frontiers | Frontiers in Environmental Science

ORIGINAL RESEARCH
published: 29 April 2022
doi: 10.3389/fenvs.2022.873730

Tween 20 Stabilized Conventional Heavy Crude Oil-In-Water Emulsions Formed by Mechanical Homogenization

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Specialty section:
This article was submitted to Toxicology, Pollution and the Environment, a section of the journal Frontiers in Environmental Science

Received: 11 February 2022
Accepted: 07 April 2022
Published: 29 April 2022

Citation:
Shen W, Koirala N, Mukherjee D, Lee K, Zhao M and Li J (2022) Tween 20 Stabilized Conventional Heavy Crude Oil-In-Water Emulsions Formed by Mechanical Homogenization. Front. Environ. Sci. 10:873730. doi: 10.3389/fenvs.2022.873730

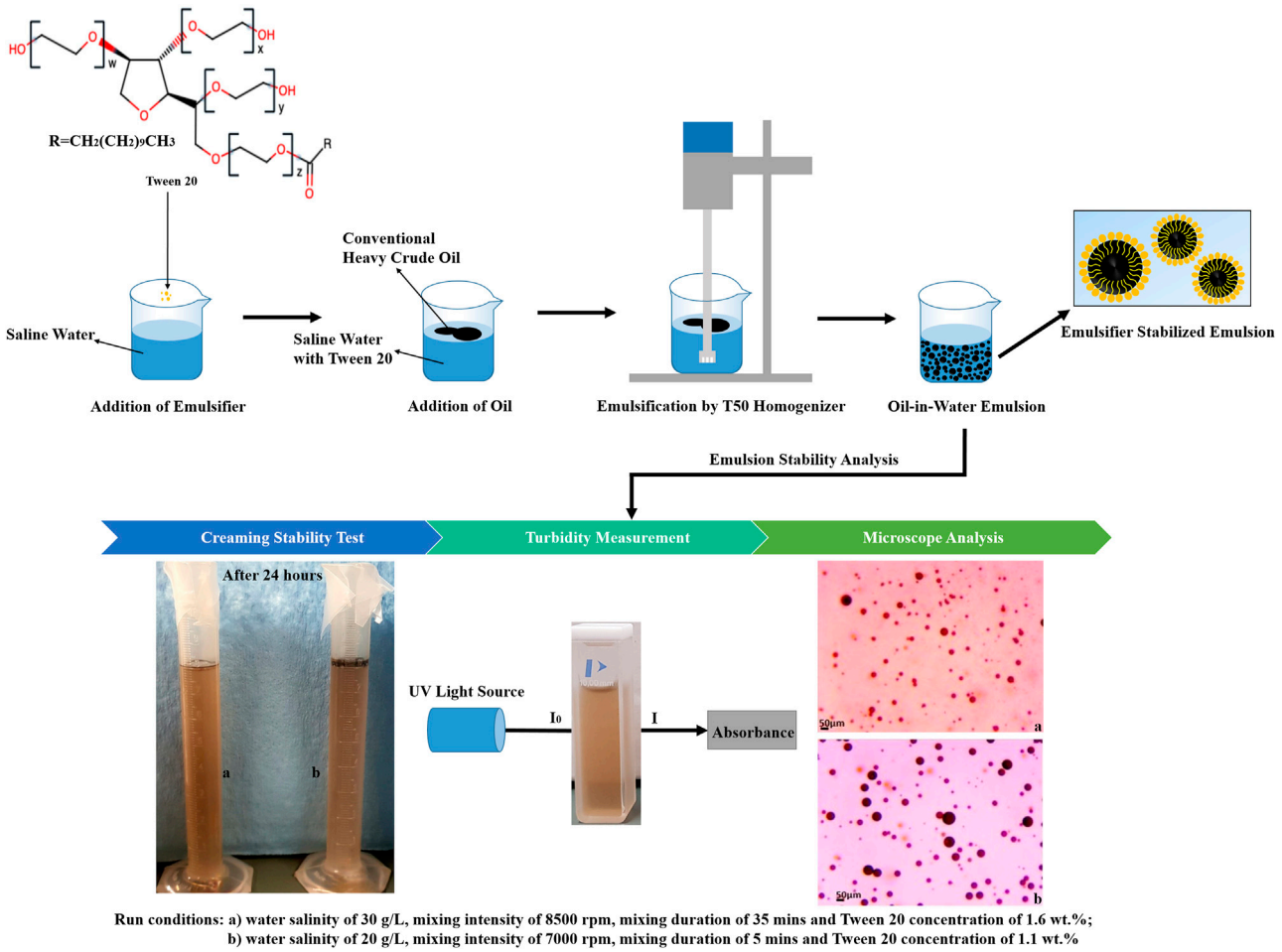
This study investigated the preparation of stable conventional heavy crude oil-in-water (O/W) emulsions by mechanical homogenization with the addition of a non-ionic surfactant, Tween-20. A four-factor, five-level central composite design was carried out to investigate the effects of four independent variables, including mixing intensity (4,000–10,000 rpm), mixing duration (5–45 min), water salinity (0–40 g/L), and the concentration of emulsifier (0.1–2.1 wt%) on the emulsion stability. Emulsion stability was determined by quantification of creaming index, turbidity change rate, and average oil droplet size. The results demonstrated that the salinity of 30 g/L, mixing intensity of 8,500 rpm, mixing duration of 35 min, and emulsifier concentration of 1.6 wt% led to the formation of the most stable emulsion.

Keywords: emulsion stability, heavy crude oil, homogenizer, O/W emulsion, surfactant, turbidity

1 INTRODUCTION

A liquid emulsion can be defined as a mixture of two immiscible liquids where one liquid is dispersed in the form of droplets in another continuous phase (Jha et al., 2014). An emulsion can be classified as oil/water (O/W), water/oil (W/O), and multiple emulsions: water/oil/water (W/O/W) or oil/water/oil (O/W/O) (Abed et al., 2019). These emulsions are essential components of many commercial products, and they have been used in different applications, including foods, cosmetics, personal care products, agrochemicals, supplements, and pharmaceuticals (Wong et al., 2015; Yukuyama et al., 2016). Emulsions are also generated during the production of crude oil (Khan et al., 2015). The occurrences of accidental and operational releases of oil in the marine environment during exploration, extraction, and transportation are well documented (Carpenter, 2019). In many cases, oil emulsions are formed from marine oil spills under the turbulent action of waves (Li et al., 2016). Similarly, oil spills in other water bodies with different salinity, such as freshwater and brackish water, also lead to the generation of emulsions (Tansel and Pascual, 2011).

Abbreviations: O/W, oil/water; W/O, water/oil; W/O/W, Water/oil/water; O/W/O, Oil/water/oil; %, Percentage; IFT, interfacial tension; NaCl, sodium chloride; mPa·s, millipascal-second; RSM, response surface methodology; CCD, central composite design; μm, micrometer; g/L, gram per liter; g/cm^3, gram per cubic centimeter; g, Gram; ppm, parts per million; wt%, Percentage by weight; CI, creaming index.



GRAPHICAL ABSTRACT |

Emulsions are not formed under normal conditions by contacting water and oil. The presence of interfacial barriers known as interfacial tension (IFT) prevents the complete mixing of two immiscible liquids, thus preventing the formation of the emulsion. Therefore, emulsion formation requires energy input: a mechanical mixing force such as shaking and stirring (Nour, 2018). During the production of crude oils, various turbulent sources of mixing energy can create shear forces. Generally, the more significant the amount of shear, the smaller the droplet size of the dispersed phase and the tighter the emulsion (Liu et al., 2016). In the ocean, waves and tides can provide adequate mixing energy to produce stable emulsions with many types of crude oils (Doerffer, 2013). Different researchers have used various equipment and methods to prepare O/W emulsions in the laboratory, including ultrasound, high shear homogenizer, high-pressure homogenizer, microfluidizer, colloid mill, and membrane emulsification (Silva et al., 2016; Taha et al., 2020).Although emulsion generation using a mechanical homogenizer is more complex and requires more energy than other processes such as sonication (Gavahian et al., 2018), mechanical homogenization is one of the commonly used techniques for preparing emulsions as it generates a large shearing and cutting force leading to the formation of fine emulsions (Yang et al., 2012; Mayer et al., 2013).

Emulsions are thermodynamically unstable, and they have a natural tendency to break down into oil and water phases over time through several processes, including gravitational separation, coalescence (or flocculation), and Ostwald ripening (Israelachvili, 2011). Gravitational separation in emulsion occurs due to the difference in density between dispersed and continuous

TABLE 1 | Properties of fresh conventional heavy crude oil.

Properties	Value	Units
API gravity	20.8	degrees (°)
Density (at 25°C)	0.926	g/cm^3
Viscosity (at 25°C)	160.9	mPa·s
Water content	599	ppm
Saturates	52.6	wt%
Aromatics	10.7	wt%
Resins	24.5	wt%
Asphaltenes	12.2	wt%

phases. When droplets float up to the top, it is referred to as creaming, and when they sink to the bottom of the vessel, it is called sedimentation. Likewise, droplet aggregation (also known as flocculation), occurs when droplets attract each other. Lastly, Ostwald ripening is the phenomenon in which smaller particles attach and dissolve on the surface of the larger particles to reach a more thermodynamically stable state, leading to the formation of larger droplets which reduces the stability of emulsions (Costa et al., 2019; Mal et al., 2021).

To increase the stability of emulsions, surfactants, also known as emulsifiers, have been widely used due to their special amphiphilic nature. A surfactant molecule consists of a hydrophilic head that interacts with water and a hydrophobic tail that is attracted by the oil (Esmaeili et al., 2019). Surfactants migrate to the oil-water interface; an energy barrier is thereby generated around the dispersed droplets to prevent them from coalescing as shown in **Figure 2**. In addition, surfactants can reduce the interfacial tension between oil and water phases so that the energy required for droplet dispersion is reduced, facilitating the emulsification process (Kumar and Mahto, 2017). Tween 20, a widely used non-ionic surfactant, is suitable for preparing O/W emulsions (Sartomo et al., 2020). The salient features of these non-ionic surfactants are, they are not affected by the salinity of the water, have a relatively low price (Saad et al., 2020), non-toxic (Alahmer et al., 2010), and lastly, emulsions prepared by these surfactants can be easily separated (Kumar and Mahto, 2017).

A better understanding of O/W emulsion formation is crucial because of the oil spills in the aquatic environment, and the properties of the generated emulsions are essential for predicting, controlling, and mitigating the environmental impacts of such emulsions in different water bodies (Payne and Phillips, 2018). Likewise, selecting appropriate technologies for treating such emulsions requires an understanding of different factors that affect the formation and stability of the emulsions (Goodarzi and Zendehboudi, 2019). The current study investigates the optimum operating conditions to prepare stable conventional heavy crude O/W emulsions using a mechanical homogenizer and an emulsifier, Tween 20. Effects of mixing intensity, mixing duration, pH, and Tween 20 concentration were studied on the stability of the prepared O/W emulsions. The stability of the prepared O/W emulsion was determined by different methods, including creaming stability test, turbidity measurement, and optical microscopy. The results from the experiments can be used to prepare stable emulsions. The prepared emulsion can then be used to assess the performance of different technologies and processes in treating stable emulsions, as treatment of stable emulsions is one of the most significant challenges in oil spill response operations.

2 MATERIALS AND METHODS

2.1 Materials

All chemicals were of analytical grade and purchased from Sigma-Aldrich (Oakville, ON, Canada). A conventional heavy crude oil recovered from the Western Canada Sedimentary Basin (WCSB) provided by Multi-Partner Oil Spill Research Initiative (MPRI) was used as the oil phase, and its physical properties are listed in **Table 1**. Test water phase samples of different salinity were prepared with ultrapure water (UPW, Milli-Q® Advantage A10) and sodium chloride (NaCl, ≥99.0%). A synthetic surfactant (Tween 20, ≥40%) was used as an emulsifier to form O/W emulsions.

2.2 Equipment

A 700 watts high-speed homogenizer (Ultra Turrax T50, IKA®-Werke GmbH and Co.) fitted with a dispersing tool (IKA Works model: S50N-G45G) were used for generating O/W emulsions. A UV-Visible spectrophotometer (Lambda 465 UV/VIS Spectrophotometer, Perkin Elmer) with the wavelength range of 190–1,100 and 1 nm optical resolution was used to measure the turbidity of the prepared emulsion samples in quartz cuvettes (10 mm pathlength and 3.5 ml capacity). A compound microscope (Fisherbrand™ AX800) equipped with a digital camera (Fisherbrand™ C-Mount Digital Camera) was used to observe the microstructure of the emulsions.

2.3 Experimental Procedures

2.3.1 Preparation of Emulsion

Homogeneous test mixtures of saline water were prepared with 0–40 g of NaCl (detailed values shown in **Table 3**) in 1 L of UPW with a magnetic stirrer. For experiments, oil concentration was fixed at 3,000 ppm, and Tween 20 concentration was varied from 0.1–2.1 wt% (details shown in **Table 3**). Tween 20 was added to the saline test solutions and mixed thoroughly by the stirrer at 1,000 rpm, 30°C for 2 min before the addition of 3 g of the fresh conventional heavy crude oil. The IKA mechanical homogenizer and its dispersing tool attachment were used for the emulsification of the crude oil under various mixing intensities (4,000–10,000 rpm) with different mixing durations (5–45 min). The crude oil was dispersed throughout the water phase by high shearing forces and mixing energy. All the experiments were conducted at room temperature. The homogenizer was turned off for 20 min after 10 min of operation to prevent the heating of the prepared emulsions when high mixing intensities and long mixing durations were applied.

2.3.2 Emulsion Stability Analysis

The stability of the emulsion was investigated by three different techniques described in the sections below.

2.3.2.1 Creaming Stability Test

An 8 ml O/W emulsion was transferred into a 10 ml graduated cylinder immediately after preparation, and it was tightly capped to avoid evaporation and stored under room temperature for 24 h (Patil and Benjakul, 2017). Emulsion stability was estimated by measuring the thickness of the creaming layer (separated oil layer). The creaming index (CI) was expressed as the percentage of the total height of the cream layer at the top over the total height of the emulsion sample according to **Eq. 1** (Campelo et al., 2017; Ferreira et al., 2010):

$$CI\,(\%) = \frac{CH}{EH} \times 100 \tag{1}$$

TABLE 2 | The independent variables and their coded levels in the factorial design.

Independent variables	Coded levels					Units
	–2	-1	0	+1	2	
Mixing Intensity	4,000	5,500	7,000	8,500	10,000	rpm
Mixing Duration	5	15	25	35	45	minute
Water Salinity	0	10	20	30	40	g/L
Tween 20 Concentration	0.1	0.6	1.1	1.6	2.1	wt%

where *CH* is the height of a cream layer, *EH* is the total height of an emulsion sample. A larger *CI* indicates that the emulsion has lower stability.

2.3.2.2 Turbidity Measurement

Turbidity measurement for the emulsion stability offers the advantage of rapid and accurate determination of emulsion stability (Alade et al., 2021). Kundu et al. (2019) the concentration of oil droplets in the emulsion. The emulsion turbidity will decrease when there is less oil droplets with larger droplet size present in the emulsion due to droplet coalescence (Kundu et al., 2013). The turbidity was measured immediately after the preparation of the emulsion and after 24 h of storage (Zhang et al., 2016). A glass pipette was used to transfer 3 ml of prepared emulsion into a 3.5 ml quartz cuvette. The turbidity was measured at a wavelength of 260 nm by the transmission of light using a UV-Visible spectrophotometer. Each measurement was conducted in triplicate and the average transmittance reading was recorded. UPW was used as a blank. The turbidity was calculated using the equation below (Sezer, 2019; Mureşan and Dănilă, 2020):

$$\tau = -\frac{1}{L} \times \ln \frac{I}{I_0} \tag{2}$$

where τ is the turbidity of the emulsion, L is the path length of the light (1 cm), I is the intensity of the light coming out of the sample, I_0 is the intensity of the initial incident light beam.

The turbidity change rate was applied to indicate the emulsion stability in terms of the following equation:

$$TC(\%) = \frac{\tau_i - \tau_f}{\tau_i} \times 100 \tag{3}$$

where τ_i is the initial turbidity of the freshly prepared emulsion, τ_f is the turbidity measured after 24 h. The higher turbidity change rate indicates that the emulsion is less stable since more oil droplets float to the top.

2.3.2.3 Microscopy Analysis

Microscopic analysis was conducted to determine the mean diameter of oil droplets and droplet size distribution to investigate the emulsion stability. The smaller oil droplet size indicates the emulsion is more stable (Abbasi et al., 2020). After the homogenization of the emulsion, a drop of each sample was placed on a glass microscope slide. A compound microscope with

TABLE 3 | Design of experiments based on response surface methodology.

Run	Independent variables			
	A: Salinity (g/L)	B: Mixing intensity (rpm)	C: Mixing duration (minute)	D: Tween 20 concentration (wt%)
1	20	7,000	25	1.1
2	30	8,500	35	0.6
3	20	7,000	25	2.1
4	20	7,000	25	1.1
5	20	7,000	25	1.1
6	30	8,500	15	0.6
7	30	5,500	35	1.6
8	10	5,500	15	0.6
9	20	7,000	25	0.1
10	30	5,500	15	0.6
11	20	7,000	5	1.1
12	30	8,500	15	1.6
13	0	7,000	25	1.1
14	10	8,500	35	1.6
15	20	10,000	25	1.1
16	10	8,500	35	0.6
17	10	8,500	15	1.6
18	40	7,000	25	1.1
19	10	8,500	15	0.6
20	20	7,000	45	1.1
21	30	8,500	35	1.6
22	10	5,500	15	1.6
23	20	4,000	25	1.1
24	10	5,500	35	0.6
25	30	5,500	35	0.6
26	10	5,500	35	1.6
27	30	5,500	15	1.6

TABLE 4 | Experimental results for three responses according to the central composite design.

Run	Independent variables				Response 1 creaming index (%) (Act. and Pre.)		Error 1	Response 2 turbidity change rate (%) (Act. and Pre.)		Error 2	Response 3 average oil droplet size (μm) (Act. and Pre.)		Error 3
	A	B	C	D									
1	20	7,000	25	1.1	10.4	9.63	0.77	10.1	9.04	1.06	33.11	32.22	0.89
2	30	8,500	35	0.6	6.5	5.75	0.75	4.84	4.48	0.36	21.92	22.36	−0.44
3	20	7,000	25	2.1	9.7	8.01	1.69	9.61	8.11	1.50	32.32	29.59	2.73
4	20	7,000	25	1.1	9.4	9.63	−0.23	9.24	9.04	0.20	32.17	32.22	−0.05
5	20	7,000	25	1.1	9.8	9.63	0.17	9.63	9.04	0.59	32.39	32.22	0.17
6	30	8,500	15	0.6	7.5	8.49	−0.99	5.5	8.09	−2.59	27.08	29.46	−2.38
7	30	5,500	35	1.6	8.4	7.74	0.66	7.34	6.79	0.55	30.99	30.53	0.46
8	10	5,500	15	0.6	15.1	14.58	0.52	17.32	16.26	1.06	42.18	41.62	0.56
9	20	7,000	25	0.1	11.2	11.26	−0.06	10.83	9.96	0.87	34.5	34.85	−0.35
10	30	5,500	15	0.6	12.5	13.17	−0.67	12.45	13.57	−1.12	40.13	40.26	−0.13
11	20	7,000	5	1.1	17.2	16.66	0.54	22.53	20.78	1.75	44.9	44.48	0.42
12	30	8,500	15	1.6	6.6	7.93	−1.33	5.12	9.41	−4.29	27.98	26.84	1.14
13	0	7,000	25	1.1	10.3	11.97	−1.67	9.81	11.73	−1.92	34.39	36.48	−2.09
14	10	8,500	35	1.6	5.4	6.34	−0.94	3.77	3.99	−0.22	24.82	26.88	−2.06
15	20	10,000	25	1.1	4.3	3.41	0.89	3.57	1.14	2.43	21.25	18.78	2.47
16	10	8,500	35	0.6	8.1	9.03	−0.93	6.46	7.17	−0.71	29.57	29.51	0.06
17	10	8,500	15	1.6	11.9	11.21	0.69	11.8	12.10	−0.30	34.38	33.99	0.39
18	40	7,000	25	1.1	8	7.29	0.71	6.25	6.35	−0.10	29.91	27.96	1.95
19	10	8,500	15	0.6	12.9	11.77	1.13	11.44	10.78	0.66	36.96	36.61	0.35
20	20	7,000	45	1.1	9.2	9.05	0.15	8.43	9.07	−0.64	31.48	30.27	1.21
21	30	8,500	35	1.6	2.2	3.06	−0.86	2.44	1.30	1.14	16.12	19.73	−3.61
22	10	5,500	15	1.6	13.7	14.01	−0.31	17.54	17.59	−0.05	38.34	39.00	−0.66
23	20	4,000	25	1.1	10.7	10.90	−0.20	10.79	12.11	−1.32	33.75	34.59	−0.84
24	10	5,500	35	0.6	12.9	11.83	1.07	13.52	12.66	0.86	36.91	34.52	2.39
25	30	5,500	35	0.6	9.9	10.43	−0.53	9.86	9.97	−0.11	32.58	33.16	−0.58
26	10	5,500	35	1.6	8.9	9.14	−0.24	7.79	9.48	−1.69	31.65	31.89	−0.24
27	30	5,500	15	1.6	11.8	12.61	−0.81	16.92	14.90	2.02	35.9	37.64	−1.74

Note: Act. = actual value; Pre. = predicted value.

TABLE 5 | ANOVA for response surface quadratic model for creaming index.

Source	Sum of squares	df	Mean square	F-value	*p*-value	
Model	260.66	8	32.58	30.98	<0.0001	significant
A	32.9	1	32.9	31.28	<0.0001	
B	84	1	84	79.86	<0.0001	
C	87.02	1	87.02	82.73	<0.0001	
D	15.84	1	15.84	15.06	0.0011	
AB	3.52	1	3.52	3.34	0.0841	
CD	4.52	1	4.52	4.29	0.0529	
B^2	9.82	1	9.82	9.33	0.0068	
C^2	16.62	1	16.62	15.8	0.0009	
Residual	18.93	18	1.05			
Lack of Fit	18.43	16	1.15	4.55	0.1951	not significant
Pure Error	0.5067	2	0.2533			
Core Total	279.59	26				

a 10× ocular lens and 20× objective lens was used to visualize the dispersed oil droplets in the water phase (the image being viewed is 200 times its actual size). Images from the digital camera, captured on a computer using SeBaView software (Laxco Inc., Bothell, WA, United States) were processed with ImageJ (National Institutes of Health, Bethesda, MD, United States) image analysis software for particle analysis. For statistical analysis, the diameter of at least 300 oil droplets was measured for each sample.

2.4 Experimental Design

The experiments were designed using the Design Expert 12.0 (Stat-Ease Inc., Minneapolis, MN, United States). Response Surface Methodology (RSM) with Central Composite Design (CCD) was used to investigate the effects of different factors, including mixing duration, mixing intensity, salinity, Tween 20 concentration on the emulsion stability (**Table 2**). The full factorial design considered four factors at five levels, and a total of 27 runs (3 replicates at the center point to evaluate the error) were employed as shown in **Table 3**.

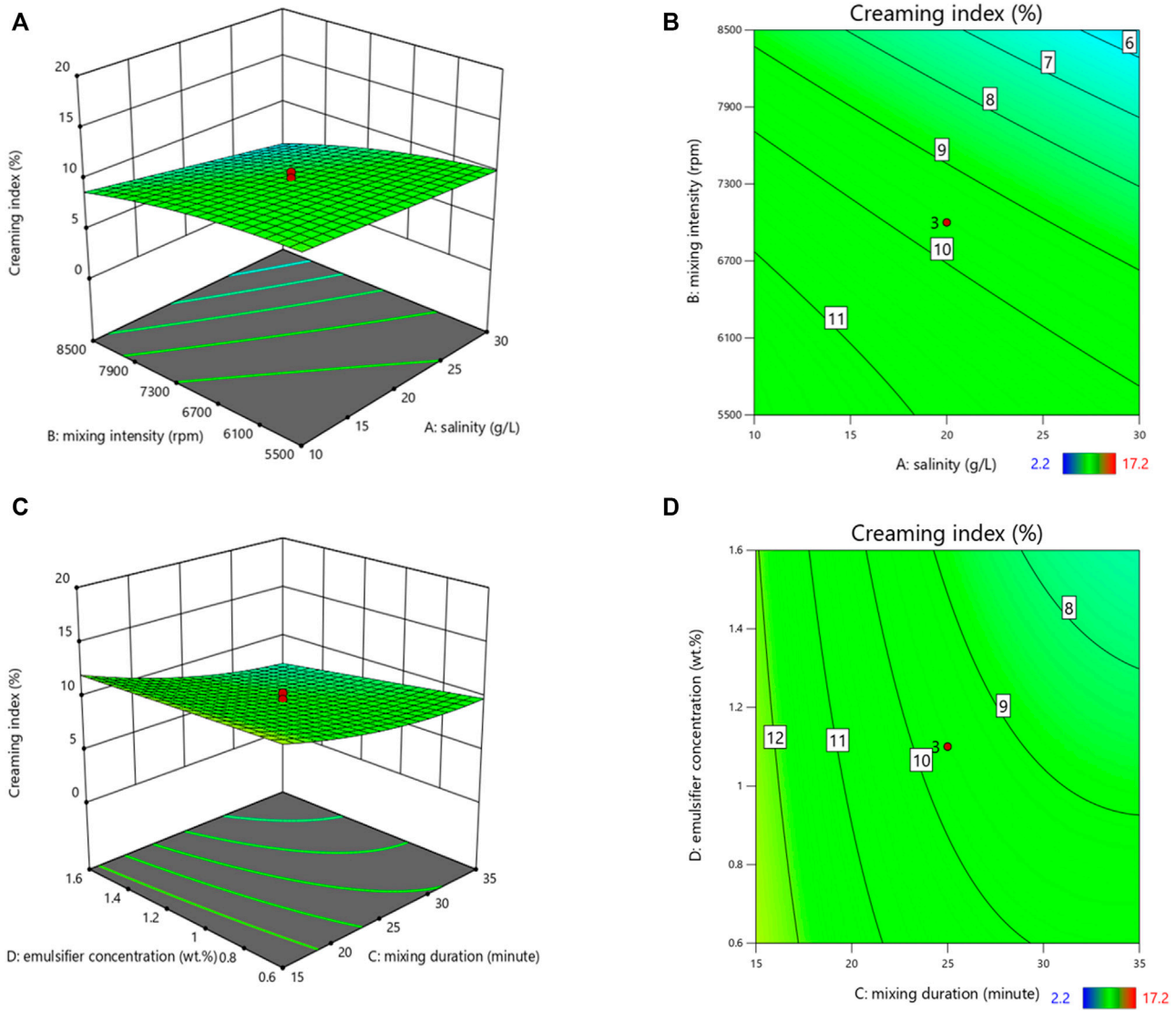


FIGURE 1 | 3D surface plots **(A,C)** and their corresponding contour plots **(B,D)** showing the effects of variables on the creaming index.

3 RESULTS AND DISCUSSION

Table 4 shows the obtained results of the response variables, including creaming index, turbidity change rate, and average oil droplet size, in each experiment.

3.1 Creaming Index

The creaming Index in this study was best described by the regression equation provided (**Table 5**), after sequential omission of the non-significant factors. The model also had a non-significant lack of fit (p = 0.195 1). To predict a quadratic polynomial model, multiple regression coefficients were made by least squares technique, and concerning the coefficient significance, the following model was proposed:

$$CI = 11.503\,28 + 0.101\,667 \times A + 0.003\,231 \times B - 0.476\,406 \times C + 1.031\,25 \times D - 0.000\,031 \times A \times B - 0.106\,25 \times C \times D - 2.752\,31 \times 10^{-7} \times B^2 + 0.008\,057 \times C^2 \quad (4)$$

where A is the salinity (g/L), B is the mixing intensity (rpm), C represents the mixing duration (minute), and D represents the Tween 20 concentration (wt%).

Results from the experiment indicated that the creaming stability was directly related to salinity, mixing intensity, mixing duration, and emulsifier concentration. Likewise, the mutual interaction between these parameters also was significant and affected the creaming Index.

TABLE 6 | ANOVA for response surface quadratic model for turbidity change rate.

Source	Sum of squares	df	Mean square	F-value	*p*-value	
Model	532.03	7	70.00	25.20	<0.0001	significant
A	43.44	1	43.44	14.41	<0.0001	
B	180.46	1	180.46	59.84	<0.0001	
C	205.74	1	205.74	68.22	<0.0001	
D	5.14	1	5.14	1.71	0.2072	
CD	20.27	1	20.27	6.72	0.0179	
B^2	9.29	1	9.29	3.08	0.0954	
C^2	55.52	1	55.52	18.41	0.0004	
Residual	57.30	19	3.02			
Lack of Fit	56.93	17	3.35	18.06	0.0537	not significant
Pure Error	0.3709	2	0.1854			
Core Total	589.33	26				

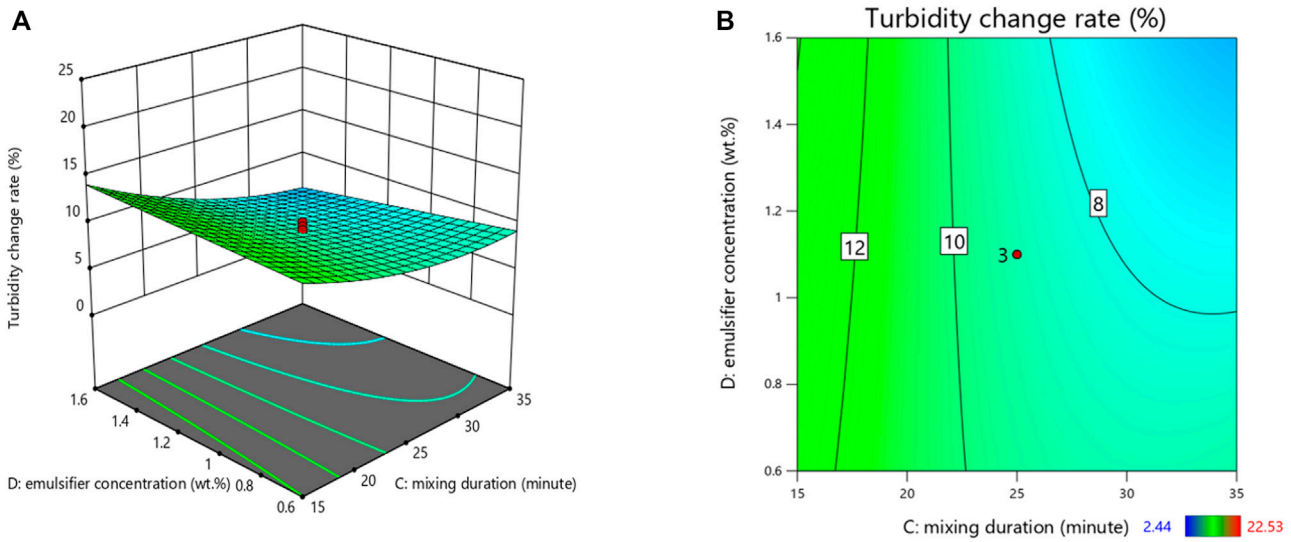


FIGURE 2 | 3D surface plot **(A)** and its corresponding contour plots **(B)** showing the effects of variables on the turbidity change rate.

It can be observed from **Figure 1** that an increase in salinity led to a decrease in CI. The experiment results were in alignment with Ling et al. (2018). They investigated the effect of salinity on the stability of W/O emulsions by measuring droplet size distribution using nuclear magnetic resonance, and demonstrated that an increase in salinity resulted in the formation of a more stable emulsion. This result is contrary to that obtained by Maaref and Ayatollahi (2018), where they prepared the W/O (medium crude oil) emulsions using a magnetic stirrer and they indicated that an increase in salt concentration led to the formation of a less stable solution due to the increased rate of aggregation and coalescence. Moreover, as the mixing duration and mixing intensity increased, the CI gradually decreased. This could be due to the production of oil droplets with smaller sizes by higher shear force, finally increasing the emulsion stability (Kundu et al., 2016). The results were also found in alignment with the results from Ashrafizadeh and Kamran (2010). Besides, it was shown from **Figure 3** that the increase in emulsifier concentration reduced CI, leading to the formation of a stable emulsion. Among all the different emulsifier concentrations used in this experiment, 1.6 wt% was the most favorable concentration for generating a stable O/W emulsion.

3.2 Turbidity Change Rate

The turbidity in this study was best described by the regression equation provided (**Table 6**), after sequential omission of the non-significant factors. To predict a quadratic polynomial model, multiple regression coefficients were made by least squares technique, and concerning the coefficient significance, the following model was proposed:

$$\begin{aligned} Turbidity\ change\ rate = {} & 22.758\,39 - 0.134\,542 \times A + 0.001\,92 \times B \\ & - 0.781\,482 \times C + 4.702\,29 \times D \\ & - 0.225\,125 \times C \times D - 2.677\,08 \times 10^{-7} \times B^2 \\ & + 0.014\,727 \times C^2 \end{aligned} \quad (5)$$

where A is the salinity (g/L), B is the mixing intensity (rpm), C represents the mixing duration (minute), and D represents the Tween 20 concentration (wt%).

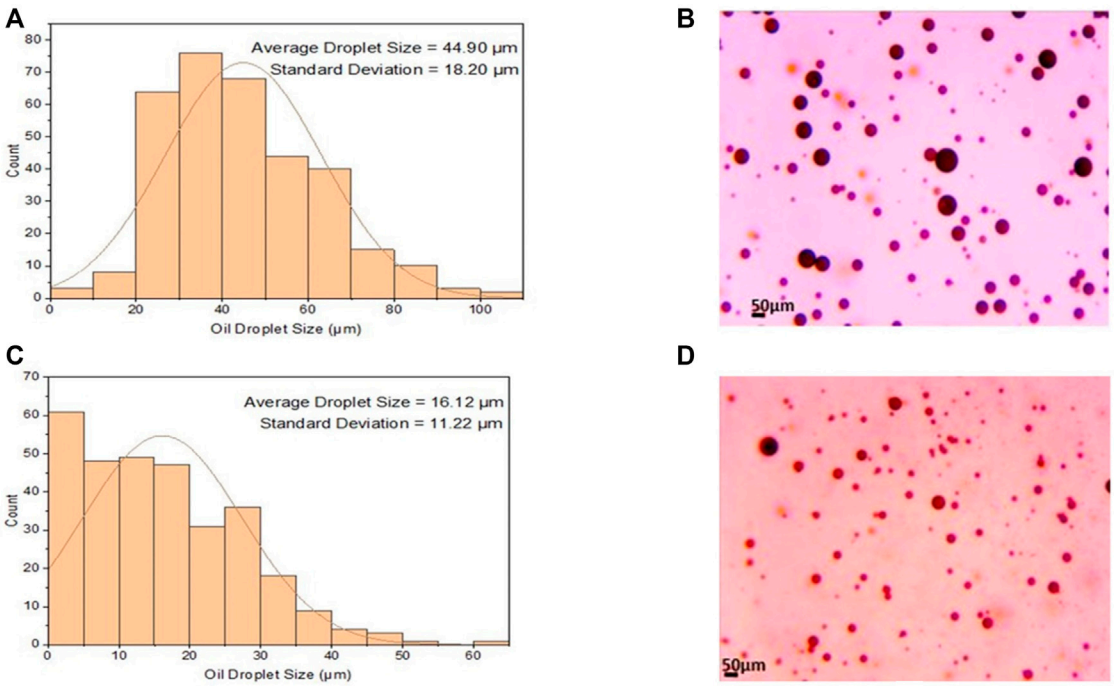


FIGURE 3 | Oil droplet size distributions and microscope images of the most stable **(C,D)** and least stable emulsions **(A,B)**.

TABLE 7 | ANOVA for response surface quadratic model for average oil droplet size.

Source	Sum of squares	df	Mean square	F-value	*p*-value	
Model	975.44	7	139.35	44.27	<0.0001	significant
A	108.67	1	108.67	34.52	<0.0001	
B	374.86	1	374.86	119.09	<0.0001	
C	302.67	1	302.67	96.16	<0.0001	
D	41.37	1	41.37	13.14	0.0018	
AB	33.5	1	33.5	10.64	0.0041	
B^2	48.98	1	48.98	15.56	0.0009	
C^2	42.56	1	42.56	13.52	0.0016	
Residual	59.81	19	3.15			
Lack of Fit	59.32	17	3.49	14.44	0.0667	not significant
Pure Error	0.4835	2	0.2417			
Core Total	1,035.24	26				

The stability of emulsions can be determined by measuring the change in turbidity with time; stable emulsion has a minor change in turbidity over time, whereas unstable emulsion has a greater change in turbidity over time. As shown in **Figure 2**, a decrease in mixing duration enhanced the rate of change in turbidity. Iqbal et al. (2020) investigated the effect of stirring duration on droplet size and emulsion stability. They found that the turbidity change rate after 24 h decreased when a longer mixing duration was applied, which is consistent with the present study. This could be attributed to a correlation between the shorter mixing time and larger, more mobile oil droplets. Hence, according to Stock's law, the pronounced upward movement of larger droplets resulted in more oil rising to the top, leading to a greater decrease in turbidity after 24 h. In addition, oil droplet size decreased with increasing Tween 20 concentrations, as the surfactant reduces droplet coalescence by producing a film at the oil-water interface. Likewise, Kundu et al. (2013) prepared O/W emulsions using diesel oil and they found that with an increase in surfactant concentration, the oil droplet size decreased. They concluded that the increased surfactant concentration resulted in an increase in the interfacial area between oil phase and water phase, which helps to form oil droplets with small size and large number thus improves the emulsion stability.

3.3 Droplet Size Index

Based on data collected (**Figure 3**), the largest average oil droplet size was 44.9 µm, which occurred at salinity of 20 g/L, mixing intensity of 7,000 rpm, mixing duration of 5 min, and emulsifier

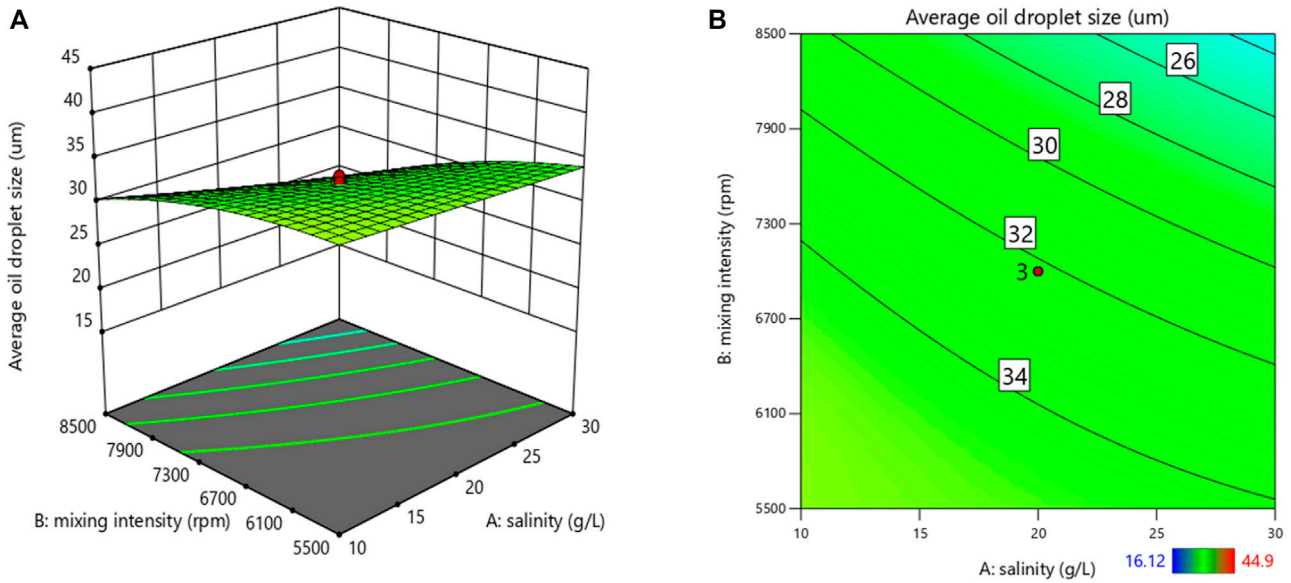


FIGURE 4 | 3D surface plot **(A)** and its corresponding contour plots **(B)** showing the effects of variables on the average oil droplet size.

concentration of 1.1 wt%. In contrast, the smallest average oil droplet size was 16.12 μm, which occurred at salinity of 30 g/L, mixing intensity of 8,000 rpm, mixing duration of 35 min, and emulsifier concentration of 1.6 wt%.

A quadratic polynomial model was proposed in terms of coded factors for predicting the average oil droplet size in **Eq. 6**.

$$\begin{aligned} \textit{Average Oil Droplet Size} = {} & 31.116\,73 + 0.462\,417 \times A + 0.007\,901 \times B \\ & - 0.999\,786 \times C - 2.625\,83 \times D \\ & - 0.000\,096 \times A \times B - 6.147\,45 \times 10^{-7} \times B^2 \\ & + 0.012\,893 \times C^2 \end{aligned} \tag{6}$$

where A is the salinity (g/L), B is the mixing intensity (rpm), C represents the mixing duration (minute), and D represents the Tween 20 concentration (wt%).

ANOVA analysis (**Table 7**) showed that the proposed model is significant with a *p*-value lower than 0.05. The R^2 is 0.942 2, which indicates that only 5.78% of the variance could not be explained by the independent variables in this regression model. The predicted R^2 of 0.863 3 is in reasonable agreement with the adjusted R^2 of 0.920 9.

As shown in **Figure 4**, the average oil droplet size in the prepared emulsion was adversely affected by the processing conditions in the mechanical homogenizer (i.e. mixing intensity), which indicated that higher mixing intensity led to more stable emulsions. Similarly, Kundu et al. (2019) also observed a decrease in oil droplet size when they increased the homogenization speed from 1,500 rpm to 4,700 rpm. They concluded that with higher mixing speed, more surfactants can be absorbed at the interface to stabilize the generated oil droplets in the emulsion. Furthermore, an increased salinity resulted in a slight decrease in the average oil droplet size which led to a more stable emulsion; a result is in line with previous studies (Esmaeili et al., 2019; Aman et al., 2015). As stated by Rocha et al. (2016), with the increase of water salinity, the IFT between the aqueous phase and oil phase decreased, reducing the driving force for droplet coalescence. In addition, increasing salinity may enhance the activity and effectiveness of natural surfactants (e.g., asphaltenes and resins) in the crude oil, which helps to stabilize the emulsion (Aman et al., 2015). Based on the study carried out by Esmaeili et al. (2019), another reason for the formation of a more stable emulsion with increased salinity might be because the addition of more salt would ionize the solution and form micelle to cover the oil droplets and prevent their coalescence.

4 CONCLUSION

In this study, conventional heavy crude oil-in-water emulsions prepared by mechanical homogenization had oil droplet sizes less than 50 μm. Second-order models were proposed to simulate the optimal formation of conventional heavy crude O/W emulsions with the minimum creaming index, lowest turbidity change rate, and smallest average oil droplet size by RSM-CCD in design expert software. The investigated independent variables all had significant effects on emulsion stability. Over the range of experimental test conditions, increase in the mixing intensity and duration as well as salinity led to increased emulsion stability and a reduction in creaming index, turbidity change rate, and average oil droplet size. In addition, it was found that the application of Tween 20 as an emulsifier had great potential in stabilizing O/W emulsions. The results showed that the combination of

30 g/L water salinity, 8,500 rpm mixing intensity, 35 min mixing duration, and 1.6 wt% emulsifier concentration achieved the most stable O/W emulsion.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

WS: Conceptualization, Methodology, Formal analysis, Investigation, Visualization, Writing—original draft preparation. NK: Conceptualization, Methodology, Formal analysis, Investigation, Visualization, Writing—original draft preparation. DM: Writing—review and editing. KL: Writing—review and editing. MZ: Writing—review and editing. JL: Supervision, Writing—review and editing.

FUNDING

This research was financially supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and Fisheries and Oceans Canada's Multi-Partner Oil Spill Research Initiative (MPRI) (Project number MPRI 4.01 and 4.03).

ACKNOWLEDGMENTS

The authors would like to thank the University of Manitoba for analyzing oil properties.

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