Abstract

Oil spilled at sea forms oil slick, which are subsequently broken up into droplets ranging from submicron to several millimeters by breaking waves. Application of dispersants to enhance the break-up process by drastically reducing the interfacial tension between oil and water has become increasingly popular in recent years. Knowledge of the resulting oil droplet size distribution is essential for determining and modeling their transport by oceanic currents and turbulence as well as their interaction with the chemical and biological environment. A major part of this experimental study involves comprehensive measurements up to five hours of the temporal evolution of the subsurface oil droplet size distributions generated by a single breaker, and their dependence on interfacial tension, oil viscosity and density, as well as breaking wave parameters. The measurements have been performed in a specially designed, 6 m long, 0.3 m wide, 0.6 m deep, totally transparent acrylic wave tank. The general entrainment processes have been visualized by high speed imaging and the temporal evolution of turbulence have been quantified using Particle Image Velocimetry (PIV). The measurements of the droplet size measurements have been performed in situ using digital inline holography at two magnifications. All early (2-10s) droplet size distributions display two distinct size ranges with different slopes. For low dispersant to oil ratios (DOR), the transition between them could be predicted based on a turbulent Weber number ($We$) in the 2-4 range, suggesting that turbulence plays an important role. For smaller droplets, all the number size distributions have power of about -2.1, and for larger droplets, the power decreases well below -3. The measured steepening of the size distribution over time is predicted by a simple model.
involving buoyant rise and turbulence dispersion. Conversely, for DOR 1:100 and 1:25 oils, the diameter of slope transition decreases from ~1mm to 46\(\mu\)m and 14\(\mu\)m respectively, much faster than the \(We\)-based prediction, and the size distribution steepens with increasing DOR. Furthermore, the concentration of micron-sized droplets of DOR 1:25 oil increase for the first ten minutes after entrainment. These phenomena are presumably caused by the observed formation and breakup of oil micro-threads associated with tip streaming whose length scale are well below the measured turbulence length scale.

In addition to subsurface oil droplet characterization, preliminary efforts have been made to elucidate the health implications of oily marine aerosol generation by breaking waves. The investigated parameters include wave energy and oil/water interfacial tension. The waves are generated every 10 s for 15 min in an attempt to mimic characteristic coastal wave periods. Results show small difference in concentration in micron range, as confirmed by two independent measurements, using an Aerodynamic Particle Sizer and digital inline holography.

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Chapter 1. Introduction
1.1. Overview

With a large amount of investment used to expand the use of renewable energy, oil nonetheless remains the world’s leading fuel. Occupying 31.9% of global energy consumption, or on average 91 million barrels daily in 2015, it will continue to be an integral source of the world’s energy, specifically for the next few generations [Dresselhaus, M.S.; Thomas, 2001; BP, 2016; U.S. Energy Information Administration, 2016]. Since 1980, a growing amount of oil has been produced or shipped at sea. Moreover, Outer Continental Shelf’s oil production has been shifting from shallow water to deep or ultra-deep water over last two decades [Anderson et al., 2012; U.S. Energy Information Administration, 2016]. Despite phenomenal success in reducing tanker and pipeline related spills over the last 40 years [Anderson et al., 2012; ITOPF, 2016], some oil inevitably has been spilled at sea. More importantly, after surveying offshore drilling history for the past 56 years, the accident rate has remained almost constant [Ismail et al., 2014], with some of these oil spills also being accompanied by gas spills of varying sizes. On average, three million gallons of oil or refined petroleum product are spilled into waters of the United States every year [National Research Council, 2005]. The most notable one is the 2010 Deepwater Horizon (DWH) oil spill, which released about 200 million gallons of crude oil into the Gulf of Mexico, approximately 60 times the annual spill rate for US waters.

Oil spills like the DWH oil spill cause extremely adverse effects to occur within marine and coastal ecosystems. When crude oil spills into the water, since two phases are immiscible, it forms a thin layer of oil slick which will then undergo a series of complicated physical, chemical and biological processes as illustrated in Figure 1.1.
[Mackay and McAuliffe, 1988; Spaulding, 1988; ASCE Task Committee, 1996; Reed et al., 1999]. In response to this, equipment such as booms and skimmers are deployed to physically control and recover the oil spills [National Research Council, 2005]. The effectiveness of this equipment is extremely dependent on the nature of the spill and oceanic conditions. After attempting to physically control the slick, oil is either recovered or in-situ burned. In-situ burning is a process limited by slick thickness, the composition of the oil, and sea conditions. At times, it is carried out by applying chemical herders, which are designed to increase the interfacial tension between the oil/water surface ($\sigma_i$) [Garrett and Barger, 1970; S.L. Ross Environmental Research Ltd., 2012; Gupta et al., 2015]. Often, these measures are only able to recover a subset of the spilled oil and rest of the oil must be dispersed quickly to a concentration that is safe to the biota. Dispersant is intended to decrease interfacial tension, promoting the spread of oil slick and enhancing the production of smaller size droplets formed by breaking waves and oceanic turbulence. In oil spill industry, dispersant become the only one tool in the toolbox to enhance the dispersion of oil spills. Since smaller droplets have much smaller quiescent rise velocity, so increasing the residence time of the oil droplets in the water column and prevent oiling the beach [National Research Council, 2005]. Though the technique is controversial [Kleindienst et al., 2015a, 2015b; Prince, 2015; Rahsepar et al., 2015; Prince et al., 2016], dispersant application has been used in many oil spills since the birth of the oil spill industry [National Research Council, 2005; Fingas, 2016]. It has become increasingly popular, less toxic, and is often used in greater quantity, particularly during large spills such as the Exxon Valdez oil spill [Shigenaka, 2014] and the DWH oil spill [King et al., 2015]. For example, the 2010 DWH oil spill used about unprecedented two
million gallons of Nalco Corexit 9500A dispersant by subsea injection and surface spraying. That’s roughly 67% of the total amount of oil spilled in US waters. Application of dispersant itself is a huge spill. Unfortunately, current major oil spill models don’t include the effects of dispersant. Thus, it is therefore essential to gain a thorough understanding of its effect on oil slick breakup due to breaking waves, and the associated droplet size distributions. Droplet size distribution is critical information after an oil spill as it is the bridge connecting small scale processes, such as droplet rise rate, dissolution and biodegradation, and large scale transport.

1.2. Dispersion of oil spill by breaking waves

Breaking waves provide the major energy source to mix up oil and water forming emulsions [Delvigne et al., 1987; Delvigne and Sweeney, 1988] as well as generate oily-marine aerosol [Ehrenhauser et al., 2014]. At times, either oil-in-water emulsions or water-in-oil emulsion will form depending on oil properties and wave conditions [Fingas, 2016]. As an example, butter is one type of water in oil emulsion, which is usually very viscous under room temperature. Milk, on the other hand, is oil in water emulsion. Formation of oil-in-water emulsions is usually called the dispersion process and begin immediately after the spill. It involves the breakup of oil droplets by breaking waves and simultaneously get entrained into the water column. Formation of water-in-oil emulsions or “chocolate mousse” is usually called the emulsification process and takes hours to days to form. It is usually formed when dispersion process is not sufficient. Stable water-in-oil emulsions usually become orders of magnitude more viscous than fresh oil and have densities approaching or slightly larger than that of sea water.
As a second transport pathway, the formation of oily-marine aerosol is less investigated and usually ignored during modeling processes [de Gouw et al., 2011; Ehrenhauser et al., 2014]. Ejection of marine aerosol into the atmosphere by breaking waves has been studied extensively for decades [Lewis and Schwartz, 2004; Veron, 2015]. Several mechanisms are involved, from the initial splashing or fragmentation of the water jet to entrainment of air into the water and resulting generation of bubbles, which rise to the surface, burst, and aerosolize droplets. The resulting aerosolized droplets are transported and dispersed in the marine atmospheric boundary layer. Depending on size of the droplets, they can be transported at various distances away from its origin. Micron sized aerosols are known to be transported inland and is the main vector for transporting bacteria and viruses across the air-sea interface [Aller et al., 2005]. Similarly, oily marine aerosol poses a health concern when the seawater surface is contaminated with crude oil. Moreover, significantly more micron-sized or submicron oil droplets are generated by applying dispersant. The potential to aerosolize these droplets poses additional health concern.

1.3. Objective

The objective of this thesis is to design and conduct a series of novel and reliable experiments to investigate and quantify the generation of subsurface and aerosolized droplets by breaking waves impacting on a controlled oil slick. The study is mainly focused on investigating the initial droplet breakup and the temporal evolution of oil droplet size distribution and their dependence on interfacial tension, oil viscosity, density, as well as breaking wave parameters. The results are intended to provide useful
information to develop new oil spill models which involves realistic account for the application of dispersant and other parameters involved.

1.4. Dissertation Outline

This dissertation consists of six chapters.

After this introduction chapter, Chapter 2 provides essential background on current study. The first part of the chapter provides brief introduction to size distribution measurement and representation. The second part presents literature review on subsurface oil droplets generation.

Chapter 3 first describes the breaking wave facility specifically designed for oil spill research at Johns Hopkins University, along with the method of generating the breaking waves used in current study. Then, details of the experimental setup and procedures are presented, including (i) flow visualization using high-speed imaging, (ii) turbulence statistics measurement using 2D PIV, and (iii) droplet size measurement using multi-resolution digital in-line holography.

Chapter 4 provides results from high-speed visualization and PIV experiments. First, macroscopic view of the breaking wave provides information on wave breaking process and droplets cloud locations and shapes. Then, microscopic views of the droplet clouds are presented. The physics underlying the observed droplets shapes and breakup processes are explained. Finally, PIV data are presented. The method of detecting the turbulent regions are firstly discussed, followed by results of temporal evolution of
energy dissipation and fluctuation velocity calculated from the 1D energy spectra for all tested waves.

Chapter 5 first reports comprehensive subsurface droplet size distributions data by varying wave energy, oil viscosity, and oil/sea water interfacial tension. Then, the preliminary results of aerosol size distribution are presented.

Chapter 6 concludes the dissertation.
Figure 1.1. Oil spill processes. [Mackay and McAuliffe, 1988]
Chapter 2.  Background and literature review
2.1 Droplet size distribution

Droplet size distribution or particle size distribution in general, can be important in understanding the physical, chemical and biological properties of the related processes. For example, settling rates, mass and heat transfer rates, entrainment and possibly coalescence rate could be inferred from the size distribution in liquid-liquid immiscible systems [Mugele and Evans, 1951]. Assume \( N \) is a continuous function of droplet diameter \( d \), representing the number concentration of all droplets smaller than \( d \), i.e. number of droplets with diameter smaller than \( d \) per unit volume. \( dN/dd \) evaluated at \( d \) is the first derivative of \( N(d) \), indicating the number concentration of droplet with diameter \( d \) per unit length. Integrating \( dN/dd \) from \( d_i \) to \( d_2 \) will give the number concentration of droplets ranging from \( d_i \) to \( d_2 \). And we call \( dN/dd \) the droplet number concentration distribution. If not otherwise mentioned, all plotted distributions in this thesis will be a discrete representation of \( dN/dd \) (i.e. \( \Delta N/\Delta d \)). The unit of the distribution will be number of droplets per unit volume per unit bin size.

Different averages of droplets system could be useful by calculating different moments of \( dN/dd \). For example, linear mean diameter (\( \bar{d}_{10} \)) can be calculated from,

\[
\bar{d}_{10} = \frac{\int_{d_1}^{d_2} d \frac{dN}{dd} dd}{\int_{d_1}^{d_2} \frac{dN}{dd} dd}
\]  

(2.1)

And volumetric mean (\( \bar{d}_{30} \)) can be calculated from,

\[
\bar{d}_{30} = \sqrt[3]{\frac{\int_{d_1}^{d_2} d^3 \frac{dN}{dd} dd}{\int_{d_1}^{d_2} \frac{dN}{dd} dd}}
\]

(2.2)
One could convert number concentration distribution into volumetric concentration distribution \( (dV/dd) \) by multiplying \( dN/dd \) by the volume the corresponding droplet \( (\pi d^3/6) \). Thus, if \( dN/dd \sim d^a \) and \( a \) changes from less than \(-3\) to larger than \(-3\) at \( d_c \), the volumetric concentration distribution increases with increasing diameter for \( d<d_c \) and decreases with increasing diameter for \( d>d_c \) and peaks at \( d_c \).

### 2.2 Relevant studies

Enormous directly relevant work seems to fall into two categories:

(i) Agitation tank experiment or numerical simulation such as [Taylor, 1934; Clay, 1940; Hinze, 1955; Acrivos, 1983; Rallison, 1984; Calabrese et al., 1986; Stone, 1994b; Gopalan et al., 2008; Liao and Lucas, 2009; Boxall et al., 2012; Perlekar et al., 2012; Skartlien et al., 2013], focus on measuring droplet size distribution or investigating breakup mechanism and transport characteristics under carefully controlled flow conditions. Some scaling trends have developed to estimate integral parameters like \( d_{50} \), which split volumetric size distribution into two equal parts or largest droplet diameter, \( d_{\text{max}} \). No unified theory to predict droplet size distribution has been developed. Besides, experiments were conducted under simple shear flow or isotropic turbulence which are not the same as breaking waves and results cannot be directly applied to predict droplet size distribution generated by breaking waves.

(ii) Second category involves measurement of oil droplet size distributions generated by waves, for example, [Delvigne et al., 1987; Delvigne and Sweeney, 1988; Lunel, 1995; Li et al., 2007, 2008b, 2009a, 2009b, 2010; Reed et al., 2009]. Results from [Delvigne et al., 1987; Delvigne and Sweeney, 1988], which are widely used in oil spill...
models [Spaulding et al., 1994; Reed et al., 1995; Lehr et al., 2002], are based on collecting oil samples at different depth and 15, 60 and 120 s after wave breaking and observing them microscopically. Their number size distributions are \( N \sim d^{-2.3} \), where \( d \) is the droplet diameter, and \( N \) is the number of droplets per diameter bin with width of \( \Delta d \). For the tested ranges of oil viscosity (8-56000 cst), density (808-992 kg/m\(^3\)) and wave energy dissipation level (1-900 J/m\(^2\)), the slope of this “power law” distribution remains unchanged. The interfacial tension remains in the 18.7-30.0 mN/m range, and the study does not investigate the effects of dispersant. The effects of oil properties and wave energy are restricted to shifts in the location of the “power law”, but not its slope. Field experiments using Phase Doppler Particle Analyzer (PDPA) by [Lunel, 1995] which include several oil-dispersant combinations and varying wind speeds, show log normal droplet size distributions. They agree with laboratory experiments involving plunging jets and waves using high-speed imaging and a light scattering based instrument (LISST-100X) [Li et al., 2007, 2008a, 2009a, 2009b, 2010; Reed et al., 2009] have conducted a series wave tank experiments investigating effects of wave energy, dispersant type, evolution with time, and temperature on the droplet sizes, also using LISST-100X. For continuously generated waves, they find that the oil-based dispersant Corexit 9500 is more effective in reducing the droplets size than the water-based dispersant SPC 1000. Their time averaged droplet size distributions change from log-normal without dispersants to bi-modal log-normal with one peak below 10 µm, and the other in the 10-100 µm range, depending on wave properties. Combining data for nonbreaking, spilling and plunging breaking waves, they introduce empirical relations for the oil concentration, \( c \), as a function of turbulence dissipation rate \( (\varepsilon) \), below the wave, \( c \sim \varepsilon^{-0.32} \) and \( c \sim \varepsilon^{-0.43} \), for
oils without and with dispersants, respectively. These studies focus on overall entrainment rate and prediction of integral parameters such as $d_{50}$ of continuous generated waves over a large parameter space.

Moreover, there is a large body of literature involving air bubbles and marine aerosol measurements in wave tanks aimed at characterizing air-sea interactions. The distributions and dynamics of bubbles both immediately after their formation [Deane and Stokes, 1999, 2002] and 10-100s seconds later [Thorpe, 1984, 1992; Medwin and Breitz, 1989; Monahan and Lu, 1990] have been measured. These distributions consist of two power “laws”, the first having a mild slope for small bubbles, and the second having a steeper slope for large bubbles. The transition between them occurs at the Hinze scale, where turbulent fragmentation stops. [Deane and Stokes, 2002] claims that the large bubbles are generated by turbulent fragmentation, and the small ones, by jets and drops impacting on the water surface. Adopting a dimensional argument proposed by [Garrett et al., 2000], they predict the -10/3 slope for large bubbles. The -3/2 slope for small bubbles is not mechanistically explained. The present paper summarizes systematic in-situ measurements using digital inline holography of the temporal evolution of droplet size distribution generated by entrainment oil slicks by single 2D breaking waves. The experiments are performed for varying wave energy and oil properties, including the orders of magnitude changes to interfacial tension caused by premixing the oil with dispersant. In accordance with phenomena occurring during bubble entrainment by waves, [Rapp and Melville, 1990; Deane and Stokes, 2002], the measurements cover both the initial rapid entrainment and fragmentation phase, when the oil forms distinct clouds, and then extend to the slower dispersion phase, when the transport of oil droplets is
dominated by turbulent diffusion and buoyant rise. The results show that dispersant changes the size distribution slopes, the characteristic droplet sizes, and increases the concentration of micron-sized droplets by orders of magnitude.
Chapter 3. Methods and materials
3.1 Wave Tank

The experiments have been carried out in a specially designed 6 m long, 0.3 m wide, 0.6 m deep and 2.5 cm thick totally transparent acrylic wave tank. The 3D model of the wave tank together with the digital inline holography are shown in Figure 3.1. The tank has four openings with size of $112 \times 15$, $112 \times 15$, $112 \times 15$, and $175 \times 30$ cm$^2$ respectively. During experiments, these openings are fully covered for health concerns. Details of the design including the wave tank assembly and parts drawings are provided in Appendix A. Figure 3.2 shows the schematic of the wave tank. To simulate oceanic condition, wave tank is filled with artificial sea water with a salinity of 33 parts per thousand. It is prepared by mixing around 68kg of Instant Ocean Aquarium Sea Salt with filtered tap water in a 500-gallon mixing tank equipped with a 2-HP pump. The mixture is simultaneously filtered by 2-step (10 µm and 1 µm) Purenex sediment filter. The mixing and filtering process usually take at least 10 hours to finish. Salinity of the water samples taken both from the bottom and top outlet of the mixing tank are measured using a salinity meter to make sure the mixture is homogenous. Breaking waves have been generated mechanically by a piston type wave maker consisting of a vertical aluminum wave plate that extends over the entire cross section of the tank. The wave plate is driven by an Exlar GS60-1010 roller-screw linear electric actuator coupled with a lever system which magnifies the displacement by 1:6, enabling a maximum wave plate stroke of 1.5 m. The linear actuator specifications are provided in Table 3.1. The actuator motion is controlled by an Emerson AC Unidrive SP-2403 along with a control software package PowerTools Pro v5.2. The communications between a laptop and the drive is established using an Ethernet cable. Calibrations involving high-speed imaging of the plate indicate
that the rms error for the wave plate displacement is less than 0.9 cm, corresponding to 2% of the condition of the present study. The coordinate system used for presenting data is also illustrated in Figure 3.2, with the origin located on the undisturbed water surface, the x-axis pointing downstream, and z-axis upward. The corresponding instantaneous velocity components are $u$ and $w$, respectively, with capitalized letter (e.g. $U$) representing spatially averaged quantities, and angle brackets, e.g. $<u>$, denoting ensemble averaging. For high speed imaging, undisturbed water depth ($h$) of 25cm and 20cm are used. For PIV and digital inline holography experiments, the same $h$ of 25cm has been maintained for all the experiments. The breaking waves are generated by translating the wave plate according to $x_p=0.5A(1-\cos2\pi ft)$, where $f, A$ and $t$ are frequency, stroke, and time, respectively.

To investigate the effect of a single wave on the time evolution of droplet size distributions, each experiment involves a single push of the wave plate for a half period, i.e. $0 \leq t \leq (2f)^{-1}$. The tank does not have a wave damper, so after breaking once, the wave remnants are reflected back and forth, as the energy is dissipated by the (measured) wave generated turbulence. The wave plate velocity, $V=dx_p/dt=A\pi f \cos2\pi ft$ and the maximum wave plate velocity $V_{\text{max}}=A\pi f$. The wave height and characteristics are controlled by varying $V_{\text{max}}$, or rather, different combinations of wave stroke and wave frequency, and the resulting waves vary from gentle spilling breakers to violent plunging breakers. Figure 3.3 illustrates the relationship between maximum wave height before breaking ($H$) and $V_{\text{max}}$ normalized by $h$ and the shallow water wave speed, $(gh)^{1/2}$, respectively. $H$ is measured using high speed imaging results. As is evident, the data collapses onto a single line. As $V_{\text{max}}$ increases, the breaking process changes from spilling to plunging. The
approximate transition is indicated by the arrows. The measured trends are compared to those of a hydraulic jump, i.e. solving $H$ and $c$ given $V_{\text{max}}$ from $(H+h)/h=\frac{1}{2}+(1+8c^2/gh)^{0.5}/2$ and $ch=(c-V_{\text{max}})(H+h)$ for $h=25\text{cm}$. As shown, the hydraulic jump relations underestimates the wave height, and the difference between them increases with increasing $V_{\text{max}}$. The rate of energy dissipation per unit width for a hydraulic jump is $E_{\text{hj}}=0.25(\rho gcH^3)/(H+h)$. A factor $\alpha$ ranging between 1.3-1.5 for a stable breaking wave on a gentle slope [Stive, 1984], or 0.01-0.04 for unstable white capping and wind-driven waves in open ocean [Longuet-Higgins and Turner, 1974] has been added to this expression in previous studies to account for the difference between breaking waves and in hydraulic jump. Another interesting feature of this plot is that the spilling and plunging breaker transition happens to be around $H/h=0.8$. By using solitary wave theory, [M'Cowan, 1893] theoretically determined the maximum value for $H/h$ to be 0.78 which is very close to our measured transition between spilling and plunging breakers.

For subsurface droplet size experiment, two plunging breakers and one spilling breaker have been selected, named as wave 1-3 respectively, for the investigation of the effects of wave energy on oil droplet size distributions, and their characteristics are summarized in Table 3.2. The wave characteristics are determined based on analysis of high-speed images involving multiple synchronized cameras, as discussed below. Included parameters are $A$, $H$, $V_{\text{max}}$, $f$, $E_{\text{hj}}$ as defined above and the location where wave breaking is initiated ($x_B$), the speed of the wave crest before breaking ($c_w$), the speed of the leading edge of the turbulent wave front after breakup averaged from $x=166$ to 307 cm ($c_t$), and the oil droplets initial intrusion depth ($z_i$). The latter refers to the visually observed deepest penetration of the oil droplets within the first 1.3s after initiation of the
wave plate. Wave speed dependent values of $E_{hj}$ are calculated using the measured $c_w$. As is evident, $E_{hj}$, $c_t$, $c_w$, $z_i$, increase with increasing wave stoke. For aerosol droplet size experiment, wave 1 and 2 are used and have been continuously generated every 10s for 15min to mimic the oceanic wave periods.

### 3.2 Oil and dispersant

In order to achieve a range of physical properties, three types of oils, namely crude oil (MC252 surrogate [Pelz et al., 2011]), fish oil (cod liver oil made by Twinlab) and motor oil (made by Service Pro) have been used to investigate the effects of viscosity. As indicated in Table 3.3, their kinematic viscosity ($\nu = \mu / \rho$, $\mu$ and $\rho$ being the dynamic viscosity and density, respectively) varies over orders of magnitude, and the fish oil is a little heavier than the other two. The viscosity data is based on measurements using several Cannon-Fenske opaque viscometers (Cannon Instrument Company). To investigate the effect of interfacial tension on the droplet size distribution, the dispersant Corexit 9500A (Nalco) has been premixed with Surrogate MC252 Oil at DOR of 1:25, 1:100, 1:500 and 0, the latter being crude oil without dispersant. The oil-dispersant mixtures will be identified by the corresponding volumetric Dispersant to Oil Ratio (DOR) level, for example, DOR 1:25 oil, in the rest of the dissertation. Surface tension with air ($\sigma$) for all oils and interfacial tension with seawater ($\sigma_i$) for oils without dispersant have been measured using the pendant drop method [Song and Springer, 1996; Murphy et al., 2015]. When a pendant droplet reaches hydrodynamic and mechanical equilibrium, gravitation force should be equal to interfacial tension force and relations between shapes and forces could be established. The interfacial tension is calculated by measuring the shape of a drop hanging from the end of a needle. A camera is equipped to
photograph the drop and is subsequently analyzed by an in-house software. Since $\sigma_i$ has been reduced significantly for dispersant and oil mixture, pedant drop method could not be formed and $\sigma_i$ for DOR 1:25, 1:100 and 1:500 oils are estimated by measuring the oblateness and quiescent rise rate of the oil droplet [Hu et al., 2000; Gopalan and Katz, 2010; Murphy et al., 2015]. The corresponding orders of magnitude changes to $\sigma_i$ are provided in Table 3.3. Values for $\sigma$ are also provided in Table 3.3.

During experiments with oils, oil is initially maintained inside a rectangular area by a slightly submerged open confinement ring centered at $x_o=150$ cm. This enclosure is connected to a pneumatic cylinder, which is synchronized with the wave plate, and pulls the oil slick confinement ring up immediately after the initiation of the wave plate motion. Two rectangular rings with streamwise and spanwise dimensions of 40×5 cm and 25.4×2.54 cm, the former for high-speed visualizations, and the latter for measuring the oil droplet size distributions. Both rings are 3D printed and have wall thickness of 0.05 cm to minimize the flow disturbance when they are pulled out from the water surface. For visualizations, 10 mL of oil is carefully injected using a syringe, and the oil is allowed to spread over the confined area, resulting in an average slick thickness of 0.5 mm. For droplet sizing experiments, the area is reduced to reach a oil concentration that could be readily measured. In this case, 3.5 mL of oil is carefully introduced into the confined area by a 5 mL syringe, resulting in an oil slick with the same 0.5 mm thickness. For each of the waves, we have also conducted experiments without oil to obtain size distributions about the entrained bubbles. After each experiment with oil, the oily water surface is first skimmed using multiple absorbent pads (New Pig). Then, the tank is drained, washed multiple times with Oil Eater Degreaser solution (Kafko International LTD.), and rinsed
and residue oil slick skimmed several times with tap water until we no longer see visual signs of an oil sheen. Details of cleaning procedure used and time consumption are summarized in Table 3.4.

3.3 High Speed Imaging and Particle Image Velocimetry

Multiple setups of high speed imaging have been implemented. For \( h = 25 \text{cm} \) waves, the high-speed visualizations have been conducted in two ways. First, the wave impingement on the oil slicks and the subsequent breakup and dispersion processes are simultaneously visualized by three \( 2016 \times 2016 \) pixels high-speed cameras (PCO.dimax) operating at 500 fps. Camera 1 is equipped with a 28mm Nikon lens and cameras 2 and 3 are equipped with 35mm Nikon lenses. The fields of view (\( FOV \)), \( 103 \times 103 \), \( 75 \times 75 \), and \( 75 \times 75 \text{cm}^2 \), are centered at \( x = 166, 243, \) and \( 307 \text{cm} \) for cameras 1, 2, and 3, respectively. The vertical location is chosen so that the initial water surface shrink to a line, i.e. at \( z = 0 \). Back lighting by a total twelve 500 Watt Halogen bulbs is used for imaging the wave motion and oil entrainment over the entire tank. The lamps are located away from the tank wall, and the light is diffused by translucent paper mounted on the wall. Second, processes of wave 2 propagation along the whole tank has been visualized by a single \( 2016 \times 2016 \) pixels high-speed camera (PCO.dimax) equipped with 35mm Nikon lenses. Eight locations centered at \( x = 148.6, 208.6, 269.3, 330.4, 391.4, 452.5, 513.5 \) with \( FOV \) of \( 75 \times 75 \text{cm}^2 \) have been chosen for visualization. Back lighting is reduced to a total six 500 Watt Halogen bulbs. For all locations, the camera is synchronized with the wave plate, operating at 250 fps. For \( h = 20 \text{cm} \) waves, the processes are visualized by two \( 2016 \times 2016 \) pixels high-speed cameras (PCO.dimax) operating at 250 fps. The \( FOV \) of \( 118 \times 118 \) and \( 90 \times 90 \text{cm}^2 \), are centered at \( x = 155 \) and 230cm for cameras 1, 2, respectively.
Camera 1 is equipped with a 28mm Nikon lens and cameras 2 is equipped with 35mm Nikon lens. Back lighting by A total eight 500 Watt Halogen bulbs are used for back lighting.

Two-dimensional PIV is used for characterizing the turbulence in the tank after wave breaking. The sample area is centered at $x_c=320\text{cm}$ and $z_c=-11.1\text{cm}$, the same location as that used for much of the droplet sizing. The $FOV$ of PIV is $4.6\times4.6\text{ cm}^2$. The flow field is illuminated by a dual head, 200 mJ/ pulse, Nd:YAG laser. The delay between exposures for waves 1 and 2 is 5ms, and that for wave 3 is 8ms. The $2016\times2016$ pixels image pairs are acquired at 15Hz for a period of 100s by a PCO.dimax camera equipped with a 60mm Nikon lens. During data analysis, the PIV images are enhanced using background subtraction, median filtering and a modified histogram equalization algorithm [Roth et al., 1999; Roth and Katz, 2001]. Images are then processed by multipass, FFT-based cross-correlations, using the LaVison© DaVis software package. The final interrogation window size is $24\times24$ pixels with 50% overlap, corresponding to a vector spacing of 0.275 mm. Based on prior measurements at similar resolutions and seeding density performed in our laboratory, the averaged uncertainty in instantaneous velocity is 0.2 pixels, corresponding to 2%. Six independent data sets have been processed for each of the three waves, each consisting of 1500 velocity distributions. The data are used for calculating the time evolution of turbulence in the tank, as discussed later.
3.4 Digital inline holography

Holography is a 3D imaging technique, which consists of recording the interference patterns between a reference beam and light scattered by objects in the volume of interest, with orders of magnitude better depth of field than conventional photography [Vikram, 1992; Hariharan, 1996]. It has been widely used to measure the 3D location, motion, and size of particles, droplets, bubbles, and planktons in numerous applications, too many to summarize in a single paper [Katz and Sheng, 2010]. In particular, size distributions have been measured by e.g. [Thompson, 1974; Katz, 1984; Ran and Katz, 1991; Malkiel et al., 1999; Fugal and Shaw, 2009; Tian et al., 2010; Gao et al., 2013; Beals et al., 2015]. In this study, the subsurface droplets size distribution is quantified using digital inline holography at magnifications (MA) of 10 and 1. Using the abovementioned PCO.dimax camera, the corresponding resolutions are 1.1 and 11.1\(\mu\)m/pixel, respectively. The light source of the digital holography system is pulsed high frequency, low energy Nd:Yag laser (CrystaLaser, model QL532-500, 61 \(\mu\)J/pulse at 1kHz). The beam is attenuated to 0.6 \(\mu\)J/pulse by a ND 2.0 filter, spatially filtered, collimated, and split into two beams. Data have been acquired at four different locations, which are identified as 1-4. The coordinates of their centers (\(x_c, z_c\)) are illustrated in Figure 3.2, and listed in Table 3.5 along with the corresponding sample volumes.

3.4.1. Optical setup

The optical setup for simultaneous measurement of droplet size distributions at \(MA=10\) in location 3 and \(MA=1\) in location 4 is illustrated in Figure 3.4. Unless specified, most of the data presented in this paper has been recorded in these sites since the main
droplets clouds pass through them. Only the $MA=1$ beam is used for locations 1 and 2, the and they are used for evaluating depth and location effects on the droplet size distributions. For the $MA=1$ measurements, a pair of relay lenses (250mm focal length, 76 mm in diameter) separated by twice their focal length are placed outside of the tank to position the plane of focus, or the hologram plane, in the middle of the tank at $y=0$. This approach is used for measuring droplets with diameters larger than 22.2 µm (2×2 pixels). For $MA=10$, the micro-droplets have to remain within about 1000 diameters from the plane of focus of the microscope objective to be resolved well [Sheng et al., 2006]. Furthermore, for the $DOR$ 1:25 and 1:100 oils, the dense cloud of micron-sized droplets could only be observed if the depth of the sample volume is restricted to no more than a few millimeters. Hence, the $MA=10$ setup involves two submerged 50mm diameter hollow tubes with windows on both sides, with an 8mm gap between them in the center of the facility. The other side of both tubes is flush with the inner wall of the tank. The measurements are performed within the central 6 mm of the tank. The 10× infinity corrected objective (Mitutoyo) coupled with a 200mm imaging tube lens is installed in one of the submerged tubes. It has a working distance of 33.5 mm and its location inside the tube is allowed for fine adjustment so the hologram plane is located at $y=0$. Since the narrow gap might prevent big droplets from entering the $FOV$, the measurements at $MA=10$ have been restricted to 2.2-143 µm (2-130 pixels) droplets. The holograms have been recorded at different frame rates, depending on objectives. Most of the data presented in this paper has been recorded at 50 frames per second (fps), but holograms have also been recorded at 500 fps to visualize the initial droplet clouds in locations 1 and 2. In addition, the evolution of droplet statistics has been measured for periods of up to
five hours using acquisition rates varying between 0.5 to 10 fps. For aerosol size distribution measurement, holography data have been acquired at 3.3 fps, $MA=10$ for 15 min at location 3.

3.4.2. Image processing

Data processing consists of three phases, namely preprocessing, reconstruction, and droplet size measurements. Preprocessing involves: (i) homogenizing the intensity distribution in each hologram using the median filtered intensity as a basis for scaling the gray levels. Larger intensity variation from for example illumination inhomogeneity could be corrected through this step. (ii) calculating the time averaged intensity of the scaled holograms and subtracting it from the instantaneous fringe patterns. Figure 3.5a-c provides sample raw hologram, enhanced hologram (or illumination corrected hologram), and background subtracted hologram. As is evident, after background subtraction, features that don’t change over time including scratches and droplets on the tank wall will be filtered out.

The holograms are then digitally reconstructed plane by plane using the Fresnel-Huygens paraxial approximation [Katz and Sheng, 2010]. A recently developed in-house GPU-based code, which speeds up the reconstruction time by 130, is used for obtaining reconstructed images along $y$ axis every $500\mu m$ for $MA=1$ and $10\mu m$ for $MA=10$. When the depth direction is defined as $z$, the digital reconstruction process of hologram is:

$$
\mathcal{U}_r(x, y, z) = \int_{\xi_a}^{\xi_b} \int_{\eta_a}^{\eta_b} \mathcal{U}_r(\xi, \eta, z = 0) \left[ \frac{\partial g}{\partial n}(x - \xi, y - \eta, z) \right] d\xi d\eta
$$

(3.1)
Here, $\bar{U}_r(\xi, \eta, z = 0)$ is the optical field of hologram, $\bar{U}_r(x, y, z)$ is the reconstructed optical field and $-\frac{\partial g}{\partial n}(x - \xi, y - \eta, z)$, the kernel function, is normal derivative of Green’s function of wave equation propagation in a homogeneous medium. Two kernel functions are frequently used, i.e. the Rayleigh-Sommerfeld formula and the Kirchhoff-Fresnel formula. In our digital reconstruction, the latter is used.

$$-\frac{\partial g}{\partial n}(x, y, z) = \frac{\exp(jkz)}{j\lambda z} \exp\left[j \frac{k}{2z} (x^2 + y^2)\right]$$

(3.2)

The convolution (3.1) can be performed either by spatial integration or by multiplication in frequency domain shown as follows:

$$\bar{U}_r(x, y, z) = \mathcal{F}^{-1}\left[\mathcal{F}(\bar{U}_r) \cdot \mathcal{F}\left(-\frac{G}{\partial n}\right)\right]$$

(3.3)

Where $\mathcal{F}$ denotes the Fourier transform. Digitally, since the (inverse) Fast Fourier Transform and multiplication are highly parallel, the reconstruction is implemented on a low cost Tesla K40c GPU board using C++ based CUDA toolkit 6.5 developed by the NIVIDIA corporation. During the reconstruction, hologram data containing both real and imaginary part is copied from CPU memory(host) to GPU global memory(device), Fourier transformed via calling the cufft complex to complex transform functions in the CUFFT library, and multiplied by the Kirchhoff-Fresnel kernel which is generated and recorded in advance on GPU,

$$\mathcal{F}\left(-\frac{G}{\partial n}\right) = j\lambda \exp\left\{-j\lambda z \pi \left[\left(\frac{m}{M\Delta x}\right)^2 + \left(\frac{n}{N\Delta x}\right)^2\right]\right\}$$

(4)
Where $M$ and $N$ are the total number of discretization points along the $x$ and $y$ axis, respectively. The result is inverse Fourier transformed using cufft complex to complex transform, yielding the reconstructed complex optical field. The reconstructed images are intensity of the optical field which is normalized to 8-bit gray scale image and transferred back to host. A simple numerically generated hologram of a single particle in space is utilized to test the accuracy of our code and a hologram image in the holographic PIV experiment whose size is 2048x2048 ($M \times N$) is used to validate the efficiency by comparing with the CPU-based reconstruction method. The clock rate and number of processors of our GPU board are 745 MHz and 2880 respectively, while the Intel i7-3770k processor for CPU has a clock rate of 3.5 GHz. Runnings of multiple plane reconstruction show that 400 ms is needed on average for a single plane reconstruction when implemented on CPU, however, the corresponding running time on GPU is reduced to around 3 ms, yielding a speed up ratio of about 130. The above calculations are based on 8-bit gray scale image. When it’s applied to 16-bit gray scale image, the data copy time from device to host and the image writing time will be doubled. These time variations are small comparing to the total time cost on CPU, however, it’s large comparing to the total time cost on GPU, resulting a reduction of speed up ratio to nearly 100.

After digital reconstruction, to identify regions of interest containing droplets, the reconstructed volumes are collapsed into planes containing the minimum intensity in each pixel, thresholding the collapsed images, and performing two-dimensional segmentation that defines the approximate boundaries of droplets. This procedure is performed in two steps, first using the original depth-compressed minimum intensity map.
Second, after high-pass filtering each of the reconstructed planes to remove the large particles, depth-compressed minimum intensity map of these filtered reconstruction map shows clear traces of small particles. The results are used for guiding the regions requiring further analysis that measures the droplet coordinates and size based on their in-focus images. The collapsed maps also serve as a convenient method for examining the temporal evolutions in droplet concentrations as clouds cross the sample area. Figure 3.6a-d provides sample image of original minimum intensity map, high-pass filtered minimum intensity map, segmented larger than 3 pixels large droplets, and segmented smaller than or equal to 3 pixel small droplets, respectively.

The size measurements are based on the in-focus reconstructed images. A circular Hough-transform [Duda and Hart, 1972; Illingworth and Kittler, 1988; Yuen et al., 1989] is used for determining the size and center of droplets larger than 5 pixels. It involves three steps, namely accumulator array computation, center estimation, radius estimation. Figure 3.7 demonstrate the basic algorithm of a circular Hough-transform. Foreground pixels with high gradient are selected as candidate pixels and are allowed to cast ‘votes' in the accumulator array. In Figure 3.7a, edge pixel will become the candidate pixels vote for accumulator arrays. Dash line shows the all possible vote from one edge pixel. Figure 3.7b shows, if there is a circle in the image, the center will stand out as the maximum in the accumulator arrays. A matlab function CircularHough are used [Peng, 2010]. The circular Hough-transform is performed for every reconstructed plane to search for possible circles. A gradient level needs to be specified to calculate the accumulator array. The threshold choices are dependent on the image quality and usually a very low threshold is chosen, so unclear circles will be detected. As a result, many circles will be
detected on different reconstruction planes. To eliminate the false one and to fine the plane of focus, six criteria involving edge gradients, as well as the mean, spatial rms, and intensity variations within the detected circles are used. A set of parameter are created based on test runs and needs to be checked and modified again for every data sets. The corresponding size uncertainty is about 1 pixel, corresponding to 1.1μm for MA=1 and 11.1μm for MA=10. For droplets smaller than 6 pixels, the Hough-transform has been found to be unreliable. Consequently, the equivalent droplet diameter is determined by applying a contrast-dependent Otsu thresholding with 0.3-0.4 scaling [Otsu, 1979] on the segmented collapsed minimum intensity images, and measuring the binary image areas. The location of in-focus plane is determined based on the planes with minimum intensity. The analysis based on the Hough transform gives the radius in discrete number of pixels, and based on visual observations, the estimated uncertainty is also about 1 pixel, corresponding to the above-mentioned magnification-dependent scales. To insure the data integrity, the detected droplets are compared visually to the compressed fields by overlaying them. The ImageJ software package [Schneider et al., 2012] is used for manually scanning, visually detecting and deleting the few false positives (1%-5%), depending on droplet concentration). Subsequently, droplets that are not detected automatically (~3%) are added by scanning through the 3D reconstructed volume, and using the Measure-And-Label plugin of ImageJ to fit circles around the focused images of the undetected droplets. To prevent bias in number size distributions, each diameter bin is chosen to include the same ratio of two between the number of discrete data points included in it and the bin width. Both increase gradually with size, and the same intervals
are used throughout this paper. The middle point for each bin is designated as the representative size.
Figure 3.1. 3D model of the wave tank and the digital inline holography setup.
Figure 3.2. Schematic of the wave tank.

Table 3.1. Performance specifications of GS 60-1010 linear actuator.

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>Stroke (mm)</td>
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</tr>
<tr>
<td>Force Ratings (N)</td>
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<tr>
<td>Maximum Velocity (mm/s)</td>
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<td>Continuous Motor Torque (N.m)</td>
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<td>Maximum Static Load (N)</td>
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<tr>
<td>Dynamic Load Rating (N)</td>
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</table>
Figure 3.3. Relationship between wave height and maximum wave plate speed. Arrows indicate the approximate transition between spilling and plunging breakers.

Table 3.2. Wave characteristics used in the present experiments.

<table>
<thead>
<tr>
<th>Wave</th>
<th>$A$ (cm)</th>
<th>$H$ (m)</th>
<th>$V_{max}$ (m/s)</th>
<th>$x_B$ (cm)</th>
<th>$c_w$ (m/s)</th>
<th>$c_t$ (m/s)</th>
<th>$f$ (Hz)</th>
<th>$z_i$ (m)</th>
<th>$E_{hj}$ (Jm$^{-1}$s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>53.3</td>
<td>0.29</td>
<td>1.26</td>
<td>144.6</td>
<td>2.41</td>
<td>2.79</td>
<td>0.75</td>
<td>0.17</td>
<td>271.5</td>
</tr>
<tr>
<td>2</td>
<td>45.7</td>
<td>0.25</td>
<td>1.08</td>
<td>139.7</td>
<td>2.21</td>
<td>2.37</td>
<td>0.75</td>
<td>0.13</td>
<td>172.2</td>
</tr>
<tr>
<td>3</td>
<td>45.7</td>
<td>0.22</td>
<td>0.90</td>
<td>134.5</td>
<td>2.02</td>
<td>2.19</td>
<td>0.625</td>
<td>0.07</td>
<td>114.1</td>
</tr>
</tbody>
</table>
Table 3.3. Measured Properties of the artificial sea water, different oil types and crude oil premixed with dispersant COREXIT 9500A at various DORs.

<table>
<thead>
<tr>
<th>Fluid</th>
<th>Density $\rho$ (kg/m$^3$)</th>
<th>Kinematic Viscosity $\nu$ (cSt)</th>
<th>Interfacial Tension with seawater, $\sigma_i$ (mN/m)</th>
<th>Surface Tension with air $\sigma$ (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Artificial seawater</td>
<td>1018.3</td>
<td>1.1</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>Crude Oil</td>
<td>877</td>
<td>9.4</td>
<td>19</td>
<td>28</td>
</tr>
<tr>
<td>Crude Oil DOR 1:500</td>
<td>877</td>
<td>10.1</td>
<td>2.35</td>
<td>28</td>
</tr>
<tr>
<td>Crude Oil DOR 1:100</td>
<td>877</td>
<td>10.6</td>
<td>1.2</td>
<td>28</td>
</tr>
<tr>
<td>Crude Oil DOR 1:25</td>
<td>877</td>
<td>12</td>
<td>0.28</td>
<td>28</td>
</tr>
<tr>
<td>Fish Oil</td>
<td>924.4</td>
<td>63.1</td>
<td>14.9</td>
<td>22.5</td>
</tr>
<tr>
<td>Motor Oil</td>
<td>877.6</td>
<td>306.5</td>
<td>19</td>
<td>24.7</td>
</tr>
</tbody>
</table>
Table 3.4. Cleaning procedures.

<table>
<thead>
<tr>
<th>Time</th>
<th>Cleaning Procedure</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 hr</td>
<td>1. Wear mask and gloves, use oil absorbent pad to skim surface oil to minimize the effort of cleaning tank walls. (1 hr)</td>
</tr>
<tr>
<td></td>
<td>2. Drain the tank fully, use oil eater to wash away residues by using micro fiber mop/towels (1.5 hr)</td>
</tr>
<tr>
<td></td>
<td>3. fill and drain the tank simultaneously to get rid of most of the surfactant and oil. (1.5 hr)</td>
</tr>
<tr>
<td>1.5 hr × (1-3 iterations)</td>
<td>Refill the tank fully and drain the tank until no oil sheen is observed. When refilling, use clean micro fiber mop to clean the tank surface, and use absorbent pad to skim surface slick.</td>
</tr>
<tr>
<td>1 hr</td>
<td>Rinse sediments on the bottom of the tank away.</td>
</tr>
<tr>
<td>1 hr</td>
<td>Optics cleaning (only for inline holography ( M4 = 10 ) experiment)</td>
</tr>
<tr>
<td>0.5 hr</td>
<td>Dry and re-grease the rail if needed.</td>
</tr>
</tbody>
</table>
Figure 3.4. The multi-resolution digital holography setup used for measuring the droplet size distributions. The sample volume centers are located at (355, 0, -11.1) for MA=10 and (320, 0, -11.1) for MA=1. The oil patch is centered at $x_o=150$ cm. Unless specified, droplet size distributions data are from these two locations.
Table 3.5. Locations and sample details of the holographic droplet sizing experiments. $x-x_o$ is the horizontal distance between the sample and the oil slick centers.

<table>
<thead>
<tr>
<th>Location</th>
<th>$x_c$ (cm)</th>
<th>$z_c$ (cm)</th>
<th>$x_c-x_o$ (cm)</th>
<th>Resolution (µm/pixel)</th>
<th>Sample depth (cm)</th>
<th>Sample Volume per hologram (mL)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>270</td>
<td>-1.1</td>
<td>121</td>
<td>1</td>
<td>11.1</td>
<td>30</td>
</tr>
<tr>
<td>2</td>
<td>270</td>
<td>-11.1</td>
<td>121</td>
<td>1</td>
<td>11.1</td>
<td>30</td>
</tr>
<tr>
<td>3</td>
<td>355</td>
<td>-11.1</td>
<td>205</td>
<td>10</td>
<td>1.1</td>
<td>0.6</td>
</tr>
<tr>
<td>4</td>
<td>320</td>
<td>-11.1</td>
<td>170</td>
<td>1</td>
<td>11.1</td>
<td>30</td>
</tr>
</tbody>
</table>
Figure 3.5. Hologram samples during preprocessing. (a) raw hologram, (b) enhanced hologram, (c) background subtracted hologram.
Figure 3.6. Sample images during segmentation. (a) original minimum intensity map, (b) high-pass filtered minimum intensity map, (c) segmentation results for larger than 3 pixels droplets, (d) segmentation results for smaller than or equal to 3 pixels droplets.
Figure 3.7. Classical CHT voting algorithm. (Mathworks)
Chapter 4. High-speed visualization and PIV
4.1 High-speed visualization

Wave repetition have been checked first using high-speed imaging results. As an example, temporal evolution of free surface for wave 2 at multiple locations over three independent runs is illustrated in Figure 4.1. The resultant non-breaking wave packet is very repeatable in time and space. The variation between runs is within 0.2 cm in space and 0.08s in time. However, it is important to note that the repeatability cannot be achieved at the breaking zone, presumably because of the highly chaotic wave breaking processes involved, as shown in Figure 4.2 for wave 2 without oil. The general feature of the wave 2 between different runs are similar, while the exact location, and concentration of the bubble plumes are not the same.

Figure 4.3 presents sample images of a plunging breaker hitting on an oil slick. The experiment is done by tuning the wave breaking exactly at the right location where we introduced a small patch of oil. A large roller is preceding to the left and finally impinges on the free surface. As the roller closes, part of the fluid parcel inside the roller will be entrained. Because of the rolling nature, a great amount of vorticity will be introduced into the otherwise irrational flow field. Another part of the fluid parcel will be peeled off upward and forward at higher speed than the wave.

Figure 4.4 compares sample images of the entrainment of 10 mL crude oil slick by the three waves listed in Table 1, with the specific locations and times indicated on each frame. Figure 4.4a, c, e illustrates the shape of the waves shortly after breakup. As is evident, waves 1 and 2 are plunging breakers, and the timings of the corresponding images are selected to show the overturning water jets impinging on the oil slicks. Wave
3 (Figure 4.4e) appears like a typical spilling breaker and image shows the wave front beginning to entrain the slick. Figures 4.4b, d, f are sample images of the bubble and oil droplet clouds shortly afterwards. For all cases, the turbulent wave fronts propagate at the speed of $c_t$, specified in Table 3.2, while plunging down and splashing up multiple times [Bonmarin, 1989], leaving a series of droplet and bubble clouds behind. After the initial plunging, most of the oil and air entrainment occur between the wave crest, which is indicated by an arrow, and the forward-most leading edge of the wave. The length of this region increases with wave height, reaching approximately 30, 20 and 10 cm for wave 1, 2 and 3, respectively.

The sample images in Figure 4.5 follow the time and streamwise evolution of the breaking and entrainment processes for wave 2 and DOR=1:25 oil. Each image shows the corresponding location and time after initiation of the wave plate motion, and all samples are intentionally presented at the same magnification. In Figure 4.5a, the plunging jet impinges on the oil slick while entraining oil and air. This process generates the first cloud of droplets and bubbles, which can be seen in the insert shortly afterwards. The initial impact is followed by multiple violent splash-ups and plunges observed before in several studies, e.g. [Bonmarin, 1989; Rapp and Melville, 1990; Perlin et al., 1996]. The first splash-up creates the largest, ~20 cm long, oil and bubble cloud, which is referred to in Figure 4.5a insert as the 2$^{nd}$ cloud shortly afterwards. Figure 4.5b shows the wave after the second splash-up and plunging, which generates the 3$^{rd}$ cloud, while the third splash-up is in progress. The fourth cloud resulting from the third splash-up and plunging, and the fourth splash-up are evident in Figure 4.5c. The process forms a series of spatially separated clouds with different sizes that migrate back and forth in the tank with the flow.
induced by the wave reflections. Consequently, the cloud pass across the holographic sample areas multiple times as they evolve under the influence of turbulence and gravity. Figure 4.5d presents the second and third clouds 0.54s after Figure 4.4c, showing that the clouds have been both displaced and spread vertically and horizontally. Figure 4.6 compares the appearance of clouds of crude and DOR 1:25 oil for the same wave and at the same time. In both cases, the wave structures and subsurface clouds have similar features, however, the droplet sizes are strikingly different. While individual crude oil droplets and bubbles are discernible in Figure 4.6a, the DOR 1:25 oil forms an opaque cloud with droplets sizes that fall well below the resolution range of the high-speed imaging system as shown in Figure 4.6b. Figure 4.7 shows a panorama ($\Delta x=440$ cm) of wave 2 entrains 50mL of crude oil from. It is generated by using one of the high-speed cameras and operating at seven consecutive $x$ locations along the tank. The magnification and vertical location of the camera remains the same. The pictures series for each column are from one run. The time difference between each row is 0.2s. As is obvious, the wave structures and the droplet cloud are consistent between columns. The splash-up and plunging becomes weaker and both wave height and the intrusion depth decreases as waves propagates downstream. After a single breaking wave, the oil slick from $x_o=150$cm has been distributed starting at around 1m from $x_o$ till the end of the tank. High speed visualizations for $h=20$cm waves are similar to what is discussed for $h=25$cm, and results are summarized in a video submitted to Gallery of Fluids Motion of 67th Annual Meeting of the American Physical Society Division of Fluid Dynamics [Li et al., 2014].
4.2 Characteristic droplet shapes

Adding dispersant drastically reduces the interfacial tension and droplets tend to deform even at quiescent rise flow conditions as illustrated in Figure 4.8. In this sample image, a syringe pump is used to inject DOR=1:25 oil slowly into the water tank forming droplets of various size. Consistent with [Gopalan and Katz, 2010], the cloud contains many instances of tip streaming and formation of long stable micro-threads presumably due to the ultralow interfacial tension. Sample microscopic views on the droplet shapes from wave experiments for several oils, as observed in the reconstructed holograms, are provided in Figure 4.9. A depth-compressed image with dense cloud of spherical crude oil droplets recorded shortly after breakup at $MA=1$ is presented in Figure 4.9a. The corresponding auto-detected droplets, ranging from 22µm to millimeter, are circled in white. As is evident, the data processing algorithms detect all the droplets, including those that are partially overlapped. In comparison, Figure 4.9b and 4.9c show droplet clouds for DOR 1:100 ($MA=1$) and 1:25 ($MA=10$) oils, respectively. Two trends resulting from adding dispersant are evident: First, for both concentrations, the fraction of large droplets (>100µm) decreases substantially. Second, consistent with the findings in [Gopalan and Katz, 2010], the breakup process of oil-dispersant mixtures involves formation of very long micro-threads, which subsequently break up into micron-scale droplets. Formation of these strings have been attributed to tip streaming, and a theoretical analysis performed by [Tseng and Prosperetti, 2015] shows that it is an inherent effect of interfacial instabilities in the neighborhood of a zero vorticity points in the lee side of the droplets.
Numerous strings with diameter smaller than 10 µm persist long after wave passage for the DOR 1:25 oil (e.g. Figure 4.9d at 100s). They eventually break up into micron-scale droplets, as demonstrated in Figure 4.9e, implying that for this case, the droplet size distributions continue to evolve well after wave passage. In [Gopalan and Katz, 2010], it is argued that the string breakup into droplets occurs when the dispersant diffuses out from the oil string, resulting in an increase in interfacial tension and development of interfacial capillary instabilities [Rayleigh, 1878, 1892]. Such capillary breakup occurs when the capillary number, $Ca=\mu_d U_d/\sigma$, where $U_d$ is a characteristic velocity scale, and subscript $d$ refers to the oil phase, falls below a critical value that depends, among other parameters, on the viscosity ratio and flow type [Stone, 1994a]. Hence, the breakup is likely to occur when the string diameter is of the same order or lower than the diffusion length scale, $(D_{ml}/U_d)^{0.5}$, where $D_m \sim 10^{-9}$ m$^2$/s is the molecular diffusion coefficient, and $l$ is a characteristic length scale. Typical values, involving e.g. the string length and turbulence intensity or droplet diameter and its quiescent rise rate would give scales smaller than 10µm. As shown in this paper, a large fraction of the droplets has substantially smaller diameter. Conversely, the order of magnitude higher interfacial tension for the DOR 1:100 oil inherently involves lower $Ca$ for the same flow conditions, and consequently, less stable and shorter-lived strings. Indeed, the DOR 1:100 oil strings break up into droplets within a few seconds ($t<9s$) after entrainment. In this case, the breakup of droplets under the influence of turbulence frequently involves the familiar filament stretching between a pair droplets (e.g. Figure 4.9f), and breakup of this filaments into small satellite droplets by capillary instability. The prevalence of this instability is evident from the wavy shape of the filament prior to fragmentation.
4.3. Evolution of turbulence after the initial wave breaking

Characterization of the turbulence intensity and energy spectra in the tank after wave breaking is needed for elucidating and modeling trends in the time evolution of droplets size distribution. The turbulence measurements are performed without oil, and the analysis avoids samples heavily populated by bubbles that obscure the field of view. Six runs are performed for each wave. During early time after breaking, the spatial distribution of turbulent regions is inherently patchy \cite{Rapp and Melville, 1990; Melville et al., 2002; Gemmrich and Farmer, 2004}. And turbulence zones are brought into and out of the \textit{FOV} of PIV measurements by the reflected waves. Figure 4.10-4.12 provides the spatially averaged horizontal velocity ($U$) for six independent runs with local positive peak and negative peaks marked with downward and upward triangle respectively. All three waves start with a positive $U$ peak as waves first pass the \textit{FOV}. The alternating negative peaks and positive peaks are presented as the waves are reflected back and forth. Because of turbulent dissipation, the magnitude of $U$ decreases to almost zero after around 70s. To identify the turbulent regions, we calculate the temporal evolution of the spatially averaged vorticity magnitude ($|\Omega|$), and choose the instantaneous maps with peak values as representatives of turbulent zones. Figure 4.13-4.15 shows $|\Omega|$ for wave 1-3, respectively. Vertical dash-dot line and dash line represent positive and negative $<U>$ peaks as shown in Figure 4.10-4.12. For the first 30s, the vorticity magnitude varies periodically, as turbulent patches are advected through the sample area by the (forward and reflected) wave-induced flow. The vorticity magnitude peaks nearly coincide in time with periods of high negative and positive $<U>$. For each run, we select the map with the highest vorticity magnitude that occurs in a time window of 1s (20\% of the sloshing
period) around the $<U>$ extrema as a representative of the turbulence generated by the 
breaking waves. These periodic variations diminish over time as the turbulence decays 
and diffuses. The chosen local peaks of $|Ω|$ are marked with downward triangle in Figure 
4.10-4.12. As an alternative to the present method for selecting the maps corresponding 
to wave turbulence, one could also use the data in several velocity maps recorded around 
the around the $<U>$ peaks. Sample comparisons between the two procedures are 
provided below.

The streamwise spatial energy spectra of the horizontal velocity component, $E_{11}(k_1)$, are 
calculated from the selected instantaneous velocity field along a series of horizontal lines 
of 168 vectors. Here, the subscripts 1 corresponds to the streamwise components and $k$ is 
the wavenumber. A discrete fast Fourier transform (fft function in Matlab) is used after 
subtracting the spatially averaged velocity from each line, without any other windowing 
functions. The instantaneous spectra obtained for the multiple lines and six runs are then 
ensemble averaged to obtain the final spectra for each wave. Only horizontal lines where 
the instantaneous spatially averaged vorticity magnitude exceeds 90% of the FOV 
spatially averaged levels are chosen for the spectrum averaging to ensure that the analysis 
does not involve laminar regions. To normalize the spectra, the dissipation rate is 
estimated by fitting the Kolmogorov -5/3 slope spectrum to wave number ranges that 
have a similar slope, and using $E_{11}(k_1)=18/55 \cdot 1.6 \cdot \varepsilon^{2/3} k_1^{-5/3}$ [Tennekes and Lumley, 1972], 
where $\varepsilon$ is the energy dissipation rate. The Kolmogorov length scale ($\eta$) is calculated 
from the estimated dissipation rate using $\eta=(\nu^3/\varepsilon)^{1/4}$. As expected, once $E_{11}(k_1)$ is 
normalized by $[\varepsilon(t)\nu^5]^{1/4}$ and the wavenumber is normalized by $\eta$, all these spectra 
collapse in the wavenumber range used for fitting the data. Results are provided for times
ranging from $t=2.3s$ to 100.1s after wave plate initiation with the corresponding $\eta$ specified for each time. Figure 4.16-4.18 provides temporal evolution of the non-dimensionalized 1D energy spectra of the streamwise velocity component, $E_{11}(k_1)$, for wave 1-3, respectively. As expected, $\eta$ increase by an order of magnitude over time, as the turbulence decays. At $t\sim100s$, $\eta\sim0.2-0.4$ mm is already consistent with typical scales found in the coastal turbulent boundary layers[\textit{Doron et al.}, 2001].

Figure 4.19 a and b present the temporal evolution of rms value of horizontal velocity fluctuations, $(u')$ and $\varepsilon$, respectively, for wave 1-3. Since one cannot readily separate between wave-induced unsteady motion (e.g. sloshing) and turbulence, one cannot simply ensemble average the velocity and subtract it from instantaneous value to obtain the velocity fluctuation. As an alternative, we calculate the streamwise component of the turbulent kinetic energy, $u'^2$, for the range of scales covered by the PIV measurements by integrating $E_{11}(k_1)$ over the available wave number range, namely,

$$u' \sim \sqrt{\int_{k_{\text{min}}}^{k_{\text{max}}} E_{11}(k_1) dk_1} \quad (4.1)$$

The magnitude of this estimate is lower than the total rms value since it does not account for scales that are larger than the field of view (4.6 cm). When the results in Figures 4.19a and b are least-square fitted to power relations, $\varepsilon \sim t^\alpha$ and $u' \sim t^\beta$, one obtains the exponents presented in the legend of each figure. Once the turbulent kinetic energy (TKE) production rate in the facility diminishes, one should expect that $\varepsilon \sim d(u'^2)/dt$, i.e. $\alpha \sim 2\beta - 1$, as discussed by [\textit{Rapp and Melville}, 1990]. Indeed, the presently measured values of $2\beta-1$ (Figure 4.19a) are within 6% of $\alpha$ for all cases. Figures 4.19a and b also provides sample data for $u'$ and $\varepsilon$ calculated using the alternative method (identified as alt.) for selecting
turbulent regions, namely by averaging results obtained from seven velocity distributions recorded during periods of \( <U> \) magnitude maxima. Although the results differ, especially at early times, the trends are quite similar, following the same trends and having the same order of magnitude.
Figure 4.1. Temporal evolution of free water surface before breaking for wave 2 at multiple locations.
Figure 4.2. Wave breaking zone between different runs for Wave 2 without oil.
Figure 4.3. Sample images of a plunging breaker hitting an oil slick.
Figure 4.4. Shapes of the three waves and droplets clouds generated after waves (a, b) 1, (c, d) 2, and (e, f) 3 entrain a 10mL crude oil slick. The timing and the location of measurement are specified on each plot. Dark arrows point to the location of the nearest wave crests to the leading edge of the breaker.
Figure 4.5. A series of sample images of wave 2 entraining oil-dispersant mixture at DOR 1:25 for the specified timing after wave initiation. (a) The initial impingement on the oil slick and formation of the first splash-up, with the insert showing the first and second oil clouds; (b) clouds appearing during the third splash-up; (c) clouds appearing during the fourth splash-up; and (d) the 2nd and 3rd clouds after wave passage.
Figure 4.6. Droplets and bubble clouds for wave 2 and slicks of: (a) crude oil; and (b) DOR 1:25 oil-dispersant mixture at $t=1.15$ s.
Figure 4.7. Panorama of wave 2 entraining crude oil using one camera.
Figure 4.8. Sample holograms showing DOR 1:25 oil forms micro-threads and “octopus” droplets
Figure 4.9. Sample reconstructed in-focus images of droplets compressed in the depth direction: (a) crude oil droplets (MA=1) with the automatically measured droplets circled; (b) Droplets and micro threads of crude oil-dispersant mixture at DOR=1:100 (MA=1); (c) superposition of 30 images of DOR=1:25 oil-dispersant mixture (MA=10); (d) three images showing capillary breakup of DOR=1:100 oil-dispersant mixture (MA=1); and long micro threads (e) before and (f) after breakup for DOR=1:25 oil-dispersant mixture (MA=10).
Figure 4.10. Temporal evolution of $U$ for wave 1 at location 4.

Figure 4.11. Temporal evolution of $U$ for wave 2 at location 4.
Figure 4.12. Temporal evolution of $U$ for wave 3 at location 4.
Figure 4.13. Temporal evolution of $|\Omega|$ for wave 1 at location 4.
Figure 4.14. Temporal evolution of $|\Omega|$ for wave 2 at location 4.
Figure 4.15. Temporal evolution of $|\Omega|$ for wave 3 at location 4.
Figure 4.16. non-dimensionalized 1D energy spectra of the streamwise velocity component, $E_{11}(k_1)$, for wave 1 at location 4.
Figure 4.17. Non-dimensionalized 1D energy spectra of the streamwise velocity component, $E_{11}(k_1)$, for wave 2 at location 4.
Figure 4.18. non-dimensionalized 1D energy spectra of the streamwise velocity component, $E_{11}(k_1)$, for wave 3 at location 4.
Figure 4.19. Temporal evolution of (a) horizontal rms velocity measured by integrating the energy spectra, (b) energy dissipation rate obtained by -5/3 slope fitting to the energy spectra for the three waves at location 4.
Chapter 5. Droplet size distribution
5.1 Subsurface droplet size distribution

5.1.1. Evaluation of temporal variation in droplet concentration

As discussed before, the droplet clouds migrate back and forth across the FOV because of wave reflections. Consequently, the droplet concentrations measured by the holography system oscillate in time, especially during the first 40-50s after wave initiation, and become homogenized at later periods. To examine the time evolution of concentrations and size distributions of droplets within the clouds, one must be careful in selecting times when these clouds are within the FOV. As an example, Figure 5.1 compares the evolution of spatially and ensemble averaged horizontal velocity over 6 runs, $<U>/V_{max}$, for wave 2 to the crude oil droplets volumetric concentration in parts per million (ppm) at location 4 ($MA=1$) for one of the runs. Here, the concentrations are evaluated by compressing the reconstructed 3D fields to 2D planes based on the minimum intensity for each pixel. Due to the location of the FOV, the data presented here corresponds to cloud No. 3 (Figure 4.4c, d). As is evident, both $<U>$ and the concentration vary quasi periodically, and the amplitude of oscillations decay in time. The decay in concentration is presumably caused by turbulent diffusion and buoyant rise of the droplets, as discussed and quantified later. The $<U>$ peak at $t=1.5s$ corresponds to the wave structure shown in Figure 4.4c passing across the FOV. The negative peak at 4.7s occurs when the reflected wave returns, followed by the next positive peaks associated with reflection from the other wall, and so on. The variations in concentration are initially complex and contain giant spikes, but become periodic at later times. For example, the small spike at $t=3s$ corresponds to the initial second splash up, which brings part (corner) of the cloud into the FOV. The spike at $4.2s<t<5.2s$ almost coincides with the $<U>$ peak associated with wave reflection,
which brings the center of the cloud into the $FOV$. Subsequently, elevated concentrations appear between the positive and negative $<U>$ peaks, implying that the forward wave and backward waves brings the cloud into and out of the $FOV$, respectively. Consequently, to characterize the droplet statistics within the clouds, we focus all the present analysis to periods elevated concentration, when the cloud passes across the field of view between the positive and negative $<U>$ peaks. For each 4.7s period and each run, out of the 250 holograms available, the statistics is based on data from 10 holograms with the highest concentrations. However, to avoid including the same droplets multiple times, the selected holograms have to be separated by at least 0.1s. The number of repeated runs is specified for each of the cases presented in this paper.

A comparison between sample temporal variation in concentration of bubbles and oil droplets for several types of oil are presented in Figure 5.2. Each line represents one run for data recorded using $MA=1$ at location 4, i.e. the data corresponds to cloud 3. Included are results obtained for bubbles (no oil), as well as crude, DOR=1:500, fish and motor oils. As is evident, the periodic variations in concentration that diminish in time persist in all the results, but the amplitudes and concentrations differ. The initial spikes in oil concentration ($t<5s$), which are associated with the plunging and splash up processes vary substantially, reflecting differences in the fraction of the clouds arriving initially to the sample area. The differences are much smaller in subsequent cycles. As expected, the bubble concentration decreases at the fastest rate, consistent with their higher buoyancy. It falls below 1 ppm after about 40 s. This trend justifies claiming that most of the particles in the clouds are oil droplets. The DOR 1:500 oil has the slowest rate of decreasing in concentration with time, followed by the fish oil, crude oil and motor oil.
As discussed later, the different rates involve effect of droplet sizes, which are substantially smaller for the DOR 1:500 oil and largest for the motor oil, as well as specific gravity, which is the highest for the fish oil. Finally, the amplitude of periodic variations and fluctuations in concentration for the DOR 1:500 oil appear to diminish at ~40 s, while the concentrations of the other oils continue to fluctuate. A possible explanation for this trend involves differences in the turbulent diffusion rates of the droplets. As shown by [Gopalan et al., 2008], the droplet turbulent diffusion rate increases with \( u'/U_q \), where \( U_q \) is the quiescent rise velocity of the droplets. Hence, small droplets are expected to dispersed by turbulence at a faster rate than large ones.

5.1.2 Effects of wave energy and location on the droplet statistics

Figure 5.3, 5.4 demonstrates the effect of wave energy on the oil droplet size distributions of crude oil at location 4 and \( MA=1 \) (Figure 5.3), and DOR=1:25 oil at location 3, and \( MA=10 \) (Figure 5.4). Each plot contains two distributions. The hollow symbols correspond to the earliest detected droplets at 0-5s, as the clouds reach the sample areas while the plunging and splash-ups occur above the sample volumes. The lines and solid symbols refer to droplets measured when the first reflected wave passes above the sample at 5-10 s, pushing clouds located slightly downstream back through the sample volumes. Each data point represents an average of multiple realizations obtained during three runs performed under the same conditions. The earliest distributions fluctuate in shape, in great part due to variations in the fraction of clouds passing through the sample volume. Conversely, for the reflected waves, while the concentrations vary with wave energy, the shapes of the size distributions appear to be quite similar with slopes averaged over all waves illustrated in each plot. In all cases, the droplet concentrations in the reflected
waves are lower than those observed initially, presumably due to the combined effects of buoyant rise and turbulent diffusion. For crude oil droplets (Figure 5.3), the concentration increases with wave energy, and in all cases, the slopes indicate that ~600 µm diameter droplets are the peak volumetric contributors to the subsurface oil concentration. For the DOR 1:25 oil (Figure 5.4), the slope changes slightly at around 10µm for all cases. The trends with wave energy are puzzling with wave 2 having the highest initial (0-5 s) concentration. The differences between waves 1 and 2 concentrations decrease for the reflected wave, but the wave 2 values are still higher. Several effects might be involved because of three possible reasons: (i) the cloud being sampled, (ii) the fraction of the cloud reaching the sample volume initially, and (iii) faster spreading of the oil in the tank because of the higher turbulent diffusion associated with wave 1. Examination of the high-speed movies indeed indicates that the clouds corresponding to wave 1 are larger, and hence more diluted than those of wave 2.

Figure 5.5 shows the effect of location on the early (2-10 s) size distribution for crude oil wave 2. Each plot is based on averaging 30 realizations obtained in three runs during periods of elevated concentration over the first 10s. As shown in Figure 3.2 and Table 3.5, FOVs 1 and 2 are located at x=270cm, but at different elevations. Figure 4.4 shows that these FOVs record the droplets located within the 2nd cloud, whereas location 4 samples the 3rd cloud. As is evident, while the concentrations vary, the shape of the size distributions remains quite similar at all locations and elevations. Within the same cloud (FOVs 1 and 2), the concentrations do not vary with depth. There is, however, a different in droplet concentration between cloud 2 and 3, with the latter being more dilute by about two times. Yet, the persistence of the size distribution slopes among locations justify
focusing our attention to effects of oil properties and time evolution in one of these clouds. Figure 5.5 also shows the bubble size distribution of wave 2 obtained during runs without oil. We have not tried to distinguish between oil droplets and bubbles during this study, but except for the largest droplets, the bubble concentration is almost an order of magnitude lower than that of the oils, even at early times. As Figure 5.2 shows, the discrepancy increases with time. Furthermore, since the slope of the bubble size distribution is very similar to that of the droplets, we treat the rest of the data presented in this paper as droplets.

5.1.3. Effects of dispersant concentration on the droplet statistics

Figure 5.6 demonstrates the effect of interfacial tension by comparing the early wave 2 results for crude oil to those of DOR 1:500, 1:100, and 1:25 oils. Here, data for $MA=1$ and 10 are combined to obtain an expanded view spanning three orders of magnitude of droplet sizes, over 9 orders of magnitude variations in number density and the corresponding to two orders of magnitude variations in interfacial tension. Results for both magnifications are presented for the DOR 1:500 and crude oil, but only $MA=10$ data are available for the DOR:100 and 1:25 oils. Each data point is based on averaging of 30 realizations recorded during three runs. The only exception is the DOR=1:100 and 1:25 oils and droplets larger than 44µm, where the number of realizations has been increased to 300 to obtain reliable statistics due to the small number of droplets in each realization. Consequently, the corresponding data points are not linked to the rest of the distributions at smaller diameters, and they are connected by dashed lines.

The number of DOR 1:25 and 1:100 oil droplets in the 2-10 µm diameter range is more than two orders of magnitude higher than those of the crude and DOR 1:500 oil, and the
difference remains substantial up to 30 µm. For the two low σi oils, the slope of size distributions appears to increase (become more negative) with increasing DOR. For the DOR 1:25 oil, nearly the same slope of about-2.9 persists up to ~10µm. For the DOR=1:100 case, the slope changes from -2.2 to -6.1, indicating that droplets in the 40-60 µm range are the peak volumetric contributors. For the crude oil, the MA=1 and 10 distributions appear to continuous with a slope of about -2.1 that persists up to 800 µm. The distributions for the DOR=1:500 oil have essentially the same slope and concentration in the 30 - 400 µm range, but there is a distinct transition to higher concentrations below about 10 µm. We do not have an explanation for this transition, but it is statistically robust. The slopes change drastically, at 500 µm for the DOR=1:500 oil, and at 800 µm for the crude oil, indicating the corresponding sizes of primary volumetric contributions. The presently observed slopes of -2.1 to -2.2, which are prevalent for a broad range of diameters and interfacial tensions (except for the DOR1:25 oil), is similar to value of -2.3 reported for crude oil by [Delvigne et al., 1987; Delvigne and Sweeney, 1988]. However, their size distributions do not show a change in slope.

The long-term evolution of micro-droplet distributions for DOR=1:25 and 1:100 oils is presented in Figure 5.7 and 5.8, respectively. These data are based on holograms acquired at 0.25-5 fps over period of up to 5hrs, and each plot is based on analysis of 30-90 holograms. For DOR=1:100 oil, the droplet concentration of all sizes decreases over time, especially over the first 10 min, and then continues at a slower pace. Considering that the early change is significantly larger with increasing droplet size, buoyant rise is a likely contributor. The rise velocity of 30 µm droplets is 70 µm /s [Clift, R., Grace, J.R., Weber, 1978], and should have a significant effect over 600s. Conversely, the rise
velocity of 3 µm droplets is less than 1 µm/s, i.e. their buoyant rise will be very limited. Hence turbulent dispersion of the droplets must also be a significant contributor, especially in the first few minutes, before the turbulence decays. The effects of buoyancy and time-dependent turbulence on the droplet concentrations are modeled and compared to data later in this paper.

The early time trends are quite different for the DOR=1:25 oil. Most notable are the changes occurring between 10s to 10min. While the concentration of droplets larger than 15µm decreases, the concentration of smaller droplets increases over time by 1.7 for to 4.5 times. Examination of the holograms involved indicates that the increase in concentration is caused by the previously mentioned micro-threading and subsequent breakup to micro-droplets (Figure 4.7), which persists for several minutes. Hence, during early times, the droplet production by micro-threading overcomes the effect of buoyant rise and turbulent diffusion for small droplets, but not for those larger than 15 µm. Between 10 min and two hours, the concentration decreases very slightly for $d<10\mu m$, and by about 2 times for larger diameters, suggesting that buoyant rise is the main contributor, and that the effect of turbulent dispersion is minimal. Between 2 and 5 hours, the concentrations decrease for by about 3 times without a clear trend with size. It should be noted that slow secondary flows with speeds of ~0.03mm/s persist in the tank for hours after each experiment.

**5.1.4. Effects of viscosity, buoyancy, and turbulence on the time evolution of droplet statistics**

Figure 5.9 demonstrates the effect of viscosity on the early 2-10s and 50-60s size distributions based on the $Ma=1$ data at location 4. During early 2-10s, an increase in
viscosity by 33 times doesn’t have a significant effect on both the slope and concentration of droplets smaller than 800μm. However, the spectral slopes change drastically for droplets larger than 800 μm, where they increase in magnitude (become more negative) with decreasing viscosity. It appears that the oil viscosity effect is restricted to mm scale droplets. Recalling that the crude and motor oils have almost the density and interfacial tension, the difference between their size distributions is most likely associated with their viscosity. Substantial literature exists on the effect of viscosity ratio on breakup of a dispersed phase in liquid-liquid two-phase flows on the droplet size distributions in a variety of settings[Calabrese et al., 1986; Stone, 1994a; Friedman et al., 2001]. A general conclusion is that the size range of droplets expands with the viscosity ratio, both in the small and large diameter ends of the distribution. This conclusion is consistent with the presently observed increase in the concentration of large droplets with oil viscosity. For the size distributions averaged over the 50s to 60s period, there is a very small change in the number of 30 - 40 μm droplets for all the oils, but the concentrations of larger droplets are significantly lower, and the those larger than ~700 μm essentially disappear. In contrast to the early samples, the concentration of fish oil droplets is now higher than that of the crude and motor oils for sizes exceeding ~100 μm, consistent with the lower buoyancy of the fish oil. The difference between the crude and motor oil concentrations remain low for this size range.

The time-dependent changes to the spectral slopes evident from Figures 5.9 motivate further analysis on the effects of buoyancy and turbulent dispersion on the transport of droplets. Figure 5.10-5.13 shows the time evolution of size distributions for the motor, fish, crude and DOR 1:500 oils, respectively. In all cases, the distributions steepen with
time, hardly changing for droplets smaller than 60 µm, and decreasing at a faster rate with increasing diameter. To explain these trends, we examine the time evolution of the size distributions predicted by a simple model involving turbulent diffusion and buoyancy, without any droplet breakup or coalescence. Assuming one dimensional diffusion in the z direction, i.e. horizontally uniform distributions, the droplets count concentration of a certain size, \( c(z, t, d) \) can be estimated from:

\[
\frac{\partial c}{\partial t} = -U_s(t) \frac{\partial c}{\partial z} + D(t) \frac{\partial^2 c}{\partial z^2} \tag{5.1}
\]

where \( U_s \) is the buoyancy-induced slip velocity, and \( D(t) \) is the turbulent diffusion coefficient. This equation does not account for many effects, such as horizontal diffusion, transport by wave, spatial inhomogeneity on the turbulence, etc. For simplicity, we assume that \( U_s \) is equal to \( U_q \). And \( U_q \) is calculated from the drag coefficients, \( C_D \) versus Reynolds number, \( Re \), curve for a solid sphere, provided in Table 5.2 of [Clift, R., Grace, J.R., Weber, 1978], and plotted in Figure 5.14, where \( C_D = 8F_D/(\pi d^2 \rho_f U_q^2) \), \( Re = U_q d/\nu_f \) and \( F_D \) is the drag force. The subscript \( f \) refers to the water. The results have been compared and validated based on data available in the same reference. The corresponding quiescent rise rates are provided in Figure 5.15. It should be noted that in reality, turbulence enhances the rise velocity of oil droplets with size falling in the inertial range of the turbulence [Friedman and Katz, 2002; Gopalan et al., 2008], and to a lesser extent also that of very small particles [Rosa et al., 2016]. The time varying turbulent diffusion coefficient can be estimated using \( D = Ku'L \), where \( L \) is the integral length scale, and \( K \) is a constant. We have selected \( K = 0.3 \), consistent with values obtained for droplets in isotropic turbulence by [Sato and Yamamoto, 1987], where \( K = 0.3-0.6 \), and by [Gopalan et al., 2008], where \( K = 0.23-0.33 \). The integral length scale is assumed to be of the same
order as the depth of the tank. The values of $u'$ correspond to periods of peak turbulence, using the power fit to its evolution for wave 2 shown in Figure 4.18. The diffusion equation is solved numerically for each droplet size using the partial differential equation solver in Matlab (function pdepe) using a spatial mesh of 0.01 cm and temporal mesh of 0.1 s. The initial condition for each diameter is the measured concentration at location 4 at 2-10s, and we assume that the oil droplet concentrations are homogeneous down to the initial intrusion depth of $z_i=-13$ cm (Table 3.2), and is equal to 0 for $z<-13$ cm. The boundary conditions are $cU_s-Dc/\partial z=0$ at $z=-25$ cm, i.e. there is no flux across the bottom boundary. The conditions at $z=0$ are questionable since rising droplets form new patches, but there are little spatial differences in concentration near the free surface (Figure 5.5). Hence, we opt to use $\partial c/\partial z=0$ at $z=0$, i.e. assume that the flux out of the domain at the free surface is only caused by buoyancy.

The computed time evolutions of droplet size distribution are presented as in Figure 5.10-5.13 as lines. In general, the simple model predicts the size- and time-dependent dilution of the droplet concentrations quite well. However, imposed by the assumed sharp transition in initial concentration at the intrusion depth, there is high downward turbulent diffusion at early period ($<15$ s), causing a decrease in concentration across all sizes. This trend diminishes at later times as the vertical concentration gradient become smooth but does not vanish because of the difference between the bottom and free surface boundary conditions. With a smooth transition, the predicted concentration of the smallest droplets detected at MA=1 hardly changes, consistent with the measured data and their low quiescent rise velocity (Figure 5.15), while the concentration of larger droplets decreases at increasing rate with size. The general agreement between the predicted and measured
size distributions suggests that the 1D model accounts for the primary contributors to the evolution of the size distribution. Namely, the changes with time are affected by upwards migration due to buoyancy and downward turbulent diffusion because of concentration gradients. Finally, Figure 5.16 compares the measured evolution total oil concentration in the sample volume over all sizes detected at MA=1, \( C_t \), to that predicted by the model. The data are normalized by the early (2-10s) experimental values, \( C_{t0} \), which is assigned to \( t=5s \). In general, trends of the simple model are similar to those of the measured values, with the initial rapid decrease occurring due to the above-mentioned imposed initial condition. Consistent with having the largest droplets, the motor oil concentration decays at the fastest rate. Conversely, the total concentration of DOR 1:500 oil, which has the lowest number of droplets larger than 600 µm, decays at the slowest rate.
5.2 Aerosol size distribution

The number size distributions of micron-scale droplets measured by APS and holography are compared in Fig. 5.17. For all cases, there is good agreement between the results of the two methods, especially for particle sizes larger than 5 µm, corresponding to droplets images larger than 4 pixels. The presence of an oil slick with or without dispersant has little effect on size distribution of the micron-sized droplets. For crude oil slicks, the number concentrations appear to be even slightly lower than those of the uncontaminated seawater (Fig. 5.17 a-b). When dispersant is added (Fig. 5.17 c-d), there is a small increase of up to 1.9 times for droplets smaller than about 5 µm. The same general trend is observed for the cases with dispersant only (Fig. 5.17 e-f).
Figure 5.1. Temporal evolution of non-dimensionalized horizontal velocity, and droplets concentration for wave 2 at location 4 ($MA=1$).
Figure 5.2. Samples showing oscillations of the total oil concentration for wave 2 at location 4 ($MA=1$) as oil clouds migrate across the field of view.
Figure 5.3. Effects of wave energy on the early (2-10s) droplet size distributions of crude oil ($MA=1$).
Figure 5.4. Effects of wave energy on the early (2-10s) droplet size distributions of DOR=1:25 oil (\(Ma=10\)).
Figure 5.5. Effects of location on the early (2-10s) droplet size distribution of crude oil for Wave 2.
Figure 5.6. Effects of oil-seawater interfacial tension on the early (2-10s) droplet size distributions for Wave 2, measured at location 3 and 4. Dashed lines are added to show approximate slopes.
Figure 5.7. Temporal variation over 2 hours of micro-droplet distributions ($MA=10$) for Wave 2 and oil-dispersant mixtures with DOR 1:100.
Figure 5.8. Temporal Variation over 5 hours of micro-droplet distributions ($MA=10$) for Wave 2 and oil-dispersant mixtures with DOR 1:25.
Figure 5.9. Effects of oil viscosity on the early 2 - 10s and 50 - 60s droplet size distributions for Wave 2, location 4, $MA=1$. 
Figure 5.10. Temporal variation (first 60s) of droplet size distributions for wave 2 (MA=1) and motor oil.
Figure 5.11. Temporal variation (first 60s) of droplet size distributions for wave 2 (MA=1) and fish oil.
Figure 5.12. Temporal variation (first 60s) of droplet size distributions for wave 2 (MA=1) and crude oil.
Figure 5.13. Temporal variation (first 60s) of droplet size distributions for wave 2 (MA=1) and DOR=1:500 oil.
Figure 5.14. Standard solid sphere Reynolds number and drag coefficient curve. Plotted diameter ranges from 2µm to 2475µm.
Figure 5.15. Quiescent rise velocity, $U_q$, calculated from drag coefficient curve for solid sphere particles with the same density as crude oil and fish oil.
Figure 5.16. Temporal evolution in total concentration for wave 2 (MA=1) and different oils.
Figure 5.17. Size distribution of airborne droplets in the micro-scale range averaged over 15 minutes. Left column shows results for continuously operated wave 3 and right column for continuously operated wave 2 for slicks containing: (a and b) crude oil (c and d) crude oil-dispersant mixture (DOR 1:25) and (e and f) dispersant only.
Chapter 6. Conclusion
Using digital holography at two magnifications, this dissertation provides detailed data on the effects of interfacial tension, viscosity, density, and wave energy on the time evolution of oil droplet size distributions generated by breaking waves. The initial entrainment generates several oils (and bubble) clouds associated with the multiple splash-ups and plunging. For oils with low (or no) dispersant concentration, the temporal evolution can be predicted by a simple model involving buoyant rise and turbulence dispersion. Trends are very different at high dispersant concentration, e.g. the DOR 1:25 oil, where the concentration of micro-scale droplets keeps increasing for the first ten minutes after entrainment due to continuing fragmentation of the micro-threads. Conversely, increasing the oil viscosity increases only the early concentration of large (mm scale) droplets. Finally, all the early size distributions have two size ranges with different slopes. Trends of the transitions between them are discussed below.

Experimentally, we have not seen even a single case of freestream droplet coalescence. Furthermore, except for the DOR 1:25 oil, where the concentration of micro-droplets increases in time as the threads break up, and to a much lesser extent for the DOR 1:100 oil, where the thread concentration is lower, there are few cases of droplet fragmentation after the initial breakup. To elucidate the dearth of turbulent fragmentation, Figure 6.1 present four characteristic droplet sizes for several cases. Here, the commonly used $d_{95}$ indicates that 95% of the total volume of oil is contained in droplets with $d \leq d_{95}$ [Clay, 1940; Hinze, 1955]. Next, $d_c$ corresponds to the point where the measured slope of the size distribution changes. It is estimated based on the intersection of two constant power lines fitted to the data (e.g. Figure 5.11). Fitted data of the slope, intercept, $d_c$ and droplet count concentration at $d = d_c$ are provided in Table 6.1. The third diameter, $d_w$, is
calculated from the critical Weber number for breakup of bubble and droplet in turbulent flows (\(We=\rho_f u'^2d/\sigma_i\)). Empirically obtained values for \(We\) vary from 1.2 to 4.7 [Hinze, 1955; Lewis and Davidson, 1982; Martínez-Bazán et al., 1999]. Selecting two characteristic values, 2 and 4, \(d_{we=4}=4\sigma_i/\rho_f u'^2\) is shown, and \(d_{we=2}\) is marked by a line on the same bar. The calculation is based on the initial values of \(u'\) (Figure 4.18a), immediately after wave breaking. The fourth size is the Hinze scale, \(d_H\), calculated from the dissipation rate, following [Deane and Stokes, 2002]. By assuming that only velocity fluctuations at scales smaller than the droplet diameter causes droplet breakup, \(u_c'^2=C\varepsilon^{2/3}d_H^{2/3}\), where \(C=2\) according to [Batchelor, 1951]. Substituting \(u_c'^2\) in the Weber number, \(d_H=2^{-3/5}(We\sigma_i/\rho_f)^{3/5}\varepsilon_c^{-2/5}\), where \(\varepsilon_c\) is the energy dissipation within the wave crest. Using empirical relation for dissipation per unit spanwise length (\(=0.009 c_w^5\rho_f/g\)) provided by [Loewen and Melville, 1991], and assuming that all the energy dissipation occurs in a cylinder with diameter equal to the initial wave roller diameter. In the present comparison, we use the initial intrusion depth provided in Table 3.2, and \(We=4\).

Several interesting features could be seen from Figure 6.1. First, except for the DOR 1:25 oil, the four characteristic droplet sizes are of the same order of magnitude. However, trends differ, e.g. the higher \(d_c\) and \(d_{95}\) for wave 1 compared to the other two waves evident also from Figure 5.3. While \(d_{we=4}\) increases with decreasing wave energy, consistent with its definition, \(d_H\) decreases slightly due to competing effects of decreasing turbulence and initial intrusion depth. As the turbulence decays over time, \(d_{we=4}\) increases, while \(d_{95}\) and \(d_c\) decrease due to the previously discussed dispersion and settling. Hence, the likelihood of breakup by turbulent shear diminishes. It should be noted that a different situation would most likely occur if the oil is continuously exposed to breaking waves.
and the turbulence does not decay, like in the Li et al. [2009] experiments. In their results, only the concentration of large droplets decreases with time, while that of small ones does not, possible, at least in part due to secondary breakup. Second, both $d_{H}$ and $d_{W_e=4}$ do not account for the effects of oil viscosity. Hence, they do not reproduce the increase in $d_{95}$ and $d_{c}$ with increasing oil viscosity (see also Figure 5.9). Third, all four characteristic droplet sizes decrease with increasing DOR, i.e. decreasing interfacial tension, but not at the same rate. The value of $d_{c}$ decreases at the fastest rate, $d_{95}$ and $d_{W_e=4}$ follow similar trends (but fluctuate), and $d_{H}$ decreases at a slowest rate due to the 3/5 power dependence on interfacial tension. The DOR 1:25 oil is the only case with significant difference between $d_{c}$ and $d_{95}$. This discrepancy is associated with the relatively mild slope change of the size distribution (from -2.9 to -5.3), implying that larger droplets are significant contributors to the total volume. Finally, the difference between $d_{c}$ and the values scaled based on turbulence parameters for the DOR 1:25 and 1:100 oils is a testimony that other processes, such as tip streaming, are involved in the breakup process.

Next, we compare the present size distributions to previously published results. For all the present cases, $N(d)\sim d^{-2.1}$ for $d<d_{c}$, with the exponent being around -2.1, except for the DOR 1:100 oil where it steepens to -2.2, and the DOR 1:25 oil, where it decreases substantially to -2.9. At larger diameters, the slopes have higher magnitudes than -3, indicating a volumetric peak at $d\sim d_{c}$. In [Delvigne et al., 1987; Delvigne and Sweeney, 1988], $N(d)\sim d^{-2.3}$ over the entire size range. A sample oil droplet size distribution is provided in Figure 6.2. Differences in sampling and data acquisition times, which in their case are longer, as well as methods used for fitting their wider-binned data might affect the small difference between trends. Their data also does not show the sharp change in
slope at $d-d_c$ observed in all the present initial distributions. Presumably, since their measurement procedures involve collection of water samples and observations at a later time under a microscope, their results might be affected by the same steepening, increase in slope magnitude, and smoothing of the transition over time evident from Figures 5.9-5.13. Figure 6.2 also provides distributions at various depths. Consistent with our results, the droplet size distributions are very similar within length scale comparable to the initial intrusion.

The data provided by [Li et al., 2009b] presented in number droplet size distribution for regular wave, spilling breaker and plunging breaker are provided in Figure 6.3-6.5. For crude oil and continuously generated waves show a slope of about -2 for spilling and plunging breakers, in agreement with the present results, and a steeper initial slope for a milder non-breaking wave, which transitions in time to -2. Steepening of the slope for all waves upon spraying the oil with dispersant is also observed from [Li et al., 2009b]. Moreover, their data show the slope change above certain diameters, which are generally smaller than the present values of $d_c$. It is not clear whether the continuous wave generation and resulting secondary droplet breakup play a role in this difference. PDPA-based field measurement by [Lunel, 1995] and LISST and photograph-based laboratory measurements by [Reed et al., 2009] show log normal distributions with peaks in the 10-500 µm range. The 10-500 µm peak is not consistent with the present observations, where the droplet number concentration increases monotonically with decreasing diameter once the magnification of our digital holographic microscopy system is increased to cover the micron scale range. This trend persists for all the present cases, irrespective of dispersant concentration. However, the present results agree with the
conclusions based on microscopic observations by [Delvigne et al., 1987; Delvigne and Sweeney, 1988], which also show a persistent increase in number with decreasing diameter for all the oils and over the entire 1-100 µm range.

One unresolved issue is the size distributions of oil-dispersant mixtures at scales falling below the present resolution range (~2 µm), possibly into the nanoscale. For example, Figure 6.6 shows microscopic image of the sample collected after the DOR 1:25 oil experiments. It clearly shows numerous ~1 µm and smaller droplets. These droplets have not been quantified in this study. In fact, a cloud of such droplets has remained suspended in the water indefinitely, consistent with phenomena observed in submerged plumes [Murphy et al., 2016]. Also, [Feng et al., 2014] has demonstrated bursting of bubbles at an oil/water/air interface with addition of surfactant results in formation of nanoemulsions. This phenomenon could also be found in wave breaking processes. In agreement, [Li et al., 2009b] show a bi-model distribution for oil-dispersant cases, with a sharp increase in the number of 1 µm droplets. The statistics and mechanisms involved in generation of submicron droplets, well below the present turbulence scales, is deferred to future studies.
Figure 6.1. Characteristic droplet size of $d_c$, $d_{95}$, $d_{We=4}$ and $d_H$. Dashed red lines indicates droplet diameter with $We=2$. 
Table 6.1. Slopes, intercept, $d_c$ and droplet count concentration at $d=d_c$.

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<th>Time</th>
<th>Slope 1 (d&lt; $d_c$)</th>
<th>Intercept 1 (d&lt; $d_c$)</th>
<th>Slope 2 (d&gt; $d_c$)</th>
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<th>$d_c$</th>
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Figure 6.2. Sample oil droplet size distribution from [Delvigne and Sweeney, 1988].
Figure 6.3. Temporal evolution of droplet size distribution under continuously generated plunging breakers with and without dispersant. (Data adapted from [Li et al., 2009b])
Figure 6.4. Temporal evolution of droplet size distribution under continuously generated spilling breakers with and without dispersant. (Data adapted from [Li et al., 2009b])
Figure 6.5. Temporal evolution of droplet size distribution under continuously generated regular waves with and without dispersant. (Data adapted from [Li et al., 2009b])
Figure 6.6. Microscopic image of the DOR 1:25 oil experiment water samples.
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Appendix A. Wave tank drawings

This appendix includes 3D assembly drawing (Figure A1) of the wave tank with parts listed in Table A1. Individual parts drawings are provided from Figure A2-A37. The tank has been designed to tilt ~3° by using four adjustable supporting jack (BJ-12 from Ellis Manufacturing Company INC, 19’’ - 31’’ in height, 80,000lb load capacity). This feature has not been used and the tank remains horizontal in all the experiments, confirmed by a level.
Figure A1. Wave tank general assembly drawing. Refer to Table A1 for individual parts.
<table>
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<th>Item #</th>
<th>Name</th>
<th>Material</th>
<th>Qty</th>
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<td>pivoting bracket</td>
<td>steel</td>
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<tr>
<td>01-00</td>
<td>movable corner</td>
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<td>02-00</td>
<td>jack support</td>
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<tr>
<td>02-01 (=03-01,04-01)</td>
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<td>steel</td>
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<td>02-02</td>
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<tr>
<td>02-03</td>
<td>C 9x15</td>
<td>steel</td>
<td>2</td>
</tr>
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<td>base 1</td>
<td>steel</td>
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</tr>
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<td>rec tube4x3 THK1/4</td>
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<td>C 9x15</td>
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<td>steel</td>
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<tr>
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<td>steel</td>
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<tr>
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<td>slotted plates</td>
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<td>C3x5 52in</td>
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</table>
Figure A3. Wave tank parts 01-00.
Figure A4. Jack support 02-00.
Figure A5. Base plate 02-01.
Figure A6. Rec tube 4”×3” THK ¼”, 02-02.
Figure A7. C9×15, 02-03.
Figure A8. Base #1, 03-00.
Figure A9. Rec tube 4”×3” Thk ¼”, 03-02.
Figure A11. Actuator support, 04-00.
Figure A12. Rec tube 4''×3'' Thk 1/4'', 04-02.
Figure A13. C 9” x 15”, 04-03.
Figure A14. Slotted plates, 04-04.
Figure A15. Vertical rod, 05-00.
Figure A16. Pivoting bracket modification, 05-01.
Figure A17. Slotted bar, 05-02.
Figure A19. Bearing Holder, 05-04.
Figure A20. Horizontal rod, 06-00.
Figure A21. Clevis bracket, 06-01.
Figure A22. Positioning block #1, 06-02.
Figure A23. I-beam for welding, 06-03.
Figure A24. Positioning block #2, 06-04.
Figure A25. APB-1HIHI(from Bimba), 06-05.
Figure A26. Wave plate, 07-00.
Figure A27. Carriage plate, 07-01.
Figure A28. Side L beam, 07-02.
Figure A29. Center L beam, 07-03.
Figure A30. Wave plate, 07-04.
Figure A31. Support, 07-05.
Figure A32. Rail support, 08-00.
Figure A33. Base plate, 08-01.
Figure A34. Support, 08-02.
Figure A35. Plate, 08-03.
Figure A36. Rail plate, 09-00.
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