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Effect of Arabic gum, xanthan gum and orange oil on flavor release from diluted orange beverage emulsion

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Abstract

The influence of main emulsion components namely Arabic gum (13–20% w/w), xanthan gum (0.3–0.20% w/w) and orange oil (10–14% w/w) on semi-quantitative headspace analysis of target volatile flavor compounds released from a model orange beverage (diluted orange beverage emulsion) was evaluated by using a three-factor circumscribed central composite design (CCCD). For optimization procedure, the peak area of 13 volatile flavor compounds (i.e. ethyl acetate, α-pinene, ethyl butyrate, β-pinene, 3-carene, myrcene, limonene, γ-terpinene, octanal, decanal, linalool, neral and geranial) were considered as response variables. The response surface analysis exhibited that the significant ($p < 0.05$) second-order polynomial regression equations were successfully fitted for all response variables except for ethyl butyrate. A satisfactory coefficient of determination (R^2) ranged from 0.831 to 0.969 (>0.8) was obtained for the response variables studied. No significant ($p > 0.05$) lack of fit was indicated for the reduced models except for the models fitted for limonene and linalool. This observation confirmed an accurate fitness of the reduced response surface models to the experimental data. The multiple response optimizations indicated that an orange beverage emulsion containing 15.87% (w/w) Arabic gum, 0.5% (w/w) xanthan gum and 10% (w/w) orange oil was predicted to provide the minimum overall flavor release.

Keywords: Semi-quantitative analysis; Orange beverage emulsion; Central composite design; Orange oil; Response surface model; Flavor release

1. Introduction

The physicochemical behavior of small molecules such as flavor compounds in food matrices is one of the most important parameters involved in their activity and sensory perception; hence, this behavior can have pronounced effects on the flavor quality of foods which are mostly emulsions, i.e. dispersed systems of oil and aqueous phases. Variation in the food matrix composition is a key factor that influences the binding and release of volatile flavor compounds (Guichard, 2002). In emulsions, both thermodynamic and kinetic mechanisms influence the flavor release from the emulsions into the gas phase. One mechanism is the partitioning of the flavor compounds between the oil and aqueous phase. Another mechanism is the diffusion of the solutes into the oil or the aqueous phase (Landy, Rogacheva, Lorient, & Voilley, 1998). Physicochemical interactions between flavor compounds and food components can affect the migration of flavor compounds in food products by modifying the nature and number of free binding sites as well as the affinity of the flavor compounds (Nongonierma, Colas, Springett, Le Quéré, & Voilley, 2007).

The term 'beverage emulsion' is used to describe a group of products that have similar composition, preparation and physicochemical properties for example fruit drinks, punches and sodas. In soft drinks, the beverage emulsion

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may provide flavor, color and cloudy appearance for the beverage, or just simply the cloudiness (Tan, 1997). Despite the large number of studies performed to study the flavor release from a variety of emulsion systems (Carey, Asquith, Linforth, & Taylor, 2002; Van Ruth & Roozen, 2000a), there is a lack of sufficient information about the interaction between volatile flavor compounds and emulsion components under accelerated condition (dilution). Since, beverage emulsions are prepared in a concentrated form and then diluted several hundred times prior to consumption (soft drink) (Tse & Reineccius, 1995). The release of volatile flavor compounds from the diluted form (e.g. soft drinks) is more intensive and much easier than the initial concentrated form. Hence, knowledge on the release of volatile compounds under accelerated condition as function of main emulsion components can be useful for the appropriate formulation of beverage emulsion leading to the desirable flavor release in the finished emulsion based product (i.e. soft drink).

In the present study, response surface methodology (RSM) was applied as an appropriate statistical design to (1) study the main and interaction effects of Arabic gum (13–20% w/w), xanthan gum (0.3–0.20% w/w) and orange oil (10–14% w/w) contents on the volatile compound release under accelerated condition (dilution) and (2) determine the optimum emulsion formulation leading to the least volatile flavor release. RSM is an empirical modeling approach for determining the relationship between response variables with the various desired criteria and the significance parameters affecting them. The main advantage of RSM is the reduced number of experimental trials needed to evaluate multiple parameters and their interactions. It should be noted that optimization of an optimum orange beverage emulsion with desirable physicochemical properties (namely viscosity, flow behavior, stability, zeta potential, conductivity, electrophoretic mobility and pH) was investigated in our previous study (Mirhosseini, Tan, Hamid, & Yusof, 2007a, 2007b). Consequently, optimization of an orange beverage emulsion in the present study allows the manufacturers for developing an optimum orange beverage emulsion with minimum volatile compound release under accelerated condition like the finished emulsion-based product (i.e. soft drink).

In fruit juice and beverage, sensory properties and organoleptic attributes of the final products are influenced by the several factors such as type and amount of acids and sweeteners under the salting out effect. In headspace analysis, the recovery of volatile flavor compounds is usually enhanced due to the 'salting out' effect. Salting out effect increases the ionic strength of aqueous solution followed by reduction in the solubility of hydrophobic or non-polar volatile flavor compounds; thus, the partitioning from the aqueous solution to the headspace can be several times higher due to decreased solubility of hydrophobic volatile flavor components in the aqueous phase. In this case, the 'salting out' effect increases the rate and intensity of volatile flavor release of non-polar flavor compounds followed by stimulating the sensory properties induced by them. Besides the salting out effect caused by the presence of acids and sugars, the sensory properties of beverage emulsion-based products (soft drinks) are intensively influenced by the volatile flavor release induced by the addition of beverage emulsion to the soft drink formulation. Since, the effect of sugars and sweeteners on flavor release from a soft drink-related model system has been studied previously (Da Porto, Cordaro, & Marcassa, 2006; Hansson, Andersson, & Leufvén, 2001); in the present study, only deionized water was used to prepare the model orange beverage in order to individually investigate only the effect of main emulsion components on the volatile compound release (Mirhosseini, Yusof, Hamid, & Tan, 2007). In this study, headspace solid-phase microextraction (HS-SPME) method developed based on our previous study (Mirhosseini et al., 2007) was used for the headspace analysis of target volatile flavor compounds. HS-SPME has also been used as a fast alternative technique to determine the flavor release in previous studies (Da Porto et al., 2006; Lubbers, Decourcelle, Vallet, & Guichard, 2004).

2. Materials and methods

2.1. Chemicals and materials

The standards of orange volatile compounds from different chemical classes (aldehydes, esters, ketones, alcohols and terpenes) including ethyl acetate (99%), α-pinene (99.5%), ethyl butyrate (99.7%), β-pinene (98.5%), 3-carene (98.5%), myrcene (95%), limonene (99%), γ-terpinene (98.5%), octanal (98%), decanal (95%), linalool (95%) and citral (95%) (neral and geranial) were supplied by Fluka (Buch, Switzerland). Arabic gum (food grade) was provided by Colloides Naturels International Co. (Rouen, France). Xanthan gum was donated by CP Kelco (Chicago, USA). Citric acid, sodium benzoate and potassium sorbate (p.a. $\geqslant$95%) were purchased from Fisher Scientific (Pittsburgh, PA). Valencia cold pressed orange oil was provided by Danisco (Cultor, Aarhus, Denmark).

2.2. Preparation of orange beverage emulsion

Twenty orange beverage emulsions composed of Arabic gum (13–20% w/w), xanthan gum (0.3–0.5% w/w), orange oil (10–14% w/w), sodium benzoate (0.1% w/w), potassium sorbate (0.1% w/w), citric acid (0.4% w/w) and deionized water were prepared for the optimization procedure based on a circumscribed central composite design (CCCD) (Table 1). As demonstrated in our previous studies (Mirhosseini et al., 2007a, 2007b; Mirhosseini et al., 2007), to prepare the water phase, sodium benzoate, potassium sorbate and citric acid were dispersed in deionized water (60 °C) using a high speed blender (Waring blender 32BL80, New Hartford, USA). While mixing,

Table 1
Matrix of the central composite design (CCD)

Treatment runs	Blocks	Arabic gum (x_1)	Xanthan gum (x_2)	Orange oil (x_3)
1	1	20.00	0.50	10.00
2	1	13.00	0.50	14.00
3	1	13.00	0.30	10.00
4 (C)	1	16.50	0.40	12.00
5	1	20.00	0.30	14.00
6 (C)	1	16.50	0.40	12.00
7	2	13.00	0.50	10.00
8	2	20.00	0.50	14.00
9	2	20.00	0.30	10.00
10	2	13.00	0.30	14.00
11 (C)	2	16.50	0.40	12.00
12 (C)	2	16.50	0.40	12.00
13 (C)	3	16.50	0.40	12.00
14 (C)	3	16.50	0.40	12.00
15	3	16.50	0.24	12.00
16	3	10.79	0.40	12.00
17	3	16.50	0.40	15.27
18	3	16.50	0.40	8.73
19	3	22.22	0.40	12.00
20	3	16.50	0.56	12.00

(C), center point.

gum Arabic was gradually added to the deionized water (60 °C) and mixed for 3 min to facilitate hydration. The Arabic gum solution was kept overnight at room temperature to fully hydrate (Buffo, Reineccius, & Oehlert, 2001). To prepare the water phase, xanthan gum solution was prepared separately by dissolving xanthan gum in deionized water and then mixed with Arabic gum solution by using a high speed blender. While mixing the water phase, the Valencia cold pressed orange oil was gradually dispersed in the water phase to provide an initial coarse emulsion. The pH of water phase was adjusted as required by using a 50% (w/w) solution of citric acid. In our preliminary study, fine emulsification (i.e. small emulsion droplet size of <1 μm and narrow particle size distribution) was achieved by subjecting the pre-emulsions to pre-homogenization using the high shear homogenizer (Silverson L4R, Buckinghamshire, UK) for 1 min and then passed through a high pressure homogenizer (APV, Crawley, UK), for three passes (30, 28 and 25 MPa). Finally, each beverage emulsion was diluted up to 1% (w/w) in order to prepare a model orange beverage.

2.3. HS-SPME procedure

In our previous work (Mirhosseini et al., 2007), the HS-SPME was developed for headspace analysis of the diluted orange beverage emulsion. In preliminary experiments, the optimization of headspace analysis condition was carried out by using different fiber type (PDMS, 100 μm; CAR/PDMS, 75 μm; PDMS/DVB, 65 μm; DVB/CAR/PDMS, 50/30 μm). According to our previous study (Mirhosseini et al., 2007), the highest overall extraction efficiency was obtained using medium-polar fiber CAR/PDMS. Previous study (Rouseff, Bazemore, Goodner, & Naim, 2001) has also reported that the highest extraction efficiency for orange volatile flavor compounds was obtained using medium-polar fiber CAR/PDMS. For SPME analysis, 5 g of the diluted beverage emulsion (1%) was transferred into a 20 ml serum vial containing a microstirring bar. Subsequently, the vial was sealed with a Teflon-lined septa and screw cap that was immersed in a water bath at 45 °C. The SPME fiber coated with CAR/PDMS (Carboxen/polydimethylsiloxane) was manually exposed to the sample headspace for 15 min at 45 °C to reach equilibrium. The sample was continuously agitated with a magnetic stirring bar during the extraction process to allow faster equilibrium time. Finally, the fiber was withdrawn into the needle holder and immediately introduced into the GC injection port and held for 8 min to be completely desorbed the volatile compounds (Mirhosseini et al., 2007).

2.4. GC conditions

As mentioned in our previous work (Mirhosseini et al., 2007), the flavor compounds of Valencia cold pressed orange oil were initially identified by using a Hewlett–Packard 6890N GC system (Wilmington, DE) equipped with Time-of-Flight Mass Spectrometer (TOFMS, Pegasus III, Leco Corp., St. Joseph, MI, USA). The volatile flavor compounds of orange beverage emulsions were then analyzed by a Hewlett–Packard 6890 GC equipped with a flame ionization detector (FID) and a DB-Wax column (J&W Science, i.d. = 0.25 mm, length = 30 m, film thickness = 0.25 μm, Supelco, MA). The GC injection port was equipped with a 0.75 mm i.d. liner (Supelco) to minimize peak broadening. For GC–FID analyses, the injection

was performed for 5 min at 250 °C in the splitless mode. Oven temperature was programmed at 45 °C for 5 min, then ramped to 51 °C at 1 °C/min and held for 5 min at 51 °C then increased to 160 °C at 5 °C/min and finally raised to 250 °C at 12 °C/min and held for 15 min at the final temperature (Mirhosseini et al., 2007). Helium was used as the carrier gas. Detector temperature was set at 270 °C. All results were expressed as the mean values of two independent trials.

2.5. Statistical design

The effect of three independent variables namely, x_1 (gum Arabic (13–20% w/w)), x_2 (xanthan gum (0.3–0.5% w/w)) and x_3 (orange oil (10–14% w/w)) on the volatile flavor release of orange flavor compounds was evaluated by using the RSM. The volatile release of each flavor compound was expressed by the peak area recorded by using GC/FID. Thus, peak area of each volatile flavor compound and total peak area were considered as response variables in the present study. In the present study, circumscribed central composite design (CCCD) as the original form of the central composite design was employed to (1) study the main and combined effects of these independent variables on response variables, (2) create empirical models between the variables and (3) optimize the proportion of main emulsion components in term of the response variables studied. Twenty treatments were assigned based on the CCCD with three independent variables at five levels of each variable involving eight factorial points, six axial points and six center points. The advantage of present CCCD was to simultaneously study the main and interaction effects of three independent variables on the response variable studied. The CCCD contains an imbedded factorial or fractional factorial design with center points that is augmented with a group of 'star points' that allows estimation of curvature. The star points establish new extremes for the low and high settings for all factors. The star points are at some distance α from the center based on the properties desired for the design and the number of factors in the design. The precise value of α and number of center point runs in the design depend on certain properties desired for the design, the number of factors involved and certain properties required for the design. Table 1 illustrates the factor settings and matrix required for the CCCD including the values corresponding to the levels of factors and treatments, assuming three-factors, each with low and high settings. As shown in Table 1, CCCD provides high quality predictions over the entire design space, but require factor settings outside the range of the factors in the factorial part. Experiments were randomized in order to minimize the effects of unexplained variability in the actual responses due to extraneous factors. The center point was repeated six times to calculate the repeatability of the method (Montgomery, 2001). In the present study, the use of blocked design with orthogonal blocking allows the estimation of individual and interaction factor effects independently of the block effects. Blocks are assumed to have no impact on the nature and shape of the response surface. As shown in Table 1, the arrangement of CCCD presented was in such a way that allows the development of the appropriate empirical equations (Montgomery, 2001; Montgomery, Runger, & Hubele, 2001).

2.6. Statistical analyses

The effect of three independent variables on semi-quantitative headspace analysis of target volatile flavor compounds released from 20 model orange beverages was determined using a response surface analysis. The generalized polynomial model proposed for predicting the response variables is given below:

$$Y_i = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \quad (1)$$

where Y_i is a predicted dependent variable, β_0 is the offset term; β_1, β_2, β_3, β_{11}, β_{22}, β_{33}, β_{12}, β_{13} and β_{23} are the corresponding parameter estimates for each linear, quadratic and interaction terms generated for the regression equation. The adequacy of the models were determined using model analysis, lack of fit test and coefficient of determination (R^2) analysis as outlined in previous study (Lee, Ye, Landen, & Eitenmiller, 2000). The terms statistically found non-significant ($p > 0.05$) were dropped from the initial models and the experimental data was refitted only to the significant ($p < 0.05$) parameters in order to obtain the final reduced model. It should be noted that some variables were kept in the reduced model despite non-significance ($p > 0.05$). Since, a quadratic or interaction term containing this variable was significant ($p < 0.05$). The experimental design matrix, data analysis and optimization procedure were performed using Minitab v. 13.2 statistical package (Minitab Inc., Pennsylvania, USA).

2.7. Optimization procedure for determining the desirable orange beverage emulsion

Graphical and numerical optimization procedures were carried out to determine the optimum level of main emulsion components leading to minimum release content of volatile flavor compounds. The graphical optimization technique was used in order to deduce workable optimum conditions (Floros & Chinnan, 1988). Therefore, three-dimensional (3D) response surface was plotted to visualize the relationship between the significant ($p < 0.05$) interaction effects of factors and response variables. Optimum levels of independent variables resulting in minimum overall flavor release were pre-established by superimposing all corresponding response surface plots. Numerical optimization was also carried out by using response optimizer (Minitab v. 13.2) to predict the exact optimum level of independent variables leading to the desirable response goals.

3. Results and discussion

3.1. Preliminary study

According to our previous study (Mirhosseini et al., 2007), 84 volatile flavor compounds were detected by GC-TOFMS in the cold pressed orange oil. Concerning the semi-quantitative analysis of headspace volatile compounds, it should be noted that more than 98% of the total flavor compounds of Valencia cold pressed orange oil composed of ethyl acetate, α-pinene, ethyl butyrate, β-pinene, 3-carene, myrcene, limonene, γ-terpinene, octanal, decanal, linalool, neral and geranial (data not shown). These flavor compounds were also chosen as the main representative volatile flavor compounds in the cold pressed orange oils (Hognadottir & Rouseff, 2003). Thus, the peak area of target volatile flavor compounds and total flavor compounds were considered as response variables in further optimization study.

3.2. The response surface analysis

The application of response surface methodology (RSM) allowed us for studying the main and possible interaction effects between the main beverage emulsion components and volatile flavor compounds. Response surface models fitted for the response variables indicated that each response variable (Y_i) was assessed as a function of linear, quadratic and interaction effects of Arabic gum (x_1), xanthan gum (x_2) and orange oil (x_3) contents. The estimated regression coefficients of three independent variables, along with the corresponding R^2, p-values and lack of fit test for the reduced response surface models are given in Table 2. The individual significance probability of each parameter term and the F-ratio are shown in Table 3. In general, the result indicated that the target volatile flavor compounds did not exhibit the same release pattern, since volatile flavor compounds may be dissolved, adsorbed, bound, entrapped, encapsulated or diffusion-limited by various food components depending on their characteristics (Kinsella, 1990).

The results clearly showed that the release pattern of target flavor compounds significantly ($p < 0.05$) varied for 20 model orange beverages (diluted orange beverage emulsions) depending on the proportion of main emulsion components. For instance, the model orange beverages containing the same quantity of orange oil, but different concentration of Arabic gum and/or xanthan gum did not show the same release content in terms of target volatile flavor compounds. This may be explained by the interaction effects between orange flavor compounds and Arabic gum as well as xanthan gum. The different degree of interaction probably depended on the physicochemical characteristics of the flavor compounds (Bylaite, Nissen, & Meyer, 2005). As shown in Table 2, a high coefficient of determination ($R^2 > 0.83$) was obtained for all polynomial regression models. Joglekar and May (1987) also suggested that R^2 should be at least 0.80 for a good fitness of a model. Thus, this finding indicated that satisfactory adjustment of the reduced response models employed for describing the release variation of target volatile flavor compounds as function of main beverage emulsion components. The ANOVA results showed that the second-order response surface equations were found to be significantly ($p < 0.05$) fitted for all response variables studied except for ethyl butyrate (Table 2). No indication of significant ($p > 0.05$) lack of fit was obtained for most of response regression models except for the ones fitted for limonene and linalool (Table 2). Thus, the further interpretations regarding the effect of main emulsion components on the volatile flavor release have not been described for limonene and linalool in the following results. As stated by Montgomery (2001), the reduced polynomial regression equations fitted to the experimental data was only a statistical empirical model in the selected ranges. It may not be true beyond the ranges of the factors. Therefore, the model cannot be extrapolated beyond these ranges.

In general, the semi-quantitative analysis of target flavor compounds released from different orange beverages was significantly ($p < 0.05$) influenced by the quantity of main beverage emulsion components studied (Table 3). The significant ($p < 0.05$) effect of components on the flavor release from the emulsion has also been reported in previous studies (Pittia, Mastrocola, & Nicoli, 2005). Among the target volatile flavor compounds, the release content of ethyl butyrate was significantly ($p < 0.05$) fitted by a first order regression equation, thus showing the variability of ethyl butyrate was significantly ($p < 0.05$) defined as a linear function of main emulsion components except for xanthan gum (Table 2). The main effect of independent variables was found to be the most significant ($p < 0.05$) terms presented in all response models. Among the independent variable effects, the main effect of orange oil was shown to be included in all reduced models (Table 3). The corresponding variables will be more significant ($p < 0.05$) if the absolute t value becomes larger and the p-value becomes smaller. In term of magnitude of F value, the independent variables exhibited the most significant ($p < 0.05$) effect on the total volatile compound release. All independent variable effects except for quadratic effect of xanthan gum were observed to be significant ($p < 0.05$) in the release of γ-terpinene. Conversely, the release of ethyl butyrate was significantly ($p < 0.05$) influenced only by the main effects of Arabic gum and orange oil (Table 3).

3.2.1. The main effect of independent variables

Among all independent variable effects, the main effect of target emulsion components was found to be the most significant ($p < 0.05$) effect on the release of target volatile flavor compounds. Therefore, they should be considered as the most important controlling factor for predicting the release variation of target flavor compounds from the diluted orange beverage emulsion (model orange beverage) to the headspace. In general, the main effects of Arabic gum and xanthan gum had the same positive or negative

Table 2
Regression coefficients, R^2, p-value and lack of fit test for the reduced response surface models

Regression coefficient	Ethyl acetate	α-Pinene	Ethyl butyrate	β-Pinene	3-Carene	Myrcene	Limonene	γ-Terpinene	Octanal	Decanal	Linalool	Neral	Geranial	Total flavor compound
b_0	−124.0	−1741	−398.9	−1169	1254	−19653	−547954	−3	−2587	−9621	5256.9	−552.9	−568	−540511
b_1	1.7	−337	62.2	47	−61	−2474	−63406	−14	809	–	368.3	36.4	101	−59387
b_2	266.9	13730	–	2909	−1435	−89350	814282	−1528	−22868	21095	–	994.1	5357	707153
b_3	12.1	890	172.1	83	−27	10781	213515	80	599	1527	−682.8	41.5	−125	215920
b_1^2	–	10	–	–	–	121	1829	1	–	–	–	−0.8	−2	1692
b_2^2	−218.0	−9748	–	–	–	–	–	–	–	−31115	–	−719.0	−5711	–
b_3^2	−0.5	−29	–	–	–	−258	−7597	−2	–	−61	55.1	−1.5	5	−7695
b_{12}	−5.0	–	–	−88	90	5254	–	37	–	–	–	−30.2	−73	–
b_{13}	–	–	–	–	2	−257	–	−2	−60	–	−29.9	–	–	–
b_{23}	–	−512	–	−134	–	–	−71987	67	1618	–	–	–	–	−67484
R^2	0.917	0.832	0.890	0.923	0.872	0.969	0.928	0.947	0.945	0.856	0.831	0.871	0.954	0.933
Regression (p-value)	0.022[a]	0.004[a]	0.001[a]	0.000[a]	0.000[a]	0.000[a]	0.000[a]	0.002[a]	0.001[a]	0.001[a]	0.001[a]	0.002[a]	0.000[a]	0.000[a]
Lack of fit (F-value)	5.01	2.90	1.88	1.57	2.64	2.39	22.40	7.51	1.41	0.43	6.41	2.52	1.51	5.90
Lack of fit (p-value)	0.108[b]	0.206[b]	0.252[b]	0.390[b]	0.229[b]	0.255[b]	0.013[b]	0.063[b]	0.429[b]	0.862[b]	0.028[b]	0.240[b]	0.399[b]	0.086[b]

[a] Significant ($p < 0.05$).
[b] Non-significant ($p > 0.05$).

Table 3
Significance probability (p-values and F-ratio) of the independent variable effects in the reduced response surface models

Variables		Main effects			Quadratic effects			Interaction effects		
		x_1	x_2	x_3	x_1^2	x_2^2	x_3^2	x_1x_2	x_1x_3	x_2x_3
Ethyl acetate	p-value	0.060[a]	0.000	0.003	–	0.002	0.003	0.027	–	–
	F-ratio	4.400	24.32	14.16	–	16.53	13.82	6.51	–	–
α-Pinene	p-value	0.003	0.003	0.002	0.003	0.011	0.004	–	–	0.030
	F-ratio	15.63	15.32	18.66	15.41	9.60	13.48	–	–	6.43
Ethyl butyrate	p-value	0.026	–	0.001	–	–	–	–	–	–
	F-ratio	6.09	–	15.25	–	–	–	–	–	–
β-Pinene	p-value	0.001	0.001	0.001	–	–	–	0.005	–	0.011
	F-ratio	20.13	18.05	20.63	–	–	–	11.92	–	9.02
3-Carene	p-value	0.000	0.000	0.026	–	–	–	0.000	0.015	–
	F-ratio	42.51	43.98	6.40	–	–	–	48.78	8.08	–
Myrcene	p-value	0.037	0.000	0.000	0.000	–	0.003	0.000	0.000	–
	F-ratio	5.78	28.62	34.72	29.93	–	14.44	27.68	26.51	–
Limonene	p-value	0.000	0.019	0.000	0.000	–	0.000	–	–	0.000
	F-ratio	37.59	7.49	72.93	34.42	–	63.36	–	–	8.57
γ-Terpinene	p-value	0.347[a]	0.002	0.014	0.016	–	0.024	0.022	0.006	0.020
	F-ratio	0.98	17.64	9.16	8.66	–	7.34	7.65	12.46	7.95
Octanal	p-value	0.008	0.025	0.217[a]	–	–	–	–	0.014	0.049
	F-ratio	10.06	6.56	1.70	–	–	–	–	8.19	4.81
Decanal	p-value	–	0.015	0.018	–	0.006	0.022	–	–	–
	F-ratio	–	7.80	7.33	–	11.04	6.74	–	–	–
Linalool	p-value	0.031	–	0.163[a]	–	–	0.007	–	0.034	–
	F-ratio	5.82	–	2.19	–	–	10.39	–	5.61	–
Neral	p-value	0.001	0.005	0.029	0.005	0.024	0.049	0.013	–	–
	F-ratio	19.05	13.26	6.49	12.90	7.05	5.02	9.22	–	–
Geranial	p-value	0.002	0.000	0.023	0.003	0.000	0.024	0.030	–	–
	F-ratio	17.62	46.51	7.16	15.41	53.74	7.06	6.43	–	–
Total flavor compounds	p-value	0.000	0.035	0.000	0.000	–	0.000	–	–	0.018
	F-ratio	33.67	5.77	76.18	30.12	–	66.39	–	–	7.70

[a] Non-significant ($p > 0.05$).

significant ($p < 0.05$) effect on the release of volatile flavor compounds in most cases. Thus, it was concluded that the release pattern (positive or negative) of target volatile flavor compounds was significantly ($p < 0.05$) governed by the concentration level of hydrocolloid rather than the type of hydrocolloid. Carey et al. (2002) investigated the release of three volatiles with different affinities from the cloud emulsions prepared by Arabic gum or modified starch. They also found that no significant difference ($p < 0.05$) was observed between volatile partitioning from cloud emulsions emulsified with Arabic gum and cloud emulsions emulsified with modified starch, at either lipid concentration. In agreement with previous study (Carey et al., 2002), it was suggested that the target volatile compounds, independent of functionality, did not significantly ($p > 0.05$) interact with the oil–water interface. Since, the orange beverage emulsion contained very low amounts of emulsifier after diluting up to 1% (w/w). Conversely, the main effects of Arabic gum and xanthan gum showed different effect pattern on total flavor release and the release of α-pinene and octanal (Table 2).

The results indicated that the main effect of orange oil was found to be the most significant ($p < 0.05$) effect on the response variables (Table 3). As shown in Table 3, the main effect of orange oil had a significant ($p < 0.05$) effect on the release variation of most of target volatile flavor compounds except for octanal and linalool. However, it had also to be kept the final reduced models fitted for octanal and linalool. Since its interaction with Arabic gum was found to be significant ($p < 0.05$) in both cases. As expected, the release of target volatile flavor compounds was positively affected by the significant ($p < 0.05$) main effect of orange oil in most cases except for 3-carene and geranial. As also shown in Table 2, total flavor compound release was also directly proportional to the main effect of orange oil content. In addition to direct presence of target volatile flavor compounds in orange oil, it may be interpreted by the positive effect of orange oil on the average droplet size. As found in our previous study (Mirhosseini et al., 2007b), it may be explained by the fact that the increase of orange oil content led to the increase in the average droplet size which may be interpreted as main reason for increasing the volatile release from the diluted orange beverage emulsion. In fact, the release of hydrophobic compounds from the matrix to the headspace is performed through transfer from oil phase to water phase and then transfer from water phase to vapor phase. When the emulsion droplet size of the emulsion increases, the total oil–water interfacial surface area is increased. Thus, the increase of the interfacial surface area may enhance the transfer rate of the hydrophobic compounds from oil phase to water phase. Van Ruth, King, and Delahunty (2000b) also reported that the increase of particle diameter increased

the release of aroma compounds from the o/w emulsions containing Tween 20. Previous researchers (Charles, Rosselin, Beck, Sauvageot, & Guichard, 2000) also found that as droplet size increased, the release of lemon and citrus aromas significantly ($p < 0.05$) increased.

Concerning the effect of hydrocolloids on the volatile release, the results indicated that the main effect of Arabic gum had both positive and negative effects on the volatile compound release depending on the type of volatile flavor compound. The main effect Arabic gum showed a significant ($p < 0.05$) negative effect on the release content of total flavor release (Tables 2 and 3). In addition, the release of α-pinene, 3-carene and myrcene was also negatively influenced by the significant ($p < 0.05$) main effect of Arabic gum. It had no significant ($p > 0.05$) effect on the release of ethyl acetate, γ-terpinene and decanal. However, it should be kept in the final reduced models fitted for ethyl acetate and γ-terpinene. Since, its interaction with xanthan gum was found to be significant ($p < 0.05$) in the reduced models. The results also indicated that the release of all volatile compounds except for ethyl butyrate was significantly ($p < 0.05$) affected by the main effect of xanthan gum (Table 3). Thus, it was included in most of the final reduced models fitted for the response variables. As shown in Table 2, the release content of ethyl acetate, α-pinene, β-pinene, decanal, neral and geranial was positively affected by the significant ($p < 0.05$) main effect of xanthan gum; while it had a negative significant ($p < 0.05$) effect on the release variation of octanal, γ-terpinene, myrcene and 3-carene. The same effect as orange oil content, the total flavor compound release was also positively influenced by the main effect of xanthan gum. The results indicated that the main effect of xanthan gum was found to be non-significant ($p > 0.05$) on the release variation of ethyl butyrate and linalool. Thus, it should not be included in their final reduced models (Tables 2 and 3).

3.2.2. The interaction effect of independent variables

In addition to the quadratic effects, the presence of significant ($p < 0.05$) interaction effects of independent variables in the final reduced models confirmed a potentially non-linear relationship (except for ethyl butyrate) between the main emulsion components and the release variation of target volatile flavor compounds (Tables 3 and 4). Fig. 1 exhibited that how significant ($p < 0.05$) interaction effects of independent variables influenced the release variation of some of target volatile flavor compounds. The presence of curvature in 3D response surface plots could be interpreted by the quadratic effects of independent variables. Besides the quadratic effect, the presence of different curvature shapes presented in the release curves (Fig. 1) exhibited that the volatile flavor release was influenced not only by the main emulsion components but also by the type and individual characteristic of each volatile compound.

As clearly shown in Table 2, the interaction effect of orange oil and xanthan gum or Arabic gum decreased the release of volatile compounds and total flavor compound release in most cases. The results indicated that the retention of volatile flavor compounds should be attributed more to the complex interaction induced by the presence of two surface active agents and volatile flavor compounds. In this case, emulsifiers interact with volatile compounds either through a binding effect (many aroma compounds are amphoteric) or by changing the mass transport properties of the liquid interfacial boundary layer (Carey et al., 2002). At least one interaction between independent variables was significantly ($p < 0.05$) fitted to most of response surface models except for the ones fitted for ethyl butyrate and decanal (Table 3). This observation confirmed that the presence of interaction effects improved the fitness of the final reduced models to the experimental data. It was also found that all interaction effects were observed to be significant ($p < 0.05$) on the release variation of γ-terpinene; whereas the release of ethyl butyrate was not significantly ($p > 0.05$) influenced by the interaction effects (Table 3).

Table 3 exhibited that the interaction between Arabic gum and xanthan gum was observed to be the most significant ($p < 0.05$) interaction effects as compared with the other interactions effects. As shown in Table 3, the interaction between Arabic gum and xanthan gum showed a significant ($p < 0.05$) effect on the release pattern of ethyl acetate, β-pinene, 3-carene, myrcene, γ-terpinene, neral and geranial; whereas, it had no significant ($p > 0.05$) effect on the total flavor compound release. It had a significant ($p < 0.05$) effect on the release of most of monoterpene hydrocarbons. The results indicated that the release of monoterpene hydrocarbons such as 3-carene, myrcene and γ-terpinene increased as the interaction effect of Arabic gum and xanthan gum increased. Conversely, it had a significant ($p < 0.05$) negative effect on the release content of ethyl acetate, β-pinene and aldehyde compounds such as neral and geranial. Except for β-pinene, this observation can reflect the effect of chemical classes on the release pattern of target flavor compounds.

The results indicated that the release of 3-carene, myrcene, γ-terpinene and octanal was significantly ($p < 0.05$) affected by the interaction between Arabic gum and orange oil (Table 3). However, it had no significant ($p > 0.05$) effect on the release behavior of ester and aldehyde compounds except for octanal. It was also found to be not significant ($p > 0.05$) in the reduced models fitted to the variation of total flavor release (Tables 2 and 3). The interaction effect of Arabic gum and orange oil exhibited a significant ($p < 0.05$) negative effect on all corresponded cases except for 3-carene. This may be contributed to the presence of an arabinogalactan attached to a polypeptide backbone (AGP) in the molecular structure of Arabic gum. Arbinogalactan–protein complex has a coil conformation with a small radius of gyration and equivalent sphere hydrodynamic radius. The hydrophobic polypeptide chain is believed to bind the hydrophobic flavor compounds; while hydrophilic arabinogalactan blocks are able to anchor the hydrophilic volatile flavor compounds.

Table 4
Comparison between experimental and predicted values based on the final reduced models

Run	Ethyl acetate [a]			α-Pinene [a]			Ethy butyrate [a]			β-Pinene [a]			3-Carene [a]			Myrcene [a]		
	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$
1	13	15	−2	1640	1723	−83	1521	1613	−92	508	518	−10	315	308	6	12996	13816	−821
2	22	21	1	1465	1514	−49	1812	1866	−54	586	562	24	282	278	4	8582	7754	828
3	15	15	−0	1580	1590	−10	1247	1178	69	421	414	6	328	344	−16	6986	6957	29
4	22	21	1	1811	1729	82	1977	1740	237	549	555	−6	300	300	−0	8616	8799	−183
5	16	17	−1	1747	1732	14	2016	2301	−285	722	726	−4	283	272	11	8314	8480	−166
6	23	21	2	1776	1729	47	1866	1740	126	544	555	−11	295	300	−5	9112	8799	313
7	15	13	1	1647	1683	−36	1700	2303	−603	472	478	−7	278	285	−7	8434	8972	−539
8	9	8	1	1514	1416	98	3141	3427	−286	542	559	−17	352	340	12	17819	17850	−31
9	8	9	−1	1346	1491	−145	2681	2739	−58	515	535	−20	220	228	−8	16067	16897	−830
10	10	9	1	1692	1693	−1	3184	2992	192	577	563	14	321	324	−3	18306	18190	116
11	12	14	−2	1757	1660	97	3494	2865	628	570	534	36	307	294	13	14854	15024	−170
12	14	14	0	1647	1660	−13	2993	2865	127	527	534	−7	288	294	−7	16478	15024	1454
13	15	14	1	1508	1698	−191	3008	3473	−465	500	504	−4	309	316	−7	15175	14594	581
14	17	14	2	1650	1698	−48	3157	3473	−316	473	504	−32	315	316	−1	14874	14594	280
15	8	7	1	1535	1473	62	3493	3473	20	538	529	9	315	306	9	14157	15029	−872
16	13	16	−4	2086	2052	34	3250	3118	132	434	439	−5	350	332	18	15023	15456	−433
17	9	10	−1	1300	1363	−63	4080	4035	45	570	599	−29	310	325	−15	12369	12994	−625
18	10	8	1	1560	1417	143	3109	2911	198	410	409	1	321	306	15	11850	10695	1155
19	13	13	0	2049	2003	46	4159	3828	331	618	569	49	290	299	−9	22613	21649	964
20	10	10	−1	1421	1403	18	3528	3473	55	491	479	12	315	325	−10	13110	14159	−1049

	Limonene[a]			γ-Terpinene[a]			Octanal [a]			Decanal [a]			Linalool [a]			Neral [a]			Geranial [a]		
	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$	Y_0	Y_i	$Y_0 - Y_i$
1	314943	332438	−17495	50	54	−4	2747	2513	234	1812	1721	91	4416	4904	−488	128	128	−0	192	209	−17
2	339491	334617	4874	82	75	7	3279	3567	−288	2370	1990	380	5478	5426	52	189	183	6	360	329	31
3	327258	334977	−7719	58	57	0	2582	2408	174	2250	2480	−230	4431	4418	13	147	157	−11	368	365	3
4	358897	346472	12425	50	57	−7	3197	2963	234	3201	2790	411	4683	4735	−52	194	187	7	359	376	−17
5	347196	351904	−4708	50	47	3	3170	3363	−194	2659	2749	−90	5283	5076	206	189	186	3	331	336	−6
6	359095	346472	12623	59	57	1	2803	2963	−160	2228	2790	−562	5005	4735	270	181	187	−6	383	376	6
7	354553	347760	6793	41	48	−7	3017	3363	−347	2345	2367	−22	4846	5010	−164	172	176	−4	303	318	−16
8	315761	307098	8663	139	144	−5	5727	5614	113	2505	2636	−131	5282	5669	−387	157	162	−5	172	188	−16
9	293980	307457	−13477	129	141	−12	5891	6143	−253	2946	3126	−181	5603	5497	106	171	179	−8	310	326	−16
10	374978	367225	7753	160	160	−0	5956	5903	53	3778	3395	383	6018	6019	−1	205	191	14	369	344	25
11	338842	340373	−1531	130	122	8	5804	5256	548	3390	3435	−45	5531	5328	203	197	200	−3	346	361	−16
12	332172	340373	−8201	139	122	17	5142	5256	−114	3431	3435	−5	5571	5328	243	207	200	7	400	361	39
13	359277	369354	−10077	103	115	−12	5410	5684	−275	3758	3985	−227	5400	5429	−29	195	200	−5	470	479	−9
14	364333	369354	−5021	96	115	−19	4957	5684	−728	3750	3985	−235	5345	5429	−84	199	200	−1	475	479	−4
15	385677	377447	8230	142	133	9	6496	6248	248	3872	3776	96	5505	5429	76	194	193	1	388	393	−5
16	437598	446580	−8983	133	135	−2	5066	5196	−131	4034	3985	49	5085	5373	−288	181	184	−4	431	458	−28
17	284145	296117	−11972	109	114	−5	7001	6505	496	3248	3556	−308	6499	6499	0	189	200	−11	508	530	−22
18	298232	280516	17716	75	63	12	5283	4863	420	3350	3117	233	5783	5535	248	180	167	13	565	538	27
19	426327	411600	14727	162	153	9	5852	6172	−320	4550	3985	565	5480	5484	−4	169	163	6	369	337	32
20	356640	361260	−4620	107	98	9	5411	5121	290	2364	2536	−172	5508	5429	79	169	168	1	270	260	10

Y_0: experimental value.
Y_i: predicted value.
$Y_0 - Y_i$: residue.
[a] No significant ($p > 0.05$) difference between experimental (Y_0) and predicted value (Y_i).

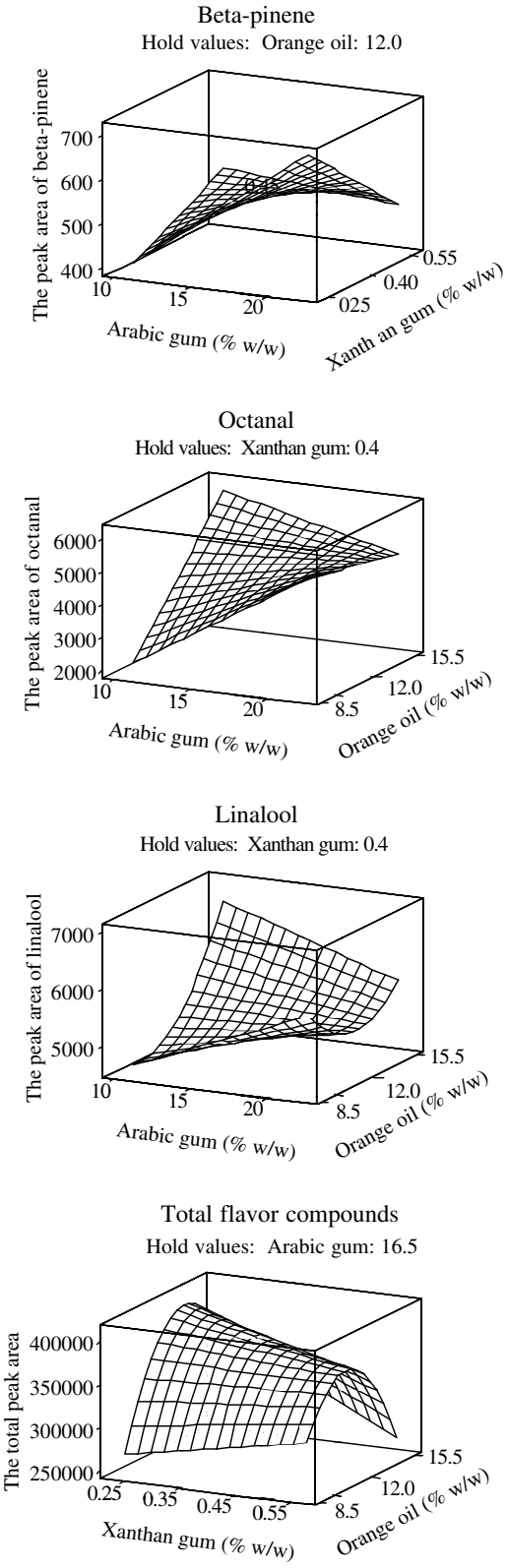

Fig. 1. Response surface plots showing the significant ($p < 0.05$) interaction effect of independent variables on the release pattern of some of target volatile flavor compounds.

The interaction between xanthan gum and orange oil was shown to be not significant ($p > 0.05$) on the release variation of 3-carene, myrcene, ester and aldehyde compounds except for octanal (Table 3). It may be explained by the fact that more than 94% of total orange flavor compounds composed of limonene. The same observation as interaction between Arabic gum and orange oil, the interaction between xanthan gum and orange oil had also a significant ($p < 0.05$) negative effect on the release of corresponded flavor compounds except for γ-terpinene and octanal. This observation may be due to the fact that xanthan gum is an ionic polysaccharide composed of a 1–4 linked β-D-glucose backbone substituted with ionized trisaccharide branches including two mannoses and one glucuronic acid on every second residue (Bylaite et al., 2005). It is a complex polysaccharide with high molecular weight including a large number of free carboxyl groups. The presence of these active sites in the structure of xanthan gum provides a great water absorption capacity. In the ordered conformation, the side chains fold back around the main chain to give a structure analogous to a double helix. In this conformation the ordered molecule is stabilized through hydrogen bonds by non-covalent side chain–main chain interactions involving hydrogen bonding (Bylaite et al., 2005). When xanthan gum is dispersed in aqueous solutions, xanthan gum undergoes a conformational transition from ordered double helix to a complex aggregates through hydrogen bonds and polymer entanglement. Xanthan gum molecules exist in solution in a rigid, ordered conformation and form a tenuous three-dimensional network (Bylaite et al., 2005). Thus, very little increase of xanthan gum content was able to reduce the overall flavor release because of its complicated network and entanglements.

3.3. Optimization procedure

The multiple response optimizations were carried out to (1) visualize the significant ($p < 0.05$) interaction effects of independent variables on the response variables and (2) to predict two set levels of main beverage emulsion components resulted in the minimum and maximum release of volatile flavor compounds from the model orange beverage. The overall optimal condition leading to the minimum overall flavor release was predicted to be obtained at combined level of 15.87% (w/w) Arabic gum, 0.5% (w/w) xanthan gum and 10% (w/w) orange oil; whereas an orange beverage emulsion containing 13% (w/w) Arabic gum, 0.36% (w/w) xanthan gum and 14% (w/w) orange oil was predicted to provide maximum overall flavor release.

As shown in the results, the high concentration of hydrocolloids along with low concentration of orange oil led to the least overall flavor release. On the other hand, as shown in our previous study (Mirhosseini et al., 2007b), the optimum region resulted in the smallest average droplet size was predicted to be obtained by an orange beverage emulsion containing of high concentration of hydrocolloids and low content of orange oil. In fact, the optimum condition led to the smallest average droplet size resulted in the least overall flavor release. Conversely, the

addition of high concentration of orange oil led to the highest emulsion turbidity and the largest average droplet size and subsequently resulted in the maximum flavor release. Thus, it was concluded that a positive correlation between average droplet size and overall flavor release in the diluted orange beverage emulsion. By comparing the present results and our previous study (Mirhosseini et al., 2007b), it was concluded that the optimum emulsion formulation resulted in the least magnitude of overall flavor release was close to the optimum region leading to the desirable physical properties except for turbidity.

As also shown in our another study (Mirhosseini et al., 2007a), the presence of high content of xanthan gum and low concentration of orange oil led to the highest negatively charged zeta potential. As predicted in the present study, the proposed emulsion formulation resulted in the least overall flavor release. Thus, it can be concluded that the magnitude of negatively charged zeta potential was positively correlated with the overall flavor release content. The comparison between the present study and our previous study (Mirhosseini et al., 2007a) also indicated that the presence of low concentration of Arabic gum and high concentration of orange oil resulted in the least pH value and highest content of overall flavor release. Hence, it can be concluded that the increase of pH may lead to decrease the overall flavor release from the diluted orange beverage emulsion to the sample's headspace.

As a main reason, the low concentration of orange oil is believed to reduce the overall flavor release in the optimum orange beverage emulsion. The effect of hydrocolloid concentration on the overall flavor release was more pronounced with xanthan gum content. The negative effect of high xanthan gum content on the overall flavor release content may be interpreted by the fact that the increase of xanthan gum content at the oil–water interface increased the resistance to the transfer of most of hydrophobic volatile flavor compounds across their initial location to the headspace. This suppressive effect of xanthan gum may be due to the reason that xanthan gum has a more distinctive hydrophobic character compared to other hydrocolloids, thus hydrogen bindings between xanthan gum and hydrophilic compounds may influence the flavor release (Bylaite et al., 2005). Secouard, Malhiac, Grisel, and Decroix (2003) also reported that the release of limonene depended on the xanthan gum concentration. They observed that the polymer chains begin to overlap in semi-dilute regime, subsequently limonene release significantly decreased.

The effect of hydrocolloids on the rate and intensity of flavor release may be explained by their effect on the viscosity and/or specific binding interactions of small molecules as a result of adsorption and complexation leading to physical entrapment, encapsulation, hydrogen bonds and any other specific or non-specific binding of flavor molecules (Carr et al., 1996; Kinsella, 1990). In the present study, the effect of hydrocolloids was more pronounced by the second mechanism. Since, the diluted orange beverage emulsions showed low viscosity as compared with the concentrate form. Thus, the reduction of release appeared to be explained by the adsorption, entrapment and binding caused by interactions between flavor compounds and matrix constituents rather than the effect of viscosity.

3.4. Validation of the reduced response models

The adequacy of response surface models was evaluated by using the *T*-test. The test was conducted to compare the experimental values with those predicted values. The closeness between the experimental and predicted values was exhibited by the low residual values. On the other hand, no significant ($p > 0.05$) difference was reported between the experimental and predicted values. Therefore, the experimental values were found to be in agreement with the predicted ones. This observation verified the adequate fitness of the response equations employed for predicting the release variation of target flavor compounds as a function of main beverage emulsion components.

Under corresponding optimum condition, the desirable orange beverage emulsion containing the predicted optimum formulation was practically prepared in order to verify the accuracy of the reduced models fitted for the volatile compound release. Thus, the volatile release of each of target volatile flavor compound in the optimum orange beverage emulsion were determined under the optimum condition and subsequently compared with those predicted values. The corresponding response values obtained from the experimental data and those ones predicted by the reduced models were observed to be close together. The validation results also indicated that no significant ($p > 0.05$) difference between the experimental and predicted values was observed, thus indicating the adequacy of final reduced models fitted by RSM.

4. Conclusions

RSM was employed to predict the optimum levels of main emulsion components leading to minimum release content of target volatile flavor compounds from the model orange beverage. In most cases, the release behavior of target volatile compounds did not follow essentially the same pattern and showed different behaviors according to their chemical classes and their different affinities to the emulsion components. In general, the interaction effects of independent variables on the release variation of target volatile compounds reflected different interaction mechanisms between orange flavor compounds and the emulsifier fraction. As expected, the combination of high concentration of xanthan gum and low content of orange oil was predicted to decrease the overall release content of target volatile flavor compounds from the model orange beverage. The negative effect of hydrocolloids may be contributed to the binding sites present in the emulsifier structure. The present optimum region (15.87% Arabic gum, 0.5% xanthan gum and 10% orange oil) confirmed that the

interaction effect of main emulsion components showed different behavior from their main effects in some cases. For instance, the results indicated that the main effect of xanthan gum had a significant ($p < 0.05$) positive effect on the volatile release of target volatile compounds; while the optimization results predicted that the highest content of xanthan gum provided the minimum overall flavor release. Thus, it was found that the CCCD was a very valuable statistical method for evaluating the main and interaction effects of the main emulsion components on the release of target flavor compounds. This study pointed out the necessity of the interaction effects between main emulsion components for developing an optimum beverage emulsion formulation with desirable flavor release. The present study indicated that the release of target volatile flavor compounds from the diluted beverage emulsion (as model beverage) could be generally modified by the proportion of main beverage emulsion components depending on the desirable goals.

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