

Composition Rather than Viscosity Modifies the Aroma Compound Retention of Flavored Stirred Yogurt

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The influence of thickening agents (modified starch/pectin mixture 0 and 7 g/L) and mechanical treatment (low, medium, and high) on the retention of esters (pentyl acetate and ethyl pentanoate), aldehydes (hexanal and (*E*)-2-hexenal), and a lactone (γ -octalactone) in low-fat flavored stirred yogurts were investigated under equilibrium conditions. In the range studied, the thickening agent and mechanical treatment had little influence on aroma compound retention compared to the decreasing effect of increasing dairy protein concentration on aldehyde retention and the “salting out” effect of carbohydrates on esters. Moreover, experiments in dynamic mode (study of the release of hexanal when yogurts were heated) showed, in the conditions studied, that heat and mass transfer coefficients were not influenced by any of the studied factors (thickening agents and mechanical treatment). These results under static and dynamic conditions are not related to the significant decreasing effect of thickening agents on apple sensory scores associated with hexanal, observed in a previous sensory study. Thus, this sensory effect of thickening agents may be due to sensory interactions between perceptions rather than physicochemical interactions.

KEYWORDS: Retention; kinetic; aroma compounds; flavor; texture; interaction; yogurt

INTRODUCTION

Yogurt is a very popular foodstuff produced throughout the world. Changes in consumption patterns have opened new growth areas for flavored low-fat yogurts. Their acceptability by the consumer is largely determined by product texture as well as flavor. Since aroma compounds generate different flavor perceptions when modifying the dairy matrices, flavorists are devoting a great deal of time to reformulating these products. Indeed, food product flavoring is a complex process because of multiple interactions which can occur between matrix and aroma compounds as well as between texture and flavor perceptions. The odorous molecules that induce aroma perception interact with the food matrix at a molecular level, modifying their retention (1, 2). Furthermore, their release during eating could be limited by the rheological properties of the matrices (3, 4) and by mastication (5) and modified by the dilution with saliva (6, 7). When eating a product, simultaneous stimulation of several senses occurs and the representation of one specific sensory modality is modified by the simultaneous inputs of other modalities (8, 9). Hence, complex sensory interactions between texture or taste perception and aroma perception may occur. To control yogurt formulation, a better understanding of the

mechanisms that govern aroma perception is necessary, taking both physicochemical and sensory interactions into consideration.

Scientific literature reveals that numerous studies have focused on texture/flavor or on matrix/aroma compound interactions, but in simple dairy food related systems. In low-fat yogurt, the main compounds which can modify the aromatic perception are milk proteins, carbohydrates, and thickening agents.

A decrease of vanillin flavor intensity in the presence of sodium caseinate or whey protein concentrate is reported by Hansen and Heinis (10). Reiners et al. (11) found an aroma compound-dependent effect of the β -lactoglobulin addition on odor perception. Thickeners, commonly added to low-fat dairy products to improve their appearance and viscosity, also influence their aromatic perception. Pangborn and Szczesniak (12) were the first authors to show that some hydrocolloids depress the odor and aroma of different aroma compounds. Later, other studies confirmed this result on gels prepared with different aroma compounds and thickeners (13–15).

According to published physicochemical studies, many low-fat yogurt constituents are shown to interact with aroma compounds. Casein is reported to retain aldehydes by chemical bonds (16) and esters by hydrophobic ones (16, 17) whereas β -lactoglobulin forms a hydrophobic pocket offering binding sites for small molecules such as aroma compounds (18, 19).

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Among thickening agents used in dairy products, pectin addition can affect the retention of some aroma compounds (20) and starch induces aroma compound retention which can form complexes with aroma compounds (21). On the contrary, sugar addition induces a release of esters when compared to water as a reference (22, 23) that could be explained by a loss of free water due to the hydration of sugar molecules.

The above-mentioned studies provide information about aroma compound/matrix interactions under equilibrium conditions. But when eating a product, equilibrium conditions are never reached due to chewing, dilution with saliva, and heating to mouth temperature. All these factors could affect aroma compound release to different degrees. Because of the difficulties involved when measuring in the mouth, attempts have been made to simulate some of the mouth processes in model systems that can be closely controlled in terms of temperature, shear rate, hydration (3) and gas flow (24). An increase in viscosity induces a decrease in the initial release rate of diacetyl from aqueous gelatin solutions (0–20%) (25). A mathematical model has been proposed by Harrison and Hills (26) to predict the dynamic flavor release from aqueous solutions containing flavor-binding polymers. According to this model, increasing solution viscosity, which determines the diffusion coefficient, decreases the mass transfer coefficient. Nahon et al. (4) confirmed this model for some of the studied aroma compounds in sucrose aqueous solutions (0–60%).

Very few studies have investigated complex dairy product systems. Cayot et al. (23) reported a release of isopentyl acetate in reconstituted skimmed milk, as compared to water. Lo et al. (27) found an aroma-dependent effect of solid nonfat concentration but no effect of thickening agent (guar gum) concentration on the aroma compound release of acidified milks.

In conclusion, the relationship between sensory data and physicochemical measurements does not always seem to be direct and depends on the product. In recent studies, Hollowood et al. (28) and Weel et al. (29) developed the hypothesis of sensory interactions between texture and flavor rather than physicochemical ones.

Therefore, according to previously published studies, it is of great importance to understand if aroma perception is determined by sensory interactions, by physicochemical interactions, or by both in order to control the flavoring of low-fat stirred yogurt. In a recent study, we examined the effect of thickening agent addition and mechanical treatment on texture, taste, and aroma perception of low-fat flavored stirred yogurt (30). We showed that the addition of thickening agents increases thickness and depresses sweetness as well as the green apple note associated with hexanal.

The aim of this study is to better understand aroma compound/dairy matrix interactions and to clarify the sensory and/or physicochemical nature of the observed interactions between aroma and texture perceptions (30). Two sources of variation of the yogurt texture were investigated: thickening agent addition and mechanical treatment. Measurements of the aroma compound retention by the dairy matrices were performed under equilibrium conditions. Moreover, a kinetic study of the release of hexanal from the different yogurt matrices was carried out to verify if yogurt viscosity influences hexanal release.

MATERIALS AND METHODS

Matrix. (a) *Yogurt Preparation.* Yogurts were prepared from a premix formulated with UHT skim milk (Lactalis, France) fortified with skim milk powder (Regilait, France) and sucrose (50 g/L). The global skim milk composition and the mix composition for yogurt preparation are given in **Table 1**.

Table 1. Main Compounds of Low-Fat Milk (44) and Mix Composition for Yogurt Preparation

	mean low-fat milk comp (g/kg)	mix comp for 1 L of skim milk ^a
water	912–903	
carbohydrates (essentially lactose)	50–52	
caseins	22–28	1 L
whey proteins (essentially β -lactoglobulin)	7–8	
salts + other compounds	8–9	
sugar (sucrose)		50 g
thickening agent (Lygomme AYS53)		0 (TA0) or 7 g (TA1)
skim milk powder (Regilait)		50 (TA0) or 43 g (TA1)
dry matter		19%

^a TA0: yogurt without thickening agent. TA1: yogurt with thickening agent

The thickening agent (Lygomme AYS53, Degussa Texturant System, France) was a mixture of pectin and modified starch and was added to the premix. When thickening agent was used, the dry matter content of the final yogurt was maintained at 19% by counterbalancing the addition of 7 g/L of the thickening agent with a decrease in milk powder addition from 50 g/L to 43 g/L. Before the fermentation step, the premix was heated at 92 °C for 5 min. The fermentation was carried out in a 7-L fermenter (SGi, France) and two fermentations have been performed. The premix were thermostated at 44.5 °C and inoculated with *Lactobacillus delbrueckii* subsp. *Bulgaricus* (0.005% DVS-LB18, Chr Hansen, France) and *Streptococcus thermophilus* (0.01% DVS-ST7 and 0.01% DVS-ST143, Chr Hansen, France). The pH of the culture was monitored during the fermentation. The fermentation was stopped at a target pH value (4.6) by pumping the yogurt through a pipe (length = 1.5 m, diameter = 6 mm) with a peristaltic pump. The yogurts were cooled immediately in a cold chamber at 4 °C. The previous pumping corresponded to the lowest mechanical treatment (MT1). The medium and the highest mechanical treatments were performed 1 day after the fermentation, after the yogurts were cooled. The products were pumped through the same type of pipe for the medium mechanical treatment level (MT2) or through a conical tip (diameter = 0.8 mm, angle = 6°) for the high mechanical treatment level (MT3). Product flavoring was done in glass vessels, at least 3 days after yogurt fermentation to prevent degradation of aldehydes by lactic bacteria (31), using a concentrated mixture of five compounds in propylene glycol. The concentration of the flavoring mixture in the yogurt was 2 g/kg. Yogurts were gently mixed with a spatula (10 rounds from the bottom to the surface), always under the same conditions. The homogeneity and the reproducibility of the yogurt aromatization have been verified by gas chromatography analysis. The reproducibility of yogurt preparation has been controlled by pH and rheological analysis (32).

(b) *Aqueous and Dairy Matrix Preparation.* Flavoring of deionized water matrices and dairy matrices was performed 48 h and at least 8.5 days before headspace analyses, respectively, using the concentrated mixture of five aroma compounds.

(c) *Yogurt Characterization.* (c.1) *Rheological Measurements.* The complex viscosity of the yogurts was measured in harmonic regime with a rheometer (Rheostress RS1 Haake, Germany), equipped with a cone-plate system (C60/2°). A stress sweep was performed with shear stress varying from 0.1 to 1 Pa, at a fixed frequency of 1 Hz. Analyses were performed at three different temperatures: 10, 30, and 50 °C. Each measurement was made in triplicate. The rheological properties of the yogurts were characterized by the complex viscosity measured at a shear stress of 0.1 Pa (η^*) without mechanical breaking of the gel during the measurement test.

(c.2) *pH Measurements.* The pH of the yogurts was measured at 4 °C with pH Mettler glass probes (Mettler Toledo, France) calibrated at 4 °C.

(c.3) *Temperature Measurements.* To quantify the heat transfer in the product, the rise in temperature in the center of the product when heating from 4 °C to 50 °C was followed with a thermocouple (Fluke Corporation, USA). Temperature rise measurements were performed on the same products and at the same time as release kinetic analyses.

Table 2. Physicochemical Properties of Aroma Compounds

aroma compd (MW)	water solubility (mg/L (25 °C)) ^a	log <i>P</i> ^b	Henry's law constant × 10 ³ (dimensionless, 25 °C) ^a
pentyl acetate (130.19)	1700 (a) (20 °C)	2.34	15.9 (d)
ethyl pentanoate (130.19)	2210 (a)	2.34	14.0 (e)
hexanal (100.16)	5640 (a) (30 °C)	1.80	8.61 (f) 8.26 (g) 8.74 (d)
(<i>E</i>)-2-hexenal (98.15)	10000 (b)	1.58	2.00 (h)
γ-octalactone (142.19)	7600 (c)	1.59	0.0498 (c)

^a Key: (a) EPI Estimation Programs Interface v3.10 (46); (b) Philippe (47); (c) Souchon (48); (d) Voustas et al. (49); (e) Hine and Mookerjee (50); (f) Buttery et al. (51); (g) Zhou and Mopper (52); and (h) Buttery et al. (53). ^b Log *P* = log of the partition coefficient of the compound between water and octanol – calculated values (44).

The probe of the thermocouple was plunged into the center of the product in 20 mL headspace vials, placed in a thermostated HS tray (11 × 21.8 × 6.8 cm³).

(d) *Flavors*. Hexanal (97%, Food Chemical Codex, (F.C.C), CAS Registry Number 66-25-1), (*E*)-2-hexenal (98%, CAS Registry Number 505-57-7), ethyl pentanoate (98%, F.C.C, CAS Registry Number 539-82-2), and pentyl acetate (99%, F.C.C, CAS Registry Number 628-63-7) were provided by Sigma-Aldrich (France). γ-Octalactone (98%, CAS Registry Number 104-50-7) was provided by Accros (France). Five-compound flavored matrices were systematically flavored with a concentrated solution of all aroma compounds in propylene glycol (99.5%, Sigma-Aldrich, France, CAS Registry Number 57-55-6). The composition of the solution was the following: pentyl acetate (5%), ethyl pentanoate (5%), hexanal (5%), (*E*)-2-hexenal (5%), γ-octalactone (25%), and propylene glycol (55%). **Table 2** gives the physicochemical properties of aroma compounds from literature.

Retention Measurement Using Headspace Analysis. (a) *Determination of Apparent Partition Coefficients*. Gas-to-matrix apparent partition coefficients were determined at 4 °C as follows:

$$k_{i,\text{gas/matrix}} = \left[\frac{C_i^{\text{gas}} \text{ (mg/L)}}{C_i^{\text{matrix}} \text{ (mg/L)}} \right]_{\text{equilibrium}}$$

where C_i^{gas} and C_i^{matrix} are the equilibrium concentrations of the compound in the gas phase and in the matrix, respectively. Vapor phase from the headspace (HS) sampler was injected with an automatic headspace sampler CombiPal (CTC Analytics, Switzerland) into a gas chromatograph equipped with a flame ionization detector (GC-FID HP6890, Germany). The aroma compounds were separated by a 30 m × 0.53 mm capillary column with a film thickness of 1 μm, type BP 20 (Interchim, France). The operating conditions for GC-FID were the following: helium (carrier gas) flow rate, 8.0 mL/min; detector temperature, 250 °C; hydrogen flow rate, 40 mL/min; air flow rate, 450 mL/min; and helium (make up) flow rate, 33 mL/min. The oven temperature was programmed from 50 °C (maintained for 2 min) to 100 °C at 5 °C/min, and from 100 °C to 180 °C (maintained for 5 min) at 10 °C/min. Each headspace analysis was done in triplicate. Twenty-milliliter vials (Chromacol, France) were used as HS sample containers. Sealed vials were systematically prepared the day before HS analysis and stocked overnight at 4 °C, to ensure that the equilibrium gas/matrix in the vial was reached.

To find the equilibrium conditions and to optimize the analysis repeatability, different ratios (0.05 to 0.5) between sample quantity and vial volume were tested. Similar $k_{i,\text{gas/matrix}}$ values were obtained under all of these conditions, but a better repeatability of C_i^{gas} was observed for small ratios of liquid quantities/vial volume. Consequently, the ratio 0.1 (2 g/20 mL vial) was fixed for all yogurt or milk HS analyses. C_i^{gas} values were calculated from peak areas, obtained by HS analyses, using response coefficients determined for each compound from aqueous solutions of known concentration (from 10 to 500 mg/kg). The initial concentration before equilibrium was assumed to be C_i^{matrix} with C_i^{gas} being negligible compared to C_i^{matrix} .

Preliminary tests did not show a significant effect of the aroma concentration on $k_{i,\text{gas/matrix}}$ (from 10 to 500 mg/kg). Hence, one concentration was tested: (1, in dairy matrix) hexanal, (*E*)-2-hexenal, ethyl pentanoate, and pentyl acetate ≈ 100 mg/kg each and γ-octalactone ≈ 500 mg/kg; (2, in aqueous matrix) hexanal, (*E*)-2-hexenal, ethyl pentanoate, and pentyl acetate ≈ 50 mg/kg each and γ-octalactone ≈ 250 mg/kg.

The matrix composition effect on aroma retention was examined by determining the apparent partition coefficients of each aroma compound for (1) aqueous solution, (2) aqueous solutions with 50 g of sucrose/L, (3) fortified (50 g of milk powder/L) heat-treated milk, (4) fortified (50 g of milk powder/L) heat-treated milk with 50 g of sucrose/L, and (5) fortified (43 g of milk powder/L) heat-treated milk with 50 g of sucrose/L and 7 g of thickening agent/L.

Thickener agent concentration and mechanical treatment effects were studied through a two-factor experimental design with yogurt having a final pH of 4.30 ± 0.02: thickening agent = 0 (TA0) and 7 g/L (TA1) and low (MT1), medium (MT2), and high (MT3) mechanical treatment.

Thickener concentrations and mechanical treatments were chosen from preliminary tests to cover a wide range of viscosity and, consequently, to induce differences in perceived texture, since the same products were used for the sensory panel (30).

(b) *Aroma Compound Release Kinetic Analyses*. Hexanal release from the product to air phase when heating the yogurts from 4 to 50 °C was followed for 110 min. Since no stirring was performed in the vial during release analyses, a higher temperature than the mouth one was tested to induce greater aroma compound release, more similar to what happens in the mouth. HS vials filled with 2 g/vial product and previously stored overnight at 4 °C were placed in the HS sample tray heated at 50 °C. Temperature measurement in the vial revealed that yogurt reached a final temperature of 45.5 ± 0.1 °C. Vapor phase from the HS vial was injected into the GC-FID every 4.3 min. One injection per vial was performed. GC-FID operating conditions were the same as those mentioned above, except for the oven temperature, which was maintained constant at 95 °C. The hexanal release kinetics were determined for different yogurts: with and without thickening agent and with low and high mechanical treatment (TA0MT1, TA0MT3, TA1MT1, and TA1MT3).

Measurement of Temperature in the Mouth. To test if heat transfer during consumption of yogurt is modified by its rheological properties or composition, mouth temperature variation was followed using T thermocouples for 14 subjects (5 women and 9 men, from 23 to 43 years old). Data acquisitions were performed with a module ADAM 4018 (Advantech, France). Two thermocouples were immobilized by inserting them into a silicone pipe. To obtain temperature measurements at different places in the mouth, one of the thermocouples protruded from the pipe by 1 mm and the other one by 5 mm. Two yogurts with different viscosity were tested: TA1/MT1 and TA0/MT3. The subject took a spoonful of yogurt into his mouth and placed the thermocouples on his tongue. Subjects were invited to masticate as they ate yogurt but not to swallow during measurement time. Two replicate and two temperature measurement trials were performed per subject: one until 10 s and the other until 30 s after having taken a spoonful in the mouth.

Quantification of Aroma Compound Concentrations in the Dairy Matrix. The aroma compound concentrations in the dairy matrix were controlled by quantification 1 and 9 days after flavoring. Their concentrations were determined by static headspace analyses of samples heated at 80 °C in stirring conditions for 30 min and by using an external calibration performed for each dairy matrix (32). The GC-FID conditions and program were the same as those for the static HS analyses.

Data Analyses. The composition effect on $k_{i,\text{gas/matrix}}$ was assessed by one-factor (matrix) analysis of variance (ANOVA). The influence of thickening agent and mechanical treatment on apparent partition coefficients was analyzed by two-factor ANOVA (TA, MT, TA×MT). The effect of yogurt viscosity on yogurt temperature in the mouth was assessed by one-factor ANOVA (product). When significant differences were revealed ($p < 0.05$), mean partition coefficients were compared using the Student-Newman-Keuls (SNK) multiple comparison test.

(a) *Determination of Heat Transfer Coefficient.* The heating of 2 g of yogurt in a vial of 20 mL can be considered as a problem of transient conduction through a plane surface corresponding to the bottom of the vial. Assuming that the Biot number (Bi) is higher than 1 and the Fourier number is higher than 0.5, the evolution of temperature inside the product is described by eq 1 (33, 34):

$$\hat{\theta}(x,t) = X(x) \exp(-Z^2 F_o) = X(x) \exp(-a_{\text{therm}} t) \quad (1)$$

where $\hat{\theta}$ is the dimensionless temperature defined by eq 2

$$\hat{\theta}(x,t) = \frac{T(x,t) - T_{\infty}}{T_i - T_{\infty}} \quad (2)$$

and T , T_i , and T_{∞} are the temperatures at time t , at $t = 0$, and at the equilibrium, respectively.

X is a dimensionless constant depending on the Biot number and the position x in the product where the temperature is calculated, a_{therm} is the thermal time constant, Z is a constant defined by the Biot number value (Z is the first root of the implicit eq 3

$$\cot Z = Z/Bi \quad (3)$$

the Fourier number F_o is a dimensionless time defined by eq 4

$$F_o = \frac{\alpha t}{R^2} \quad (4)$$

where t is the time (in seconds), α the thermal diffusivity (in $\text{m}^2 \cdot \text{s}^{-1}$) and R (in m) the depth of the product in the vial. In our case, $R = 5.5 \times 10^{-3}$ m and $\alpha = 1.35 \times 10^{-3} \text{ m}^2 \cdot \text{K}^{-1}$ (35). Considering the numerical values, the assumption $F_o > 0.5$ means that $t > 110$ s. A nonlinear (exponential) regression procedure was used to estimate the three parameters, T_{∞} , X , and a_{therm} (and their associated standard deviation) from eqs 1 and 2 and experimental values of temperatures measured in the yogurt. The value of T_i was set at the initial value measured, $T_i = 4.1$ °C. The estimation of parameters was carried out by considering the experimental points obtained for time values higher than 110 s.

The estimated thermal time constants were higher than 0.0056 s^{-1} , corresponding to Bi values higher than 2.0 by considering eqs 3 and 4. Indeed, the assumption $Bi > 1$ was verified.

(b) *Determination of the Mass Transfer Coefficient.* The mass transfer of aroma compounds in a semisolid food is similar to heat transfer. Equations 1 and 2 have to be adapted by replacing the temperature by the concentration of the aroma compound. If we consider the mean concentration in the gas phase, the parameter X is set at 1. The a_{mass} parameter is a mass diffusion time constant related to the compound diffusivity in the liquid or semisolid phase and in the gas phase. This parameter is of great interest for comparing the diffusion phenomena for different products. The following equation describes the evolution of the gas concentration:

$$\frac{C(t) - C_{\infty}}{C_i - C_{\infty}} = \exp(-a_{\text{mass}} t) \quad (5)$$

By considering the $k_{\text{gas/liq}}$ partition coefficient between the gas phase and the liquid phase and dividing the upper and the lower parts of eq 5 by C_{liq} , we obtain eq 6:

$$\frac{k_{\text{gas/liq}}(t) - m_{\infty}}{k_i - k_{\infty}} = \exp(-a_{\text{mass}} t) \quad (6)$$

On the basis of our experimental conditions, k_i is the value of the partition coefficient at equilibrium at 4.1 °C and k_{∞} is the value of the partition coefficient at equilibrium at 45.2 °C.

Generally, the studies that deal with kinetics and flavor release consider the initial slope of the evolution of the partition coefficient versus time. In our study, we preferred to compare the values of the a_{mass} parameter that is independent of the partition coefficient and that is more closely related to the diffusion of the aroma compound.

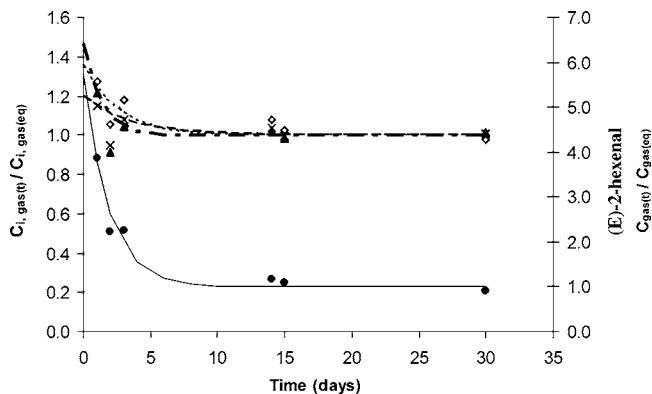


Figure 1. Stabilization time at 4 °C of aroma compounds in yogurt (thickening agent 0 gL^{-1} /low mechanical treatment, TA0/MT1): \diamond , hexanal; \blacktriangle , pentyl acetate; \times , ethyl pentanoate; and \bullet , (E)-2-hexenal. The lines are fitted values of an exponential law.

A nonlinear regression procedure was used to estimate the a_{mass} parameter and its standard deviation (eq 5) from experimental values of aroma compound in the gas phase for the vial containing 2 g of yogurt.

RESULTS AND DISCUSSION

Flavored Matrix Stabilization. Since yogurt is a viscous and complex multicomponent system, stored at a temperature of 4 °C, physicochemical interactions between aroma compounds and dairy matrix stabilize slowly. To determine the time necessary for the stabilization of the flavored matrix, the headspace concentration of the five aroma compounds above the yogurt was followed over 30 days at 4 °C after adding the flavoring agent. The results were expressed as relative headspace concentrations corresponding to the ratio of the gas concentrations at the analyzed time ($C_{i,\text{gas}}(t)$) and at the equilibrium time ($C_{i,\text{gas}}(\text{eq}) = (C_{i,\text{gas}}(t=15 \text{ days}) + C_{i,\text{gas}}(t=30 \text{ days}))/2$) (Figure 1).

A decrease of aroma compound headspace concentration was observed over a period of time (Figure 1), with the exception of γ -octalactone. It was not due to aroma compound loss or degradation since no significant difference was found between initial aroma compound concentration in the dairy matrix and aroma compound concentration, 9 days after flavoring. This decrease can be explained by the low diffusivity of aroma compounds in yogurt at 4 °C. Indeed, during the first days after flavoring, the physicochemical interactions between aroma compounds and the matrix were not yet established, leading to the highest headspace concentrations. A high variability of headspace analyses performed during the first days, partly due to the nonhomogeneity of the matrix, confirmed this explanation.

To reach 95% of the equilibrium value, the five aroma compounds studied in the low-fat stirred yogurt needed less than 4.5 days at 4 °C, with the exception of (E)-2-hexenal with the longest time of 8.5 days. It was assumed that the interactions of (E)-2-hexenal with dairy matrix constituents were greater than those of the other aroma compounds. Since most of the reported studies were performed at temperatures higher than 20 °C and in stirred samples, few authors have dealt with this problem. Consequently, each measurement was performed after a fixed delay of 8.5 days to provide a homogeneous flavoring.

Dairy Protein, Sucrose, and Thickening Agent Effects on Aroma Compound Retention. To better understand the effect of the different constituents on the aroma compound retention in the yogurt matrix, a systematic study was first performed on a liquid matrix containing the main constituents of the yogurt matrix (sucrose, milk, and thickening agent). The gas/matrix

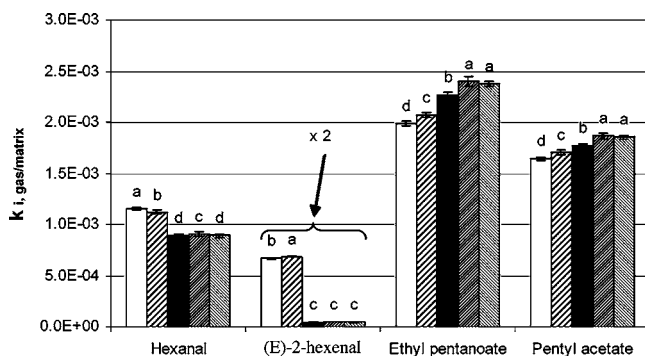


Figure 2. Effect of dairy protein, sugar, and thickening agent addition on $k_{i, \text{gas/matrix}}$ at 4 °C: 1st bar from left, water; 2nd bar from left, water + sucrose; 3rd bar from left, milk; 4th bar from left, milk + sucrose; 5th bar from left, milk + sucrose + thickening agent. The lines indicate the confidence intervals. Different labels (a, b, c, and d) indicate means that significantly differ at $p < 0.05$ (SNK test).

partition coefficients of the aroma compounds were determined in these different liquid matrices at 4 °C and compared to those in water (as a reference).

ANOVA results did not reveal a significant matrix composition effect on $k_{\text{gas/matrix}}$ of γ -octalactone, the less volatile aroma compound of the series studied, probably due to relative high standard deviations.

The retention of the aldehydes was significantly higher in the dairy matrices than in water, but at different levels (**Figure 2**). The (*E*)-2-hexenal partition coefficient decreased by about 94%, whereas the hexanal partition coefficient only decreased by about 22%. Considering that the carbohydrates (sucrose) have a small or nonsignificant effect (in water as well as in milk), a specific interaction between (*E*)-2-hexenal and the dairy proteins could be demonstrated. This interaction would also explain the slower stabilization time of (*E*)-2-hexenal in yogurts.

As for aldehyde/dairy protein interactions, several studies have been carried out on simple food-related systems: aqueous solution of dairy proteins (casein or β -lactoglobulin) and at pH values close to those of milk (6 to 7). The binding activity of caseins has been observed as being dependent on protein concentration. A high retention of heptanal from aqueous solutions with casein content varying from 0% to 12% is reported by Fischer and Widder (16). These authors explained these results by chemical bonds resulting from reactions of aldehydes with the free amino group of proteins (36), producing reversible Schiff bases. This mechanism can be suggested for the matrices studied in this work. Even at low concentrations, the β -lactoglobulin effect could also be taken into account. It is well established that its hydrophobic core is a binding site for small molecules such as aroma compounds (19, 37, 38). Unsaturated aldehydes are reported to bind into the hydrophobic core of β -lactoglobulin (39).

Regarding esters, a slight but significant increase of their gas/milk partition coefficient (<15%) compared to the gas/water partition coefficient was observed, expressing a tendency of milk medium to release esters. Many authors have reported hydrophobic interactions between caseins and esters with $\log P$ values similar to those studied in this paper (16, 17). Esters are also reported to bind into the hydrophobic core of β -lactoglobulin (19, 40). Hence, considering the high hydrophobicity of our esters (**Table 2**), retention by dairy protein would have been expected. On the contrary, a "salting out" effect was observed in the multicomponent milk medium. Apparently, lactose, another main constituent (**Table 1**, 35% more concentrated than

dairy proteins), must be taken into account. Since the same "salting out" effect was also observed when adding sucrose to water, the result on milk matrix is probably due more to the sugar effect than to the effect of dairy protein. In reconstituted skim milk (10% milk powder), the observations of Cayot et al. (23) are in agreement with our results regarding esters. The apparent gas-liquid partition coefficient of isopentyl acetate ($\log P = 2.26$) increased ($\approx 46\%$ of mass fraction) in milk as compared to water.

Sucrose addition had little influence on aldehyde retention (<2.7%) and decreased it slightly for esters (<6%). These results are in agreement with other studies. Kieckbusch and King (22) reported a sharp increase of $k_{\text{gas/sucrose solutions}}$ of C_1 - C_5 acetates with increasing carbohydrate content. The authors attributed these results to the loss of free water due to the hydration of sugar molecules. Cayot et al. (23) reported that sucrose addition (100 g/L) increases the $k_{\text{gas/matrix}}$ of isopentyl acetate (pH 6.5-6.8), compared to water.

As shown in **Figure 2**, thickening agents (pectin and starch) did not seem to affect the retention or release of aroma compounds by dairy liquid matrices: no significant effects of thickening agent addition on $k_{i, \text{gas/milk}}$ were found, except for hexanal for which a little effect was observed. This could be explained by the fact that the added pectin needs an acid pH to play a thickening role. As a general result, aroma compounds of the same chemical class showed similar trends with regard for the different matrices. Thus, in liquid matrices, the observed retention of aldehydes by a dairy matrix would essentially be explained by interactions with caseins (chemical), especially for the (*E*)-2-hexenal. A possible effect of the whey proteins could also be involved in their retention. Dairy protein effect on esters is less apparent in the milk. The presence of sugars enhances ester release from the studied matrices. The less volatile compound, γ -octalactone, is not affected by dairy proteins or the presence of sugars. Finally, among the additives studied, the texturing agent (TA) at a concentration of 7 g/L has little effect compared to that of proteins or sugars.

Thickening Agent and Mechanical Treatment Effect. (a) *Static Study.* On the basis of the yogurt matrices, an experimental design was developed to study the effect of each factor (thickening agent and mechanical treatment) as well as the interaction between these factors, at a fixed temperature of 4 °C, on the retention of the flavor compounds from the dairy matrices. As shown in **Figure 3**, the mechanical treatment had a stronger influence on the complex viscosity of yogurt than the addition of a thickening agent.

Moreover, a limited effect of the thickening agent and the mechanical treatment on the gas/yogurt partition coefficient of the five studied aroma compounds was observed (**Table 3**).

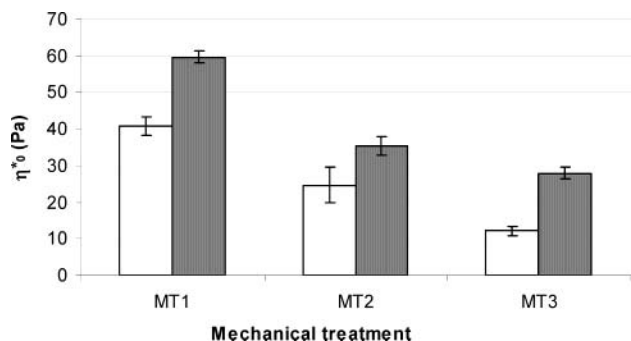
Two-factor ANOVA (TA, MT) with interaction shows a significant influence of the thickening agent on $k_{\text{gas/yogurt}}$ of hexanal alone, whereas the observed mean decrease was less than 5%. The mechanical treatment effect was significant for γ -octalactone: with increasing mechanical treatment, $k_{\text{gas/yogurt}}$ increased (16% and 30%). This result could be explained by a specific interaction between γ -octalactone and a fixation site on casein network modified by the mechanical treatment. This assumption should be verified. A significant interaction between mechanical treatment and thickening agent was revealed for pentyl acetate, showing that mechanical treatment effect depends on the thickening agent level.

Pectin, one of the components of the thickening agent, was reported to increase retention of esters and aldehydes but only when gel structure is created (20, 41). According to the authors,

Table 3. Partition Coefficients of Aroma Compounds for Yogurts ($k_{i,\text{gas/matrix}}$) with Different Levels of Thickening Agents and Mechanical Treatment and Their Confidence Intervals ($T = 4\text{ }^{\circ}\text{C}$)^a

	$k_{i,\text{gas/matrix}}$					
	TA0/MT1	TA0/MT2	TA0/MT3	TA1/MT1	TA1/MT2	TA1/MT3
hexanal ($\times 10^3$)	1.05 \pm 0.06 (B)	1.05 \pm 0.02 (B)	1.04 \pm 0.04 (B)	0.96 \pm 0.06 (A)	1.02 \pm 0.04 (A)	1.03 \pm 0.06 (A)
(E)-2-hexenal ($\times 10^5$)	4.22 \pm 0.39	4.20 \pm 0.10	4.37 \pm 0.23	3.81 \pm 0.46	4.34 \pm 0.46	4.27 \pm 0.52
ethyl pentanoate ($\times 10^3$)	2.32 \pm 0.13	2.28 \pm 0.04	2.22 \pm 0.10	2.10 \pm 0.17	2.25 \pm 0.08	2.34 \pm 0.12
pentyl acetate ($\times 10^3$)	1.83 \pm 0.11 (a)	1.82 \pm 0.03 (ab)	1.81 \pm 0.04 (b)	1.66 \pm 0.17 (a)	1.79 \pm 0.07 (ab)	1.88 \pm 0.12 (b)
γ -octalactone ($\times 10^6$)	1.68 \pm 0.07 (a)	1.95 \pm 0.32 (b)	2.18 \pm 0.16 (b)	1.72 \pm 0.08 (a)	2.00 \pm 0.18 (b)	2.20 \pm 0.66 (b)

^a Labels A, B and a, b indicate means that significantly differ at $p < 0.05$ (SNK test) by thickening agent concentration and mechanical treatment level, respectively.

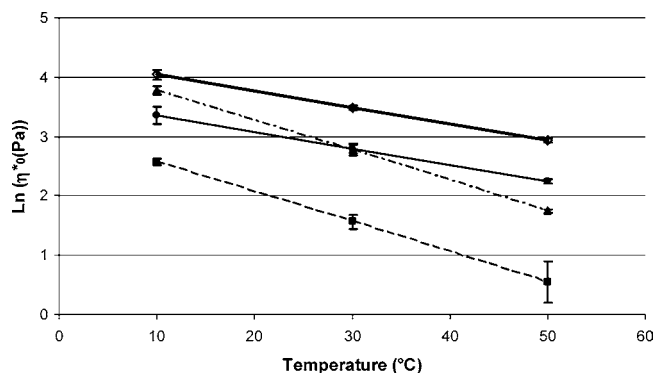
**Figure 3.** Complex viscosity and confidence intervals at low shear stress of 0.1 Pa (η^*) of yogurts at 10 °C: □, without thickening agent; and ■, with 7 g/L of thickening agent.

the aroma compound retention is related to the entangled pectin network, which inhibits migration of aroma molecules from the inner part of the matrix to the surface. As for starch, the other component of the thickening agent used in our experimental plan, it has been shown that it can retain aroma compounds (42). However, it must be pointed out that these studies considered very high concentrations of starch (7%) and matrix flavoring was performed before cooking at high temperatures, favoring complex formation.

In the yogurt matrix studied, the high content of dairy proteins and sugars compared to the thickening agent concentration probably masked the thickening agent and the mechanical treatment effects on the equilibrium properties of these compounds. Lo et al. (27) reported similar results by determining partition coefficients of acetaldehyde, ethanol, and diacetyl in acidified milk with varying concentrations of guar gum (0.1% and 0.5%) and solid nonfat (SNF, 6–12%). They showed that SNF changes the partition coefficients of aroma compounds, but that guar gum concentration had no effect.

(b) *Kinetics of Aroma Compound Release from Yogurt.* To get closer to the mouth conditions, a release study of hexanal under dynamic conditions was carried out. Heat and mass transfer were observed “in vitro” with use of a simple and original setup. Hexanal was chosen in reference to a sensory study (30), which showed that the addition of thickening agents in yogurt depresses the green apple note associated with this compound.

Since yogurts were heated at high temperatures, measurements of their complex viscosity were performed at 10, 30, and 50 °C, to ensure that rheological differences between them really existed. As shown in **Figure 4**, differences persisted at 50 °C, especially between yogurts resulting from the combination thickening agent/low mechanical treatment (TA1/MT1) and those without thickening agent/high mechanical treatment (TA0/MT3). The thermal rheological behavior of yogurt was determined, to a greater extent, by the presence of a thickening agent rather than by the applied mechanical treatment: complex

**Figure 4.** Thermal rheological behavior of yogurts and confidence interval: ▲, TA0/MT1; ■, TA0/MT3; ◇, TA1/MT1; and ●, TA1/MT3 in reference to **Table 3**.

viscosity decreased more slowly in the presence of the thickening agent, regardless of the mechanical treatment. Consequently, thickened yogurt seemed to be more resistant to temperature increase.

(b.1) *Heat Transfer.* When eating a product, several processes could affect the aroma compound release to different extents: (a) mastication and spreading into the oral cavity, which increase the exchange surface between the matrix and the air phase; (b) dilution by saliva; and (c) changes in temperature.

Temperature measurements were performed by heating the product from 4 °C to 50 °C to compare the thermal behavior of yogurt with different rheological properties. The temperature inside the sample placed in vials was recorded every 0.5 min for 30 min.

The thermal time constant values were determined from eqs 1 and 2. These constants were equal to $0.0052 \pm 0.00020\text{ min}^{-1}$ for product TA0MT3 (yogurt without thickening agent and with high mechanical treatment) and to $0.00556 \pm 0.00018\text{ min}^{-1}$ for product TA1MT1 (yogurt with thickening agent and with low mechanical treatment). They did not vary with yogurt, indicating that the thermal properties were identical although the rheological properties were different.

These results were confirmed by yogurt temperature measurements in the mouth when eating the yogurts with the highest viscosity (thickening agent/low mechanical treatment) and the lowest viscosity (no thickening agent/high mechanical treatment) (**Table 4**).

No significant difference (one factor ANOVA: product) was observed between the mean yogurt temperatures for 5 to 10 s after taking a spoonful of the two different yogurts. However, large inter-individual differences in yogurt temperature during consumption were observed. For example, extreme yogurt temperatures over 5 to 10 s were 10 °C for one subject and 24 °C for another one.

(b.2) *Mass Transfer.* The measurement of the headspace concentration over a yogurt sample going from one equilibrium

Table 4. Yogurt Temperature Variation in the Mouth during Eating by a Panel of 14 Subjects for Two Different Yogurts^a

	lowest panel temp (°C)		mean panel temp (°C)		highest panel temp (°C)	
	between 5 and 10 s	between 10 and 30 s	between 5 and 10 s	between 10 and 30 s	between 5 and 10 s	between 10 and 30 s
TA0/MT3	10.4	15.7	20.1	22.6	25.0	27.0
TA1/MT1	10.2	13.2	18.8	21.8	24.0	27.0

^a TA0/MT3 = yogurt without thickening agent and with high mechanical treatment; TA1/MT1 = yogurt with 7 g/L of thickening agent and with low mechanical treatment.

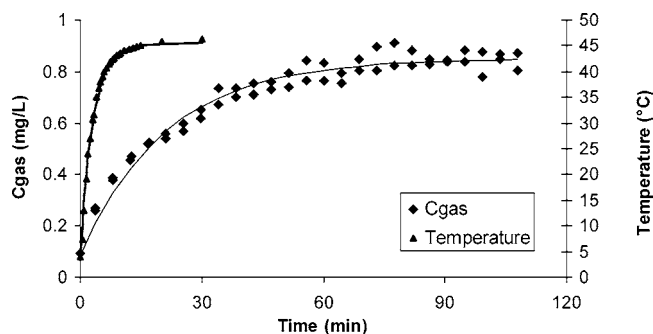


Figure 5. Yogurt temperature (\blacktriangle) and headspace hexanal concentration (\blacklozenge) kinetics of yogurt without thickening agents and with high mechanical treatment (TA0/MT3) with heating from 4 to 45 °C. The lines are fitted values of eqs 1 and 5, respectively.

stage (at 4 °C) to another equilibrium stage (at 50 °C) was also carried out. Headspace concentration was determined every 4.3 min for 110 min, at the same time that the temperature was measured. The gas concentration variation gave quantitative information related to the kinetics of release from matrix to the gas phase.

As expected, the mass transfer is less rapid than the heat transfer (**Figure 5**). Headspace concentration takes more than 17 min to reach 63% of the final concentration. Considering that the heat transfer was a nonlimiting step, the mass transfer was specifically studied.

The hexanal diffusion time constants was determined from eq 5 for the four yogurts, varying in composition (0 and 7 g/L) and in mechanical treatment level (low and high). These constants were 0.047 ± 0.0042 , 0.054 ± 0.008 , 0.037 ± 0.009 , and $0.059 \pm 0.013 \text{ min}^{-1}$ respectively for products TA0MT3, TA0MT1, TA1MT1, and TA1MT3. No notable differences were observed between the different products. These results indicated that the diffusion properties were similar although the rheological properties were different.

According to published studies, several authors have modeled flavor release of simple matrices under simulated mouth conditions, but reported results are contradictory concerning the role of viscosity on aroma compound release. Harrison and Hills (26) have developed a mathematical model based on the penetration theory to describe flavor release from liquid mixtures containing aroma-binding macromolecules such as gelatin. According to their theory, the mass transfer coefficient is inversely proportional to the square root of viscosity. This model was used by Bakker et al. (25) to interpret their experimental data and fitted adequately with release of diacetyl from stirred gelatin solution with different gelatin concentrations inducing varying rheological properties. Initial release slopes decrease with increasing gelatin concentration. The observations of Nahon et al. (4) concerning hexanal are in agreement with our results. These authors modeled flavor release from aqueous solutions containing concentrations of sucrose ranging from 0% to 60%

and reported that increasing the sucrose concentration hardly influences the ratio of release of hexanal compared to water matrix.

Concerning low-fat stirred yogurts, as shown above, the kinetics of hexanal release under dynamic conditions was not dependent on thickening agent composition and product viscosity, in the range of yogurt studied.

(c) *Comparison with Sensory Analyses.* Sensory analyses (30) demonstrated that thickened yogurts were perceived as less intense in grass/green apple note. Physicochemical analyses revealed a limited increasing effect of thickening agent on hexanal retention at equilibrium (<5%) and no effect on release under dynamic conditions. Consequently, it is very unlikely that the decrease in the headspace concentration of 0.3×10^{-3} ppm (5%) may be perceived by subjects. Hence, the sensory analysis results could not be explained by specific interactions between the thickening agent and hexanal. This result tends to support the hypothesis that the texture of yogurt, rather than the flavor headspace concentration itself, determines perception of flavor intensity.

Previous studies explain the lower aroma perception intensity by specific interactions between associated aroma compounds and matrix compounds (12, 43). In more recent studies, authors provided sensory explanations rather than physicochemical ones. Hollowood et al. (28) found that, for concentrations of hydroxy propyl methyl cellulose (HPMC, 0.5% to 1.2%), the perception of strawberry and almond notes decreases. In parallel, these authors have measured the benzaldehyde and ethyl butyrate (marker of strawberry aroma) concentration on the breath during consumption. They observed no concentration variation of both aroma compounds on the breath during consumption, regardless of the HPMC concentration. Consequently, they suggested that the perception of viscosity itself or the effect of HPMC on stimulation of taste receptors may affect overall flavor perception and interaction may occur at a neurological level or at a perceptual level. Weel et al. (29) reported similar results for five whey protein gels, containing β -lactoglobulin and α -lactalbumin and flavored with ethylbutyrate or diacetyl.

These studies (28, 29) tend to confirm that the texture of gels determines perception of flavor intensity rather than the in-nose flavor concentration. The same conclusions can be suggested for the flavored low-fat stirred yogurts studied in this paper.

Furthermore, physiological phenomena, as the yogurt spreading in mouth, could be another assumption to explain the sensory results. Indeed, the aroma compound released quantities depend on the exchange surface developed in the mouth. It could be interesting to determine the impact of the yogurt rheological properties on this parameter, and then, the influence of this parameter on the sensory perception of yogurts.

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