controlled excluding the above factors affecting the rate of flavor release, coupled with static headspace gas chromatography in order to study effects of oil droplet coalescence on the flavor release from emulsions.

# 2. Materials and methods

# 2.1. Materials

Ethyl acetate (abbreviated to ethyl ester carbon number 4; EE4), ethyl propanoate (EE5), ethyl butyrate (EE6), ethyl pentanoate (EE7) and ethyl hexanoate (EE8) as flavor compounds and hexadecane (purity>97.0%) as oil phase were purchased from Wako Pure Chemical Industries, Ltd (Osaka, Japan). The octanol-water partition coefficient (Log P) value of the flavor compounds is 0.642, 1.24, 1.80, 2.32 and 2.80, respectively according to the literatures [22–24]. Physicochemical properties of the used flavor compounds are summarized in Table 1. NaCAS (Sodium caseinate 180, Fonterra Co-operative Group) and OSA as emulsifiers were kindly gifted by Riken Vitamin Co., Ltd. (Tokyo, Japan) and San-Ei Gen F.F.I., Inc. (Osaka, Japan), respectively. Diglycerol ester of mono-oleic acid (DO) and an amylase, "Biozyme A" (AMY) as coalescence promoters were manufactured by Riken Vitamin Co., Ltd. (Tokyo, Japan) and Amano Enzyme Inc. (Nagoya, Japan), respectively. Enzymatic activity of the amylase was 7000 unit/g or higher. NaCAS and OSA were used in combination with DO and AMY, respectively (see Section 3).

# 2.2. Sample preparation

### 2.2.1. Solution

Stock emulsifier solutions were prepared by dissolving the emulsifiers (NaCAS and OSA) into 20 mM phosphate buffer solution (pH 7.0) using a stirrer in a water bath at 60 °C. Flavored emulsifier solutions were prepared by gently mixing the stock emulsifier solutions with a small aliquot of flavor compound, followed by overnight storage in a water bath set at 25 °C.

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