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Making Oil-in-Water Emulsions by Ultrasound and Stability Evaluation Using Taguchi Method

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Oil-in-water emulsions containing 40% wt sunflower oil were prepared using ultrasound with the frequency of 30 kHz. The effect of sonication time, stabilizer concentration, NaCl, and pH of aqueous phase on the stability and particle size distribution of samples was investigated using Taguchi statistical method. The results showed that increasing sonication time decreased mean diameter of droplets and narrowed droplet size distribution curves. NaCl was found to have a positive effect on the stability of samples. More stable emulsions were prepared when using xanthan and pectin together at pH 4.

Keywords Emulsification, particle size distribution, pectin, Taguchi method, ultrasound, xanthan

1. INTRODUCTION

Many food products such as milk, cream, beverages, dressings, dips, sauces, butters, and desserts are oil-in-water emulsions that consist of small lipid droplets dispersed in an aqueous medium.[1–3] These products are thermodynamically are unstable. Hence, it is of importance for manufacturers to produce food emulsions of high stability with no or minimal changes in the structure or consistency during storage. Stability of an emulsion depends on many parameters of which size of droplets is of crucial. This has been studied for many years leading to development of new concepts and technologies. It has long been known that ultrasound is capable of making fine emulsions.[4–9] However, yet it has not been widely used in the food industry for some reasons. The disintegration effect of ultrasound is due to the bubbles collapsing at the interface of two immiscible liquids disrupting one phase into another.

In the present work, the stability was monitored as a function of sonication time, ionic strength and pH of aqueous phase, surfactant, and stabilizer concentration and the optimum conditions to make emulsions with reasonable stability were determined using Taguchi's robust design methodology, which was first introduced by Dr. Genichi Taguchi. It is a process/product optimization method based on eight steps of planning, conducting, and evaluating results of matrix experiments to determine the best level of control factors on characteristic properties and hence optimal conditions for any complex process.[10] It has been used in many areas of manufacturing since the 1960 s,[11] with many discrete product engineering and manufacturing companies using it to great effect. This technique is an alternative to standard full factorial designs. Since it reduces the number of experiments, it is easier to use, faster and at the same time accurate and reliable, saving time and cost.[10] Taguchi method can determine the experimental conditions having the least effect on the desired characteristic by calculating a term called signal to noise (S/N) ratio. The experimental conditions having the maximum S/N ratio are considered to be the optimal condition as the experimental variables are inversely proportional to the S/N ratio.[12]

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2. MATERIALS AND METHODS

2.1. Materials

Sodium chloride, glacial acetic acid, and soy bean lecithin were purchased from Merck Co. (Germany). Xanthan from *Xanthomonas campestris* and pectin from citrus peel were supplied by Sigma Chemical Co. (Germany). Sun flower oil was obtained from local market.

2.2. Experimental Procedure

2.2.1. Preparation of Aqueous Phase

Aqueous phase was prepared by dissolving 1 g lecithin and different proportions of xanthan, pectin, and NaCl in distilled water adjusted to pH 3 or 4 by glacial acetic acid. Table 1 shows proportion of each constituent aqueous phase for various formulas used in the experiment.

TABLE 1
Constituents of various formulas used for emulsion making

Compound	Formula 1	Formula 2	Formula 3	Formula 4	Formula 5	Formula 6
Xanthan (g)	0.0	0.0	0.0	0.4	0.0	0.4
Pectin (g)	0.0	0.0	0.4	0.4	0.4	0.0
NaCl (g)	0.0	0.4	0.4	0.4	0.0	0.0
Water (g)	59.0	58.6	58.2	57.8	58.6	58.6
Oil (g)	40.0	40.0	40.0	40.0	40.0	40
Lecithin (g)	1.0	1.0	1.0	1.0	1.0	1.0

2.2.2. Preparation of Emulsion

The aqueous phase and sunflower oil was mixed together in a beaker. An amount of 5 ml aliquot of the mixture was introduced into a round bottom glass tube of 14 mm internal diameter and 100 mm length. Sonication was carried out using Dr. Hielscher ultrasonic processor, Model UP 50 H, Germany, with a tapered titanium sonotrode of 3 mm diameter. The tip of sonotrode was placed 1 cm below the surface of mixture. All samples were sonified in ice water for 60 and 120 seconds.

2.2.3. Determination of Particle Size Distribution

Particle size distribution of samples was measured using Fritsch laser diffraction analyzer, Model Analysette 22 (Germany).

2.3. Experimental Design

Taguchi L8 orthogonal array design was used to screen the effect of pH, sonication time, and xanthan, pectin, and sodium chloride concentration on the particle size distribution of samples. Table 2 shows the L8 orthogonal array design used in this study. Statistical analysis of variance (ANOVA) was performed to determine the significance of process parameters at the confidence level of 0.05. Data were analyzed using Minitab software (version 14.1, UK, 2003).

3. RESULTS AND DISCUSSION

Table 3 shows the results of ANOVA for arithmetic mean diameter of droplets. As can be seen the effect of all experimental variables, except for NaCl, on arithmetic diameter of oil droplets was significant. Xanthan was found to have the most influence followed by the time of sonication, pH and pectin, respectively. The main effect of experimental factors on arithmetic diameter is graphically shown in Figure 1. It is very clear that on going from lower to higher level of the factors, the droplets diameter decreases. Comparing the slopes of lines reveals that the line representing xanthan is steeper than the others, confirming this polysaccharide influenced the droplet diameters the most as explained above. There was an interaction between NaCl and xanthan and also pH and NaCl as illustrated in Figures 2a and 2b. The lines for NaCl and pH in Figure 2b are nearly parallel, that indicate there was little interaction between these two factors. It was therefore pooled in the table of ANOVA (Table 3).

TABLE 2
Taguchi's L8 orthogonal array design for five 2-level factors and 2-interaction

Experiment number	Factors*						
	pH	NaCl	pH*NaCl	Xanthan	Time	NaCl*Xanthan	Pectin
1	1	1	1	1	1	1	1
2	1	1	1	2	2	2	2
3	1	2	2	1	1	2	2
4	1	2	2	2	2	1	1
5	2	1	2	1	2	1	2
6	2	1	2	2	1	2	1
7	2	2	1	1	2	2	1
8	2	2	1	2	1	1	2

*: Levels 1 and 2 represent the lower and higher values assigned to experimental factors in Table 1.

TABLE 3
ANOVA table for arithmetic mean diameter of droplets

Source	Significant at 0.05	DF	SS	Mean square	F value	Pr > F
pH	*	1	2.25781250	2.25781250	409.58	0.0314
NaCl		1	0.02531250	0.02531250	4.59	0.2780
pH*NaCl		1	0.00551250	0.00551250	Pooled	
Xanthan	*	1	8.10031250	8.10031250	1469.44	0.0166
NaCl*xanthan		1	0.20161250	0.20161250	36.57	0.1043
Time	*	1	2.40901250	2.40901250	437.01	0.0304
Pectin	*	1	1.48781250	1.48781250	269.90	0.0387

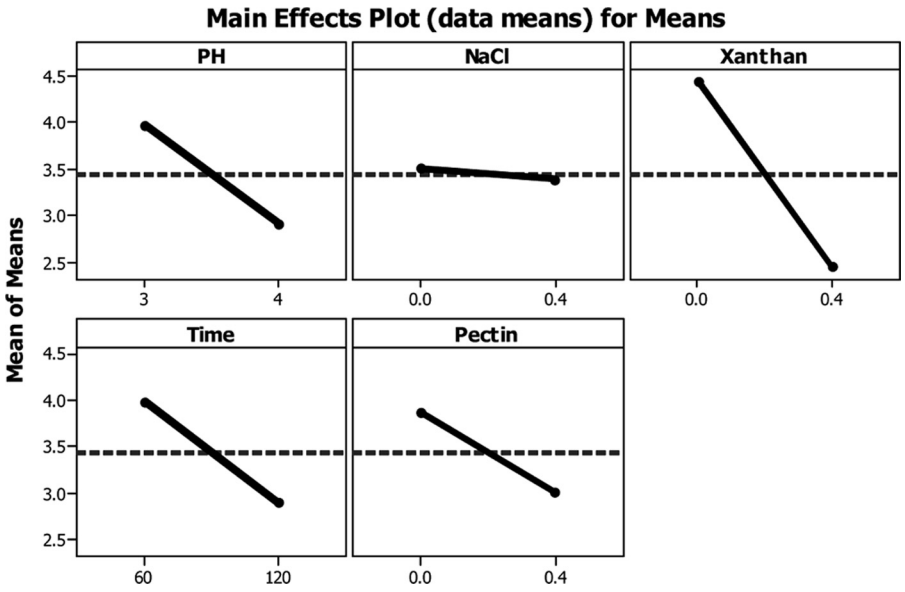

FIG. 1. Main effect plots for means (arithmetic mean diameter).

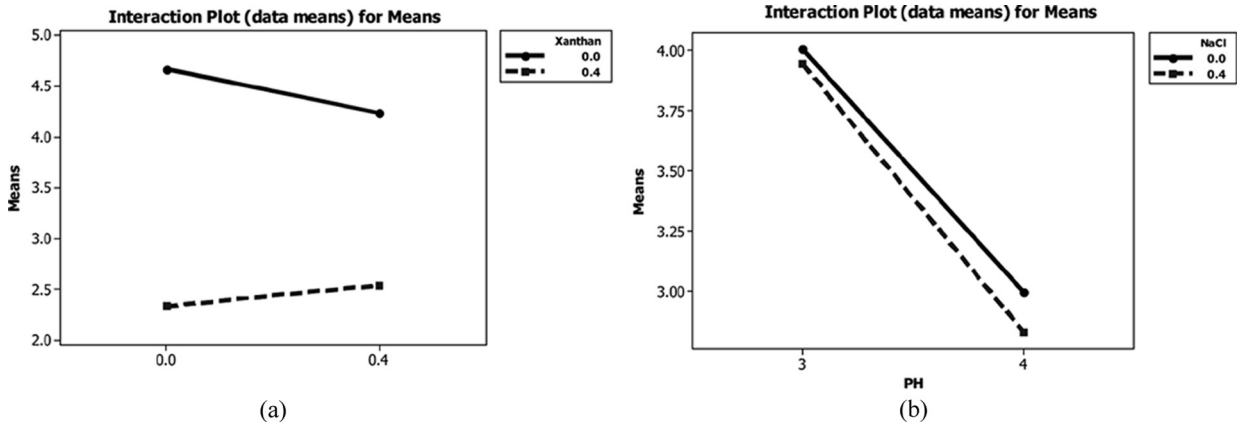

FIG. 2. Interaction plots for means (arithmetic mean diameter): (a) xanthan*NaCl; (b) NaCl*pH.

TABLE 4
ANOVA table for specific surface area of droplets

Source	DF	SS	Mean square	F value
pH	1	0.28125000	0.28125000	5.99
NaCl*Xanthan	1	0.08076180	0.08076180	1.72
NaCl	1	0.04694048	0.04694048	Pooled
Xanthan	1	0.41023682	0.41023682	8.74
pH*NaCl	1	0.31402813	0.31402813	6.69
Time	1	0.75731125	0.75731125	16.13
Pectin	1	0.17006112	0.17006112	3.62

For specific surface of droplets, the software did not clearly indicate how significantly the experimental variables influenced this characteristic, which is likely due to the lack of normality of data. However, the F values given in Table 4 can be alternatively used as an indicator to identify had more important contribution to the specific surface area. From Table 4, it is obvious that the time of sonication had the most impact on the specific surface area of droplets followed by xanthan, pH and pectin, respectively. As was shown for arithmetic mean diameter, the second level of experimental parameters increased the specific surface area (Figure 3). It was also found that NaCl had a considerable effect on the specific surface area, while it almost applied no impact on arithmetic mean diameter. This could be due to the different algorithms used for calculating arithmetic mean diameter and specific surface area by the particle sizer. The same trends they would show if Sauter diameter ($d_{3,2}$) was employed. However, it was pooled when running analysis of variance (Table 4) as its impact on the specific surface area was dramatically less than the other parameters. Figure 4 shows the interaction between pH and NaCl and also xanthan and NaCl. As can be seen, there is a strong interaction between pH and NaCl as the lines intersect, whereas the interaction between NaCl and xanthan is very small and hence it was pooled in the table of ANOVA. It has been reported by Scherze et al. that presence of NaCl during water-in-oil emulsification led to significantly smaller water droplets (larger specific surface area).[13] This is in agreement with the results presented here. Since in lecithin stabilized emulsions the stability mainly comes from the electrostatic repulsion barrier it puts on the emulsion droplets,[14] it seems to be likely that NaCl affects the magnitude of the electrostatic repulsion and hence coalescence and specific surface area of droplets. It may also change the hydrophobicity/lipophilicity balance and the hydration shell around the polar head groups of surfactant molecules.[15] With an increase in the lipophilicity of the surfactant and increased hydrophobic interactions, a relatively rigid surfactant molecule layer would be formed. As was shown before, increasing pH had a beneficial effect on both arithmetic mean diameter and specific surface area of droplets. It seems to be due to changes in the degree of dissociation of carboxylic groups on xanthan and pectin molecules as a function of hydrogen ion concentration in the aqueous phase. The side chains of the xanthan molecules tend to have an appreciable negative charge as the pH goes up. This increases the electrostatic repulsion between droplets and thus the stability of emulsion. The same explanation can be applied to the positive effect of pectin on the stability of emulsion, although it was less significant than xanthan in this study. The pKa value of pectin is around 3.5 so that it starts to

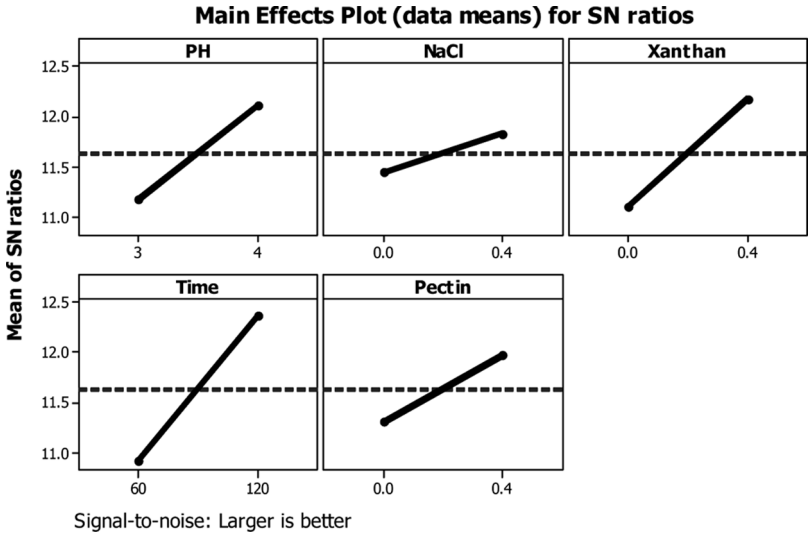

FIG. 3. Main effect plots for means (specific surface area).

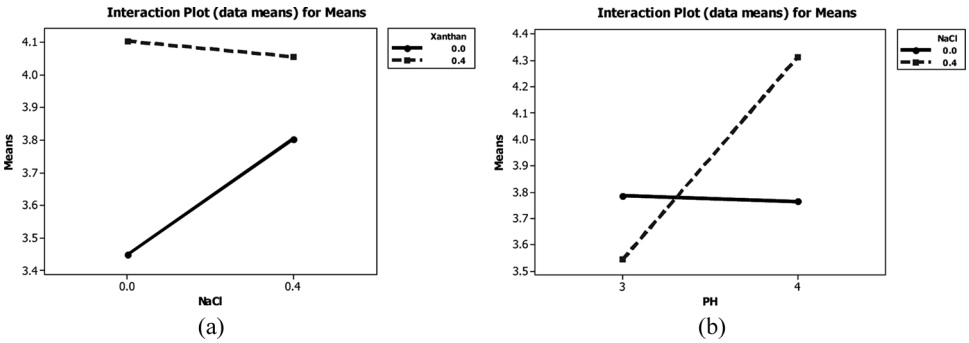

FIG. 4. Interaction plots for means (specific surface area): (a) xanthan*NaCl; (b) NaCl*pH.

lose its negative charge as the pH lowered around or lower this value. This would reflect itself by increasing the coalescence of droplets and accordingly smaller specific surface areas. As can be seen in Table 5, the lowest values was obtained for specific surface area in the absence of xanthan and pectin, that indicate the presence of these polysaccharides is of importance for emulsion stability. The stabilizing effect not only applies by increasing the electrostatic repulsion between droplets, but also by increasing the viscosity of the continuous phase and hence preventing droplets movement which will lead to coalescence, creaming, and eventually emulsion breakdown. It seems that xanthan and pectin have synergistic effect when they are used both together. As is shown in Table 5, simultaneous presence of pectin and xanthan resulted in the largest specific surface area after sonication for 120 seconds. Regarding the time of sonication, it is obvious from the data given in Table 5 that increasing sonication time resulted in larger specific surface areas and smaller arithmetic mean diameters, which are both beneficial to emulsion stability. The increase of sonication time increases the number of collapsing bubbles and hence the shear rate required to disrupt the dispersed phase into continuous one. It should be noted that prolonged sonication may lead to demulsification, which was not observed in this study.

4. CONCLUSIONS

It was shown in this study that Taguchi method can be safely used for stability evaluation of emulsions. The larger the better performance characteristic was taken for specific surface area and the smaller the better was used for arithmetic mean diameter. Both were helpful in identifying the optimum experimental conditions. This technique considerably reduces the number of experiments and gives similar results which could be obtained with full factorial design.

The presence of salt was shown to have positive effect on the stability, although yet further research is required to establish the upper limit of NaCl. Moreover, xanthan was found to have synergetic effect on pectin in stabilizing the emulsion especially at higher pH values. Time of sonication plays a crucial rule in order to make stable fine emulsions and can be studied further.

TABLE 5
The L8 orthogonal array with responses

	Factor					Response	
Experiment no	pH	NaCl	Xanthan	Time	Pectin	Arithmetic mean diameter (μm)	Specific surface area (m^2)
1	3	0	0	60	0	6.15	3.0059
2	3	0	.4	120	.4	1.86	4.5667
3	3	.4	0	60	.4	4.91	3.2554
4	3	.4	.4	120	0	2.98	3.8311
5	4	0	0	120	.4	3.18	3.8916
6	4	0	.4	60	0	2.18	3.6385
7	4	.4	0	120	0	3.56	4.3504
8	4	.4	.4	60	.4	2.10	4.2786

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